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A Study of Optical Nonlinearities at the Single-Photon Level for Quantum Logic

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A Study of Optical Nonlinearities at the Single-Photon Level for Quantum Logic

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Abstract

In this dissertation, we shall focus on theoretically studying quantum nonlinear optical schemes to construct a conditional phase gate at the single-photon level. With an aim to develop analytical models, we shall carry out a rigorous quantized multimode field analysis of some of these schemes involving only the interacting field operators. More specifically, we shall first study the three-wave mixing process involving two single-photons in a second-order nonlinear medium ($\chi^{(2)}$) under two different cases viz. when the photons are traveling with equal velocities and when they are traveling with different velocities, and explore the possibility of using them for building a conditional phase phase gate.

Finally, we shall study the interaction of single-photon wavepackets with a realistic atomic system viz. an ensemble of five-level atoms, to construct a phase gate. We will particularly look at the "giant Kerr" effect in electromagnetically induced transparency to explore the possibility of using this scheme for achieving a conditional phase shift and to understand how the bandwidth gets restricted naturally in such an atomic system.

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This is the most important section of my dissertation. I could not have brought this work to fruition without the long and continuous support of several people. Ideally speaking, this should be a separate chapter in itself, in order to do complete justice to everyone's contribution to my work and life, as a graduate student. But the need to keep the length of my dissertation within a manageable limit has necessitated me to only briefly describe, in a few pages, the unstinting support that I received from all the significant people who have made this work possible.

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needed platform for me to learn the basics of this subject at one place. I am grateful to Dr. Gea-Banacloche more than words can express, for all his help and support.

Throughout this dissertation, I use first person plural, i.e. "we" instead of "I" for exposition. This is to remind the readers that this work is not my own. At every stage, Dr. Gea-Banacloche's invaluable assistance and insights have played a vital role in resolving the conundrums in this dissertation. But for his active participation and forethought, my labor would not have fructified.

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Finally, I wish to express my heartfelt thanks and indebtedness to my parents for their unconditional love and affection. It would not be possible for me to thank my mother in words, for all her support and help, without demanding anything from me. I am at loss of words to express my deepest sense of gratitude to her. She has given me absolute freedom to pursue what is close to my heart. She has always been the backbone of my strength and the primary source of my happiness. I dedicate my dissertation to her and to the memory of my father, with utmost love and affection.

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A synopsis on quantum logical operations

1.1 Introduction

Quantum information science is one of the most splendid mansions in the vast estate of modern physics. It is a magnificent edifice built upon the solid foundations of quantum mechanics. The field of quantum information science addresses a variety of problems, both of fundamental nature and of high practical importance. It encompasses a wide range of sub-fields such as quantum computing, quantum error correction, quantum cryptography, quantum entanglement, quantum communication etc. Each of these sub-fields is a challenging endeavor in its own right with several decades of intensive research having been put into it. Needless to say, no single research document can do adequate justice to all of these areas, in all its detail and rigor.

This dissertation focuses on a very specific problem in quantum computing, viz. exploring the theoretical aspects of constructing quantum logical gates with photons as qubits [1]. Qubits are the basic units for quantum computing and are analogous to the bits in a classical computer. It is important to note that there are many candidates for qubits in addition to photons such as trapped ions [2], electron spin, superconducting qubits [3] etc. However, photons are one of the most effective carriers of quantum information and they have certain inherent advantages over the other systems. One of them is that photons interact weakly with the environment. In addition to this, the information encoded in photons can be transmitted to a long distance with minimum loss. Furthermore, quantum logical operations involving photons can be performed at room temperature which may not

be possible in other systems.

1.2 Controlled NOT gate

Quantum logical gates are the fundamental building blocks of quantum circuits analogous to the digital gates in a classical computer. The fundamental quantum logical operation is the Controlled NOT (CNOT) gate which is given by the following transformation [4]:

$$|00\rangle \rightarrow |00\rangle,$$

$$|01\rangle \rightarrow |01\rangle,$$

$$|10\rangle \rightarrow |11\rangle,$$

$$|11\rangle \rightarrow |10\rangle,$$

$$(1.1)$$

where in the generic state $|AB\rangle$, $|A\rangle$ is the control qubit and $|B\rangle$ is the target qubit. In the gate operation, if the control qubit is $|1\rangle$, the target qubit gets flipped. We can see from Eq. (1.1) that both the input states $|10\rangle$ and $|11\rangle$ get transformed to $|11\rangle$ and $|10\rangle$, respectively. This is a universal quantum gate, i.e. all the other quantum logical gates can be constructed using the CNOT gate, which is the reason why most of the efforts in quantum computing are devoted to the construction of the CNOT gate. The single-qubit gates with photons are pretty much trivial and the challenge is in getting a universal two-qubit gate such as CNOT. The success of this endeavor will facilitate the operation of other quantum logical gates from this universal gate.

Using photons as qubits, the construction of a CNOT gate is a two-step process. It is a combination of a conditional phase (CPHASE) gate and two Hadamard gates. We shall denote the qubits $|0\rangle$ and $|1\rangle$ by the following column vectors:

$$|0\rangle \equiv \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

and

$$|1\rangle \equiv \begin{pmatrix} 0 \\ 1 \end{pmatrix}.$$

The Hadamard gate (U_H) is defined as

$$U_H|0\rangle \equiv \frac{1}{\sqrt{2}} (|0\rangle + |1\rangle),$$

$$U_H|1\rangle \equiv \frac{1}{\sqrt{2}} (|0\rangle - |1\rangle). \tag{1.2}$$

In the $(|0\rangle, |1\rangle)$ basis, the Hadamard gate can be explicitly represented by the following 2×2 matrix:

$$U_H = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}. \tag{1.3}$$

It can be easily seen that U_H is unitary.

The conditional phase (CPHASE) gate is described by the following transformation [4]:

$$|00\rangle \rightarrow |00\rangle,$$

$$|01\rangle \rightarrow |01\rangle,$$

$$|10\rangle \rightarrow |10\rangle,$$

$$|11\rangle \rightarrow -|11\rangle. \tag{1.4}$$

Since the CPHASE gate involves two qubits, the matrix corresponding to the transformation in Eq. (1.4) will be 4×4 which is represented as

$$U_{CPHASE} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}. \tag{1.5}$$

Next, we shall explicitly construct a matrix representation for the CNOT gate. Since we have two qubits, i.e. the control and the target, it is obvious that this gate has to be represented by a 4×4 matrix. We shall consider a general matrix of the form:

$$U_{CNOT} = \begin{pmatrix} c_{11} & c_{12} & c_{13} & c_{14} \\ c_{21} & c_{22} & c_{23} & c_{24} \\ c_{31} & c_{32} & c_{33} & c_{34} \\ c_{41} & c_{42} & c_{43} & c_{44} \end{pmatrix},$$

where we have to determine all the matrix elements. We shall use the transformation in Eq. (1.1) to compute the matrix elements and hence construct the matrix for the CNOT gate.

The column vector for the state $|00\rangle$ can be expressed as

$$|00\rangle = |0\rangle \otimes |0\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \otimes \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}.$$

Eq. (1.1) tells us that $U_{CNOT}|00\rangle = |00\rangle$. We can now express this transformation explicitly in terms of the matrices for U_{CNOT} and the state $|00\rangle$.

$$\begin{pmatrix} c_{11} & c_{12} & c_{13} & c_{14} \\ c_{21} & c_{22} & c_{23} & c_{24} \\ c_{31} & c_{32} & c_{33} & c_{34} \\ c_{41} & c_{42} & c_{43} & c_{44} \end{pmatrix} \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}, \tag{1.6}$$

which then yields

$$\begin{pmatrix} c_{11} \\ c_{21} \\ c_{31} \\ c_{41} \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}. \tag{1.7}$$

This implies that $c_{11} = 1$ and $c_{21} = c_{31} = c_{41} = 0$.

The second transformation in Eq. (1.1) can be expressed as $U_{CNOT}|01\rangle = |01\rangle$. The column vector for the state $|01\rangle$ can be written as

$$|00\rangle = |0\rangle \otimes |0\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \otimes \begin{pmatrix} 0 \\ 1 \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}.$$

The transformation for the state $|01\rangle$, in terms of the matrices, is given by

$$
\begin{pmatrix} 1 & c_{12} & c_{13} & c_{14} \\ 0 & c_{22} & c_{23} & c_{24} \\ 0 & c_{32} & c_{33} & c_{34} \\ 0 & c_{42} & c_{43} & c_{44} \end{pmatrix} \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}, \tag{1.8}
$$

which then gives us

$$
\begin{pmatrix} c_{12} \\ c_{22} \\ c_{32} \\ c_{42} \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}. \tag{1.9}
$$

This implies that $c_{22} = 1$ and $c_{12} = c_{32} = c_{42} = 0$.

The third transformation in Eq. (1.1) is given by $U_{CNOT}|10\rangle = |11\rangle$. The column vectors for the states $|10\rangle$ and $|11\rangle$ can be written as

$$
|10\rangle = |1\rangle \otimes |0\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \otimes \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix}
$$

and

$$
|11\rangle = |1\rangle \otimes |1\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \otimes \begin{pmatrix} 0 \\ 1 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}.
$$

The matrix representation of the transformation for the state $|10\rangle$ is given by

$$
\begin{pmatrix} 1 & 0 & c_{13} & c_{14} \\ 0 & 1 & c_{23} & c_{24} \\ 0 & 0 & c_{33} & c_{34} \\ 0 & 0 & c_{43} & c_{44} \end{pmatrix} \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}, \tag{1.10}
$$

which then yields

$$\begin{pmatrix} c_{13} \\ c_{23} \\ c_{33} \\ c_{43} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}. \tag{1.11}$$

This implies that $c_{43} = 1$ and $c_{13} = c_{23} = c_{33} = 0$.

The final transformation in Eq. (1.1) is $U_{CNOT}|11\rangle = |10\rangle$ which in the matrix

representation can be written as

$$\begin{pmatrix} 1 & 0 & 0 & c_{14} \\ 0 & 1 & 0 & c_{24} \\ 0 & 0 & 0 & c_{34} \\ 0 & 0 & 1 & c_{44} \end{pmatrix} \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix}, \tag{1.12}$$

from which we obtain

$$\begin{pmatrix} c_{14} \\ c_{24} \\ c_{34} \\ c_{44} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix}. \tag{1.13}$$

This implies that $c_{34} = 1$ and $c_{14} = c_{24} = c_{44} = 0$.

We have thus, computed all the matrix elements of U_{CNOT}. Now we have an explicit

matrix representation for the CNOT gate that is given by

$$U_{CNOT} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix}. \tag{1.14}$$

It can be easily seen that U_{CNOT} is unitary and furthermore, Eq. (1.14) can be

decomposed into the following form:

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$$\underbrace{\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix}}_{U_{CNOT}} = \underbrace{\frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & -1 & 0 & 0 \\ 0 & 0 & 1 & 1 \\ 0 & 0 & 1 & -1 \end{pmatrix}}_{I \otimes U_H} \underbrace{\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}}_{U_{CPHASE}} \underbrace{\frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & -1 & 0 & 0 \\ 0 & 0 & 1 & 1 \\ 0 & 0 & 1 & -1 \end{pmatrix}}_{I \otimes U_H}, \quad (1.15)$$

where

$$I \otimes U_H = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \otimes \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 & 0 & 0 \\ 1 & -1 & 0 & 0 \\ 0 & 0 & 1 & 1 \\ 0 & 0 & 1 & -1 \end{pmatrix}.$$

We mentioned earlier that a CNOT gate is a combination of a CPHASE gate and two Hadamard gates, and Eq. (1.15) shows this result explicitly. In order to appreciate this point better and see more clearly how these two transformations indeed act on the qubits themselves, we shall work out an example, i.e. we shall see how the CPHASE and the Hadamard gates act on the state $|10\rangle$ to perform the CNOT operation. From Eq. (1.1), we can see that $|10\rangle \xrightarrow{\text{CNOT}} |11\rangle$. In the state $|10\rangle$, $|1\rangle$ is the control qubit and $|0\rangle$ is the target qubit. In order to perform the CNOT operation on the state $|10\rangle$ using a CPHASE gate, we must first perform the Hadamard transform [see Eq. (1.2)] on the target qubit.

$$|10\rangle \equiv |1\rangle \otimes |0\rangle \xrightarrow{\text{Hadamard}} |1\rangle \otimes [U_H|0\rangle] = |1\rangle \otimes \frac{1}{\sqrt{2}} (|0\rangle + |1\rangle) = \underbrace{\frac{1}{\sqrt{2}} (|10\rangle + |11\rangle)}_{|\psi\rangle}. \quad (1.16)$$

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Next, we shall apply the CPHASE transformation [see Eq. (1.4)] on the state $|\psi\rangle$ which yields

$$|\psi\rangle \equiv \frac{1}{\sqrt{2}} \left(|10\rangle + |11\rangle\right) \xrightarrow{\text{CPHASE}} \frac{1}{\sqrt{2}}(|10\rangle - |11\rangle) = \underbrace{\frac{1}{\sqrt{2}} |1\rangle \otimes (|0\rangle - |1\rangle)}_{|\psi'\rangle}. \qquad (1.17)$$

Finally, we shall once again apply the Hadamard transform [see Eq. (1.2)] on the target qubit in the state $|\psi'\rangle$ which is $|0\rangle - |1\rangle$. Following this step, we obtain

$$|\psi'\rangle \equiv \frac{1}{\sqrt{2}} |1\rangle \otimes (|0\rangle - |1\rangle) \xrightarrow{\text{Hadamard}} \frac{1}{\sqrt{2}} |1\rangle \otimes \left([U_H|0\rangle] - [U_H|1\rangle]\right)$$

$$= \frac{1}{2} |1\rangle \otimes [|0\rangle + |1\rangle - |0\rangle + |1\rangle] = |1\rangle \otimes |1\rangle \equiv |11\rangle, \qquad (1.18)$$

which is the desired output, i.e. $|10\rangle \xrightarrow{\text{CNOT}} |11\rangle$. From Eqs. (1.16), (1.17) and (1.18), we can clearly see that a combination of a CPHASE transform and two Hadamard transforms indeed perform the CNOT gate operation for the state $|10\rangle$. We can follow the same procedure and perform the gate operation for the other states in Eq. (1.1).

1.3   Schemes to perform quantum logic with photons

Despite the many advantages that the photons have as qubits for quantum logical operations, there are some serious challenges to their physical realization. The major challenge that needs to be surmounted is the realization of a CPHASE gate at the single-photon level. The fourth transformation in Eq. (1.4), i.e. $|11\rangle \rightarrow -|11\rangle$, is extremely

hard to realize in a lab using photons. This transformation tells us that if we send two photons into the system, we want the two outgoing photons to acquire a phase π with respect to the incoming ones. It is important to note that the photons do not interact with each other directly. This means we need a suitable medium to mediate the interaction between the photons to achieve the desired phase shift without distorting the state. One such mechanism is to use a nonlinear optical medium toward this goal.

In the subsequent chapters, we shall study the interaction of single-photons with a nonlinear medium. To be specific, we will study the three wave mixing process involving two single-photons in a second-order nonlinear medium $(\chi^{(2)})$ and see under what conditions such schemes would work to construct a CPHASE gate. The method we adopt to solve this problem is to first characterize the medium by a pertinent Hamiltonian and then derive the relevant equations of motion for the system. Following this, we shall solve the dynamical equations analytically in an appropriate limit (with an aim to develop an analytical model) and analyze the solution to determine the possibility of using nonlinear optical schemes to construct a phase gate at the single-photon level. Many researchers have done extensive work in exploring the "Kerr" medium (i.e. third-order nonlinearity) for quantum logical operations with single-photons. We shall compare and contrast our results in the subsequent chapters with those for the third-order ones.

In chapters 2 and 3, we shall consider the "macroscopic" picture of the nonlinear medium. By "macroscopic", we mean that we won't be concerned with the internal constituents of the medium. Here, we shall use an appropriate Hamiltonian for the nonlinear medium involving only the interacting field operators and in our model, the only parameter associated with the medium that will figure in our analysis is the nonlinear

coupling strength. With this approach, we shall explore the theoretical possibilities of constructing a phase gate. We shall discuss both the positive and negative results and in addition, we shall explore ways to circumvent the objections to the possibility of conditional phase shifts on single-photons raised earlier.

We shall take the positive results from the macroscopic model, and search for a realistic atomic system that could help us physically realize a phase gate experimentally. This will be our endeavor in chapter 4. Here, we shall consider an ensemble of five-level atoms interacting with single-photon wavepackets to construct a conditional phase gate. We will be looking at the "giant Kerr" effect in electromagnetically induced transparency (EIT) which is equivalent to a conventional third-order nonlinear medium, in an atomic system. We shall develop a model based on adiabatic perturbation theory. We shall conclude our work with a discussion on the promises and challenges in this model toward the physical realization of a conditional phase gate with single-photons as qubits.

Chapter 2

Conditional phase gate based on second-order nonlinearity

2.1 Introduction

The exploitation of optical nonlinearities to implement quantum logical gates at the single-photon level has been an active problem of investigation over the past decade with considerable research efforts having been invested into it. The primary goal in this endeavor is to achieve a controlled phase shift, described by the transformation [4]:

$$|00\rangle \rightarrow |00\rangle,$$

$$|01\rangle \rightarrow e^{i\phi_1}|01\rangle,$$

$$|10\rangle \rightarrow e^{i\phi_1}|10\rangle,$$

$$|11\rangle \rightarrow e^{i\phi_2}|11\rangle,$$

$$(2.1)$$

where the useful phase for quantum logic is $\phi = \phi_2 - 2\phi_1$. In the ideal case, we want $\phi = \pi$. This operation is equivalent to a CNOT gate since we can indeed construct this gate with a controlled phase shift and a Hadamard transform. All the efforts to explore nonlinear optical schemes are to achieve this controlled phase shift by using the nonlinear medium to mediate photon-photon interaction.

Initially, the proposals for controlled phase shifts involved Kerr-type nonlinearities, which are conventionally classified as "third-order" because the susceptibility arises from terms that are cubic in the field, in the expansion of the polarization of the nonlinear

medium. However, Shapiro [5, 6, 7] in a seminal paper on conventional Kerr nonlinearity argued that when the multimode nature of a finite wavepacket interacting with a finite-bandwidth medium is considered, there is always an unavoidable trade-off between the desired phase shift and the achievable fidelity. By "conventional", it is meant that Shapiro's model for Kerr nonlinearity was a direct generalization of classical nonlinear optics to the quantized-field picture.

The primary cause for the degradation of fidelity in Shapiro's formalism is the phase noise arising from the Langevin operators introduced in the theory to preserve the commutation realtions of the field operators. Speaking in mathematical terms, the origin of this difficulty is the need to limit the medium's bandiwdth in order to avoid divergence in the theory. Physically, the finite bandwidth is connected to a finite response time for the medium, which is what we expect for a real optical system [8, 9, 10, 11]. The finite response time is incorporated in the theory by assigning a memory to the nonlinear index of refraction [5] and following this, the dynamical equations for the quantized field are written down. This, however, has a profound ramification. The free-field commutators obtained by solving these evolution equations no longer preserve the canonical commutation relation unless the "Langevin noise terms" are added.

The effect of the phase noise is negligible when the duration of the pulses are much larger than the response time of the medium. This is indeed the "large bandwidth limit". In this limit, Shapiro had found that the cross-Kerr phase shift goes to zero. We can visualize this in the following way: in a long pulse, the probability to find two photons within the same narrow time window corresponding to the response time of the medium is negligible which means that two photons will not even interact with the medium. This is

the reason why the optical nonlinearity vanishes in this limit.

These noise terms can also be neglected in the opposite limit (i.e. the short-pulse limit) because here, we send a broadband wavepacket through a narrowband optical medium and what happens is that the medium will either reject or absorb the incoming wavepackets. So once again, there is no phase shift.

Thus, the only relevant regime is when the bandwidth of the medium and the pulse are evenly matched in the frequency space. However, over here, the phase noise cannot be neglected and this indeed degrades the gate performance.

This view was further strengthened by Gea-Banacloche [12] in a Hamiltonian treatment of the "giant-Kerr effect". The main obstacle here to the high performance of the conditional phase gate is the spectral entanglement of the outgoing photons. The origin of this mechanism is the following: In order to get a large phase shift, the photons must interact very strongly with the nonlinear medium. This means that the incoming photons get destroyed and recreated inside the medium several times. However, when two photons co-propagate with equal velocities, the only constraint for the entire process is the conservation of momentum (or phase matching), which is the same as the conservation of energy, in this case. Thus, the only mathematical condition to be satisfied by the outgoing photons with frequencies ω_1 and ω_2 is $\omega_1 + \omega_2 = \omega_1' + \omega_2'$, where ω_1' and ω_2' are the frequencies of the incoming photons. This results in an entangled spectrum (for the final state) of the form

$$f(\omega_1, \omega_2) \sim \int d\omega' \; f_0(\omega') \; f_0(\omega', \omega_1 + \omega_2 - \omega')$$

in terms of the the incoming spectrum f_0 of the individual photon.

Thus, it has been concluded that the difficulties to the realization of such phase gates in Kerr media with high fidelity are due to time-nonlocality of the conventional nonlinear media and spectral entanglement of the final state.

A few years ago, Langford *et al.* [13] proposed a scheme for constructing a phase gate based on 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, the basic process would be

$$|011\rangle_{abc} \rightarrow -i|100\rangle_{abc} \rightarrow -|011\rangle_{abc}. \tag{2.2}$$

Here, we start with a two-photon state, i.e. a b, c pair, which annihilates inside the nonlinear medium to create an a photon. This is the parametric up-conversion process. Still later, the a photon annihilates to create a new b, c pair which is the down-conversion process. We want the final two-photon state to pick up a phase π with respect to the initial one, as shown in Eq. (2.2). This can be accomplished with the Hamiltonian

$$\hat{H} = \hbar\epsilon \ (\hat{a}^\dagger \ \hat{b} \ \hat{c} + \hat{a} \ \hat{b}^\dagger \ \hat{c}^\dagger), \tag{2.3}$$

where ϵ is the strength of the nonlinear coupling. The description provided by the Hamiltonian in Eq. (2.3) is a "single mode" picture because only one mode operator is assigned to each of the three photons involved. We can in fact understand the process described in Eq. (2.2) quantitatively by solving the equations of motion for this system. The state, in the single mode representation, is written as

$$|\psi(t)\rangle = \xi_a(t)\ \hat{a}^\dagger|0\rangle_a|0\rangle_b|0\rangle_c + \xi_{bc}(t)\ |0\rangle_a\ \hat{b}^\dagger|0\rangle_b\ \hat{c}^\dagger|0\rangle_c. \tag{2.4}$$

On inserting Eqs. (2.3) and (2.4) in the Schrödinger equation: $|\dot{\psi}\rangle = -(i/\hbar)\hat{H}|\psi\rangle$, we

get the following pair of differential equations for the a and the b, c photons:

$$\dot{\xi}_a = -i\epsilon\ \xi_{bc},$$

$$\dot{\xi}_{bc} = -i\epsilon\ \xi_a, \tag{2.5}$$

whose solutions are written as $\xi_a(t) = -i\ \sin(\epsilon t)$ and $\xi_{bc}(t) = \cos(\epsilon t)$ with the initial

condition that there is no a photon at $t = 0$.

Thus, we can formally write the state of the system as

$$|\psi(t)\rangle = -i\ \sin(\epsilon t)\ |100\rangle_{abc} + \cos(\epsilon t)\ |011\rangle_{abc}. \tag{2.6}$$

From Eq. (2.6), we can clearly see that beginning with the state $|011\rangle_{abc}$, evolution

under the Hamiltonian in Eq. (2.3) produces the middle state, $-i|100\rangle_{abc}$ at time $t = \pi/2\epsilon$,

and the final state $-|011\rangle_{abc}$ at $t = \pi/\epsilon$. It is evident that the states without an a photon

and with only one b or c photon are not affected by this Hamiltonian.

It should, however, be noted that a single-mode description is not adequate to describe

a traveling wavepacket. Our aim in this chapter is to find out what happens when we study

the scheme suggested by Langford *et al.* in a multimode framework which is an appropriate

description for single-photon pulses in a nonlinear medium.

One way to generalize the treatment based on Eq. (2.3) to a multimode framework is to simply replace the single-mode operators a, b and c by their corresponding multimode representations such as

$$\hat{a} \rightarrow \hat{A}(t) = \int d\omega \; \hat{a}_\omega \; e^{-i\omega t}, \tag{2.7}$$

where ω represents a deviation around the central frequency associated with the "a"-type modes. Similarly, we can define the multimode operators for the b and the c photons too. Nonetheless, a direct substitution of Eq. (2.7) into the Hamiltonian leads to diverging integrals in the model. In order to obtain finite results, it is imperative to account for the finite bandwidth that any real nonlinear medium must have. One way to accomplish this is to truncate the spectrum of the fields by hand. This is done by introducing upper and lower cutoffs in the integrals, in Eq. (2.7). This procedure can be justified by arguing that the medium has a finite transparency window, and absorbs all the spectral components outside its bandwidth. This is the approach that we shall pursue in the next section.

Another approach to study this problem is to place the nonlinear medium inside an optical cavity whose decay rate provides a natural bandwidth for the system. We shall work out this case in detail, in section 2.3.

2.2 Free space configuration

In this section, we shall study the evolution and propagation of a two-photon state through a second-order nonlinear $[\chi^{(2)}]$ medium. As mentioned in the previous section, we group the modes involved into three sets, denoted by the indices a, b and c. We shall use the

continuous mode formalism to solve this problem.

We assume that the pulse incident on the medium has one b and one c photon traveling in the same direction, and no a photon. As this pulse travels through the medium, the b and the c photons are annihilated to create an a photon. Later, the a photon is annihilated to create a new $b - c$ pair. We choose the length of the medium such that the interaction stops at this point, i.e. just when the new b, c pulse leaves the medium. Furthermore, we assume that the three photons viz. a, b and c have the same speed.

Under these assumptions, the Hamiltonian of the system, in the Schrödinger picture, is written as

$$\hat{H} = \hat{H}_0 + \hat{H}_{int},$$

$$\hat{H}_0 = \hbar v \int dk \ k \ \hat{a}_k^\dagger \ \hat{a}_k + \hbar v \int dk \ k \ \hat{b}_k^\dagger \ \hat{b}_k + \hbar v \int dk \ k \ \hat{c}_k^\dagger \ \hat{c}_k,$$

$$\hat{H}_{int} = \hbar \epsilon \int_{z_0}^{z_0+l} dz \int_{-k_{max}}^{k_{max}} dk_a \int_{-k_{max}}^{k_{max}} dk_b \int_{-k_{max}}^{k_{max}} dk_c \ e^{i(k_a - k_b - k_c)z}$$

$$\times \ \hat{a}(k_a) \ \hat{b}^\dagger(k_b) \ \hat{c}^\dagger(k_c) + H.c., \tag{2.8}$$

where $\hat{H}_0$ is the Hamiltonian of the free field, $\hat{H}_{int}$ is the Hamiltonian corresponding to the second-order optical nonlinearity. Here we have labeled the modes by wavevectors instead of frequency for notational convenience. We will solve this problem in the interaction picture. The next step, obviously, is to transform $\hat{H}_{int}$ from the Schrödinger picture to the interaction picture and the unitary transformation that does this is

$$\hat{H}_{int}^{I} = e^{i\hat{H}_0 t/\hbar} \; \hat{H}_{int} \; e^{-i\hat{H}_0 t/\hbar},$$

where the superscript I symbolically denotes the interaction picture. This

transformation yields us the following expression for $\hat{H}_{int}$ in the interaction picture:

$$
\begin{aligned}
\hat{H}_{int}^{I} = \hbar\epsilon & \int_{-k_{max}}^{k_{max}} dk_a \int_{-k_{max}}^{k_{max}} dk_b \int_{-k_{max}}^{k_{max}} dk_c \int_{z_0}^{z_0+l} dz \; e^{i(k_a - k_b - k_c)z} \\
& \times e^{ivt \int dk' \; k' \; \hat{a}^\dagger(k')\hat{a}(k')} \; \hat{a}(k_a) \; e^{-ivt \int dk' \; k' \; \hat{a}^\dagger(k')\hat{a}(k')} \\
& \times e^{ivt \int dk' \; k' \; \hat{b}^\dagger(k')\hat{b}(k')} \; \hat{b}^\dagger(k_b) \; e^{-ivt \int dk' \; k' \; \hat{b}^\dagger(k')\hat{b}(k')} \\
& \times e^{ivt \int dk' \; k' \; \hat{c}^\dagger(k')\hat{c}(k')} \; \hat{c}^\dagger(k_c) \; e^{-ivt \int dk' \; k' \; \hat{c}^\dagger(k')\hat{c}(k')} + H.c., \\
\hat{H}_{int}^{I} = \hbar\epsilon & \int_{z_0}^{z_0+l} dz \int_{-k_{max}}^{k_{max}} dk_a \int_{-k_{max}}^{k_{max}} dk_b \int_{-k_{max}}^{k_{max}} dk_c \; e^{-i(vt-z)(k_a - k_b - k_c)} \\
& \times \hat{a}(k_a) \; \hat{b}^\dagger(k_b) \; \hat{c}^\dagger(k_c) + H.c.,
\end{aligned}
\tag{2.9}
$$

where

$$e^{ivt \int dk' \; k' \; \hat{a}^\dagger(k')\hat{a}(k')} \; \hat{a}(k_a) \; e^{-ivt \int dk' \; k' \; \hat{a}^\dagger(k')\hat{a}(k')} = \hat{a}(k_a) \; e^{-ivk_a t},$$

$$e^{ivt \int dk' \; k' \; \hat{b}^\dagger(k')\hat{b}(k')} \; \hat{b}^\dagger(k_b) \; e^{-ivt \int dk' \; k' \; \hat{b}^\dagger(k')\hat{b}(k')} = \hat{b}(k_b) \; e^{-ivk_b t},$$

and

$$e^{ivt \int dk' \; k' \; \hat{c}^\dagger(k')\hat{c}(k')} \; \hat{c}^\dagger(k_c) \; e^{-ivt \int dk' \; k' \; \hat{c}^\dagger(k')\hat{c}(k')} = \hat{c}(k_c) \; e^{-ivk_c t}.$$

This is the natural multimode generalization of the Hamiltonian in Eq.(2.3), under the assumptions that the interaction takes place between two co-propagating wavepackets traveling at the same velocity v, in a medium of length l. The bandwidth cutoff actually makes the interaction nonlocal in space. The field operator (say for the a photon)

$$\int_{-k_{max}}^{k_{max}} \hat{a}(k_a) \ e^{-i(vt-z)k_a} \ dk_a$$

does not act at single space-time point, but over a range of values of $vt - z$, of the order of $1/2k_{max}$. In other words, the truncated field operators in Eq. (2.9) annihilate and create a photon, not necessarily precisely at z, but rather in a region of space of width $1/2k_{max}$.

The most general field state under our assumptions (only one b and one c photons or only one a photon) is written as

$$|\psi(t)\rangle = \int dk_1 \ \xi_a(k_1,t) \ \hat{a}^\dagger(k_1)|0\rangle_a|0\rangle_b|0\rangle_c + \int dk_2 \int dk_3 \ \xi_{bc}(k_2,k_3,t) \ |0\rangle_a$$
$$\times \ \hat{b}^\dagger(k_2)|0\rangle_b \ \hat{c}^\dagger(k_3)|0\rangle_c. \tag{2.10}$$

We shall now use Eqs. (2.9) and (2.10) to write down the Schrödinger equation: $|\dot{\psi}\rangle = -(i/\hbar)\hat{H}^I_{int}|\psi\rangle$, to get the equations of motion for the a and the b, c pulses. We have assumed that the medium has a finite bandwidth $2k_{max}$, so that photons with frequencies which lie outside this window do not contribute to the time evolution.

This procedure yields the following pair of differential equations:

$$\frac{\partial}{\partial t}\xi_a(k_a, t) = -i\epsilon \int_{z_0}^{z_0+l} dz \int_{-k_{max}}^{k_{max}} dk_b \int_{-k_{max}}^{k_{max}} dk_c \ e^{i(vt-z)(k_a-k_b-k_c)} \ \xi_{bc}(k_b, k_c, t),$$

$$\frac{\partial}{\partial t}\xi_{bc}(k_b, k_c, t) = -i\epsilon \int_{z_0}^{z_0+l} dz \int_{-k_{max}}^{k_{max}} dk_a \ e^{-i(vt-z)(k_a-k_b-k_c)} \ \xi_a(k_a, t). \tag{2.11}$$

In order to solve this system of equations, we will introduce "envelope functions" $f(t, z)$

and $g(t, z)$ for the a and the b, c pulses, respectively, which are defined as

$$f(t, z) \equiv \int_{-k_{max}}^{k_{max}} dk \ \xi_a(k, t) \ e^{-i(vt-z)k},$$

$$g(t, z) \equiv \int_{-k_{max}}^{k_{max}} dk' \int_{-k_{max}}^{k_{max}} dk'' \ \xi_{bc}(k', k'', t) \ e^{-i(vt-z)(k'+k'')}. \tag{2.12}$$

In terms of these two envelope functions, Eq. (2.11) can be rewritten as

$$\frac{\partial}{\partial t}\xi_a(k_a, t) = -i\epsilon \int_{z_0}^{z_0+l} dz \ e^{i(vt-z)k_a} \ g(t, z),$$

$$\frac{\partial}{\partial t}\xi_{bc}(k_b, k_c, t) = -i\epsilon \int_{z_0}^{z_0+l} dz \ e^{i(vt-z)(k_b+k_c)} \ f(t, z). \tag{2.13}$$

Clearly, the functions $f(t, z)$ and $g(t, z)$ satisfy the following propagation equation:

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z}\right) f(t, z) = \int_{-k_{max}}^{k_{max}} dk \; \dot{\xi}_a(k, t) \; e^{-i(vt-z)k},$$

$$= i\epsilon \int_{z_0}^{z_0+l} dz' \; g(t, z') \int_{-k_{max}}^{k_{max}} dk \; e^{ik(z-z')}, \qquad (2.14)$$

where we have substituted for $\dot{\xi}_a$ from the first of Eqs. (2.13). If we assume that the "acceptance bandwidth" of the medium, $2k_{max}$, is very large for the a photon, then we could approximate the integral over k on the right hand side of Eq. (2.14) by a δ function, $2\pi\delta(z - z')$. However, to justify this approximation, $1/(2k_{max})$ should be much smaller than both l and the spatial width of the b, c pulse. This simplifies Eq. (2.14) to

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z}\right) f(t, z) \simeq -2\pi i\epsilon \; g(t, z) \; \text{rect}(z, z_0, z_0 + l), \qquad (2.15)$$

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.

Next, we shall get the propagation equation for $g(t, z)$. Clearly, we have

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z}\right) g(t, z) = \int_{-k_{max}}^{k_{max}} dk' \int_{-k_{max}}^{k_{max}} dk'' \; \dot{\xi}_{bc}(k', k'', t) \; e^{-i(vt-z)(k'+k'')},$$

$$= -i\epsilon \int_{z_0}^{z_0+l} dz' \; f(t, z') \int_{-k_{max}}^{k_{max}} dk' \; e^{ik'(z-z')} \int_{-k_{max}}^{k_{max}} dk'' \; e^{ik''(z-z')}, \qquad (2.16)$$

where we have substituted for $\dot{\xi}_{bc}$ in the previous equation from the second of Eqs. (2.13). Here we cannot simply let k_{max} go to infinity, since at least one of the integrals in Eq. (2.16) will diverge. We can, nonetheless, under the same assumptions as before, replace one of the integrals over k on the right hand side of the previous equation by $2\pi\delta(z - z')$, following which the other integral will just have the value $2k_{max}$, resulting in a more compact equation for $g(t, z)$:

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z}\right) g(t, z) = -4\pi i\epsilon \; k_{max} \; f(t, z) \; \text{rect}(z, z_0, z_0 + l). \tag{2.17}$$

We now have to solve the system in Eqs. (2.15) and (2.17). To start with, we shall make the following coordinate transformations: $t' = t - (z - z_0)/v$ and $z' = z$. In terms of these new coordinates,

$$\frac{\partial}{\partial t} + v\frac{\partial}{\partial z} = v\frac{\partial}{\partial z'}.$$

Thus, Eqs. (2.15) and (2.17) can be rewritten as

$$\frac{\partial}{\partial z'}f(t', z') = -\frac{2\pi i\epsilon}{v}g(t', z'), \tag{2.18}$$

and

$$\frac{\partial}{\partial z'}g(t', z') = -\frac{4\pi i\epsilon k_{max}}{v}f(t', z'). \tag{2.19}$$

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It is evident that Eqs. (2.18) and (2.19) are coupled. The standard trick is to decouple them and then solve for the envelope functions. We shall differentiate both sides of Eq. (2.18) with respect to z' and substitute for $\partial g/\partial z'$ from Eq. (2.19), which would yield

$$\frac{\partial^2}{\partial z'^2} f(t', z') = -\frac{8\pi^2 \epsilon^2 k_{max}}{v^2} f(t', z') = -\frac{\Omega^2}{v^2} f(t', z'), \tag{2.20}$$

where $\Omega^2 \equiv 8\pi^2 \epsilon^2 k_{max}$. The formal solution to this differential equation is

$$f(t', z') = A \cos\left[\frac{\Omega}{v}(z' - z_0)\right] + B \sin\left[\frac{\Omega}{v}(z' - z_0)\right], \tag{2.21}$$

where A and B are quantities which are independent of z' but may well depend on t'. At $z' = z_0$, $A = f(t', z_0)$.

Next,

$$\frac{\partial}{\partial z'} f(t', z') = -\frac{\Omega}{v} A \sin\left[\frac{\Omega}{v}(z' - z_0)\right] + \frac{\Omega}{v} B \cos\left[\frac{\Omega}{v}(z' - z_0)\right]. \tag{2.22}$$

From the previous equation, we see that at $z' = z_0$, $B = (v/\Omega)(\partial f/\partial z')_{z'=z_0}$, and from Eq. (2.18), we get $B = -(2\pi i\epsilon/\Omega)g(t', z_0)$. Note that $f(t', z_0) = 0$, since there is no a photon until the interaction begins inside the medium [see Eq.(2.12)], which makes A vanish, i.e. $A = 0$. This simplifies the solution in Eq.(2.21) to

$$f(t', z') = -\frac{2\pi i\epsilon}{\Omega} g(t', z' = z_0) \sin\left[\frac{\Omega}{v}(z' - z_0)\right]. \tag{2.23}$$

Our next task is to express the envelope function f in the previous equation, in terms of the original coordinates (t, z). Noting that $t' = t - (z - z_0)/v$ and $z' = z$, we can see that at t=0 and some constant t', $z_{initial} = -vt' + z_0$. On substituting for t' in terms of the original coordinates, we get $z_{initial} = z - vt$, at $t = 0$. Thus, in terms of the original coordinates, Eq. (2.23) gives us following solution, in the region $z_0 \leq z \leq z_0 + l$,

$$
\begin{aligned}
f(t, z) &= -\frac{2\pi i \epsilon}{\Omega} \, g(0, z - vt) \, \sin\left[\frac{\Omega}{v}(z - z_0)\right], \\
&= -\frac{i\Omega}{4\pi \epsilon k_{max}} \, g(0, z - vt) \, \sin\left[\frac{\Omega}{v}(z - z_0)\right],
\end{aligned}
\tag{2.24}
$$

where we have substituted explicitly for Ω in the previous equation. On substituting the formal solution for $f(t, z)$ from Eq. (2.24) in Eq. (2.15), we get

$$
g(t, z) = g(0, z - vt) \, \cos\left[\frac{\Omega}{v}(z - z_0)\right].
\tag{2.25}
$$

Our goal here is to obtain an explicit expression for $\xi_{bc}(k_b, k_c, t)$. So as a first step toward this goal, we shall substitute for $f(t, z)$ from Eq. (2.24) in the second of Eqs. (2.13) to get

$$
\frac{\partial}{\partial t}\xi_{bc}(k_b, k_c, t) = -\frac{\omega}{4\pi k_{max}} \int_{z_0}^{z_0+l} dz \, e^{i(vt-z)(k_b+k_c)} \, g(0, z - vt)
$$
$$
\times \sin\left[\frac{\Omega}{v}(z - z_0)\right],
\tag{2.26}
$$

and then formally integrate both sides of Eq. (2.26) with respect to time.

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This yields,

$$
\xi_{bc}(k_b, k_c, t) = \xi_{bc}(k_b, k_c, 0) - \frac{\Omega}{4\pi k_{max}} \int_{z_0}^{z_0+l} dz \ e^{-iz(k_b+k_c)} \ \sin\left[\frac{\Omega}{v}(z - z_0)\right]
$$

$$
\times \int_0^t d\tau \ e^{i(k_b+k_c)v\tau} \ g(0, z - v\tau). \tag{2.27}
$$

Now the integral with respect to τ in Eq. (2.27) involves the spatial profile of the b, c pulse. From the second of Eqs. (2.12), we get

$$
g(0, z - v\tau) = \int_{-k_{max}}^{k_{max}} dk' \int_{-k_{max}}^{k_{max}} dk'' \ \xi_{bc}(k', k'', 0) \ e^{-i(v\tau - z)(k'+k'')}. \tag{2.28}
$$

The b and the c photons enter the medium (i.e. $z = z_0$) at time $t = 0$. Note that the variable z in Eq. (2.28) is confined to the range: $z_0 \leq z \leq z_0 + l$. This means that for $t \leq 0$, $g(0, z - vt)$ should be negligible, in which case we can harmlessly extend the lower limit of integration over τ to $-\infty$. Furthermore, since we are ultimately interested in the state long after the interaction with the medium is over, we can also formally extend the upper limit of integration to ∞. So, now on inserting Eq. (2.28) in Eq. (2.27), we find that

$$
\int_0^t d\tau \ e^{i(k_b+k_c)v\tau} \ g(0, z - v\tau) = \int_{-k_{max}}^{k_{max}} dk' \int_{-k_{max}}^{k_{max}} dk'' \ \xi_{bc}(k', k'', 0) \ e^{iz(k'+k'')}
$$

$$
\times \underbrace{\int_{-\infty}^{\infty} d\tau \ e^{i(k_b+k_c-k'-k'')v\tau}}_{(2\pi/v) \ \delta(k_b+k_c-k'-k'')}. \tag{2.29}
$$

The δ function allows us to replace $\exp[iz(k' + k'')]$ in Eq. (2.29) by $\exp[iz(k_b + k_c)]$ which will then cancel out its complex conjugate in Eq. (2.27).

Thus, on substituting Eq. (2.29) in Eq. (2.27), we obtain, in the long-time limit $(t \to \infty)$

$$\xi_{bc}(k_b, k_c, t \to \infty) = \xi_{bc}(k_b, k_c, 0) - \frac{\Omega}{2vk_{max}} \int_{-k_{max}}^{k_{max}} dk' \int_{-k_{max}}^{k_{max}} dk'' \, \xi_{bc}(k', k'', 0)$$
$$\times \, \delta(k_b + k_c - k' - k'') \int_{z_0}^{z_0+l} dz \, \sin\left[\frac{\Omega}{v}(z - z_0)\right]. \qquad (2.30)$$

Our next task is to evaluate the integral over z which gives us

$$\int_{z_0}^{z_0+l} dz \, \sin\left[\frac{\Omega}{v}(z - z_0)\right] = \frac{v}{\Omega}\left[1 - \cos\left(\frac{\Omega l}{v}\right)\right]. \qquad (2.31)$$

We want to ensure that there is no a photon after the interaction with the medium is over, i.e. for $z > z_0 + l$, for all t. This means that we specifically want $f(t, z_0 + l) = 0$, in Eq. (2.24). Consequently, this implies that $\sin(\Omega l/v) = 0$ which means the integral in Eq. (2.30) is either zero or $2v/\Omega$ and when we incorporate this result in Eq. (2.30), we end up with

$$\xi_{bc}(k_b, k_c, t \to \infty) = \xi_{bc}(k_b, k_c, 0) - \frac{1}{k_{max}} \int_{-k_{max}}^{k_{max}} dk' \int_{-k_{max}}^{k_{max}} dk'' \, \xi_{bc}(k', k'', 0)$$
$$\times \, \delta(k_b + k_c - k' - k''). \qquad (2.32)$$

From this result, the major obstacle to obtain a high fidelity, i.e. a large overlap with the initial state, is immediately apparent. The two outgoing b and c photons with momenta

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k_b and k_c may be created from an initial pair having any momenta, k' and k'', subject to the constraint $k' + k'' = k_b + k_c$. This is indeed the condition for momentum conservation. This means that the final state may not spectrally resemble the initial state, very much at all. Even if we assume that the initial state is factorizable, the final state given by Eq. (2.32) is entangled in momentum. This is exactly the same problem one confronts in schemes involving Kerr nonlinearities.

The expression for the final state in Eq. (2.32) may be simplified a little by introducing two new variables η and Λ such that $k_b = (\eta + \Lambda)/2$ and $k_c = (\eta - \Lambda)/2$. This makes $dk' \, dk'' = (1/2) \, d\Lambda' \, d\eta'$. In terms of these new variables, Eq. (2.32) can be rewritten as

$$\xi_{bc}(\eta, \Lambda, t \to \infty) = \xi_{bc}(\eta, \Lambda, 0) - \frac{1}{2k_{max}} \int_{-2k_{max}+|\eta'|}^{2k_{max}+|\eta'|} d\Lambda' \int_{-2k_{max}}^{2k_{max}} d\eta' \; \xi_{bc}(\eta', \Lambda', 0)$$

$$\times \, \delta(\eta - \eta'). \qquad (2.33)$$

On enforcing the δ function, we get a slightly more compact expression for the final state given by

$$\xi_{bc}(\eta, \Lambda, t \to \infty) = \xi_{bc}(\eta, \Lambda, 0) - \frac{1}{2k_{max}} \int_{-2k_{max}+|\eta|}^{2k_{max}+|\eta|} d\Lambda' \; \xi_{bc}(\eta, \Lambda', 0). \qquad (2.34)$$

We can define a combined phase and fidelity, by the following quantity:

$$\sqrt{F}e^{i\phi} = \langle \psi(0)|\psi(t)\rangle$$

$$= \int_{-k_{max}}^{k_{max}} dk_b \int_{-k_{max}}^{k_{max}} dk_c \; \xi_{bc}^*(k_b, k_c, 0) \; \xi_{bc}(k_b, k_c, t \to \infty)$$

$$= \frac{1}{2} \int_{-2k_{max}}^{2k_{max}} d\eta \int_{-2k_{max}+|\eta|}^{2k_{max}-|\eta|} d\Lambda \; \xi_{bc}^*(\eta, \Lambda, 0) \; \xi_{bc}(\eta, \Lambda, t \to \infty). \qquad (2.35)$$

Fidelity is the overlap of the final state with the initial one. Note that the right-hand side of this equation is always a real quantity. Unlike in a Kerr medium, the phase ϕ, here, can take only two values, zero or π. The quantity in Eq. (2.35) would be equal to -1 for the ideal transformation in Eq. (2.2).

On substituting Eq. (2.34) in Eq. (2.35), we get

$$\sqrt{F}e^{i\phi} = \frac{1}{2} \int_{-2k_{max}}^{2k_{max}} d\eta \int_{-2k_{max}+|\eta|}^{2k_{max}-|\eta|} d\Lambda \; |\xi_{bc}(\eta, \Lambda, 0)|^2 - \frac{1}{4k_{max}} \int_{-2k_{max}}^{2k_{max}} d\eta$$

$$\times \left| \int_{-2k_{max}+|\eta|}^{2k_{max}-|\eta|} d\Lambda \; \xi_{bc}(\eta, \Lambda, 0) \right|^2 . \qquad (2.36)$$

This is the most general expression for a combined fidelity and phase, for an arbitrary initial state. Next we shall calculate the fidelity for two different kinds of initial pulses viz. a Gaussian and a hyperbolic secant. First we will do the calculation for a Gaussian pulse of the form $\xi_{bc}(\eta, \Lambda, 0) = 1/(\sigma\sqrt{\pi}) \; e^{-(\eta^2+\Lambda^2)/4\sigma^2}$.

For this case, Eq. (2.36) can be rewritten as

$$\sqrt{F}e^{i\phi} = \underbrace{\frac{1}{2\pi\sigma^2} \int_{-2k_{max}}^{2k_{max}} d\eta \int_{-2k_{max}+|\eta|}^{2k_{max}-|\eta|} d\Lambda \ e^{-(\eta^2+\Lambda^2)/2\sigma^2}}_{I_1}$$

$$-\underbrace{\frac{1}{4\pi\sigma^2 k_{max}} \int_{-2k_{max}}^{2k_{max}} d\eta \left| \int_{-2k_{max}+|\eta|}^{2k_{max}-|\eta|} d\Lambda \ e^{-(\eta^2+\Lambda^2)/2\sigma^2} \right|^2}_{I_2}. \qquad (2.37)$$

Our next task is to evaluate the two integrals I_1 and I_2.

$$I_1 = \frac{2}{\pi\sigma^2} \int_0^{2k_{max}} d\eta \ e^{-\eta^2/2\sigma^2} \underbrace{\int_0^{2k_{max}-\eta} d\Lambda \ e^{-\Lambda^2/2\sigma^2}}_{I_{12}}, \qquad (2.38)$$

where

$$I_{12} = \sigma\sqrt{\frac{\pi}{2}} \ \mathrm{erf}\left(\frac{2k_{max}-\eta}{\sigma\sqrt{2}}\right).$$

In Eq. (2.38), we have exploited a property of the definite integrals for an even integrand, and in the second integral, we have simply set $\Lambda/\sigma\sqrt{2} \equiv y$ and invoked the definition of error function:

$$\mathrm{erf}(x) \equiv \frac{2}{\sqrt{\pi}} \int_0^x dz \ e^{-z^2}.$$

From this intermediate result, it is very clear that it is not possible to get an exact expression for I_1. We have to leave the final result as an integral involving error function.

On explicitly substituting for I_{12} in Eq. (2.38), we obtain

$$
\begin{aligned}
I_1 &= \frac{\sqrt{2}}{\sigma\sqrt{\pi}} \int_0^{2k_{max}} d\eta \; e^{-\eta^2/2\sigma^2} \; \text{erf}\left(\frac{2k_{max} - \eta}{\sigma\sqrt{2}}\right) \\
&= \frac{2}{\sqrt{\pi}} \int_0^{\sqrt{2}k_{max}/\sigma} dx \; e^{-x^2} \text{erf}\left(\frac{\sqrt{2}k_{max}}{\sigma} - x\right),
\end{aligned}
\tag{2.39}
$$

where in the second step of the previous equation, we have set $\eta/\sqrt{2}\sigma \equiv x$.

Following this, we shall now evaluate I_2.

$$
I_2 = \frac{2}{\pi\sigma^2 k_{max}} \int_0^{2k_{max}} d\eta \; e^{-\eta^2/2\sigma^2} \left|\underbrace{\int_0^{2k_{max}-\eta} d\Lambda \; e^{-\Lambda^2/4\sigma^2}}_{I_{21}}\right|^2,
\tag{2.40}
$$

where

$$
I_{21} = \sigma\sqrt{\pi} \; \text{erf}\left(\frac{2k_{max} - \eta}{2\sigma}\right).
$$

We have followed the same procedure to evaluate I_{21} as we did for I_{12}. Just like I_1, it is not possible to get an exact expression for I_2. On substituting explicitly for I_{21} in Eq. (2.40), we get

$$
\begin{aligned}
I_2 &= \frac{2}{k_{max}} \int_0^{2k_{max}} d\eta \; e^{-\eta^2/2\sigma^2} \left|\text{erf}\left(\frac{2k_{max} - \eta}{2\sigma}\right)\right|^2 \\
&= \frac{2\sqrt{2}\sigma}{k_{max}} \int_0^{\sqrt{2}k_{max}/\sigma} dx \; e^{-x^2} \left|\text{erf}\left[\frac{1}{\sqrt{2}}\left(\frac{\sqrt{2}k_{max}}{\sigma} - x\right)\right]\right|^2,
\end{aligned}
\tag{2.41}
$$

where in the second step of the previous equation, we have once again set $\eta/\sqrt{2}\sigma \equiv x$.

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On substituting Eqs. (2.39) and (2.41) in Eq. (2.37), we obtain

$$\sqrt{F}e^{i\phi} = \frac{2}{\sqrt{\pi}} \int_0^{\sqrt{2}k_{max}/\sigma} dx \ e^{-x^2} \ \text{erf}\left(\frac{\sqrt{2}k_{max}}{\sigma} - x\right) - 2\sqrt{2} \frac{\sigma}{k_{max}}$$
$$\times \int_0^{\sqrt{2}k_{max}/\sigma} dx \ e^{-x^2} \left| \text{erf}\left[\frac{1}{\sqrt{2}}\left(\frac{\sqrt{2}k_{max}}{\sigma} - x\right)\right]\right|^2. \quad (2.42)$$

For a normalized Gaussian pulse, $\xi(k) = (1/\sigma^{1/2}\pi^{1/4}) \ e^{-k^2/2\sigma^2}$, the variance of k is related to σ by $\langle k^2 \rangle = \sigma^2/2$. This result is straightforward.

$$\langle k^2 \rangle = \int_{-\infty}^{\infty} dk \ k^2 \ |\xi(k)|^2 = \frac{1}{\sigma\sqrt{\pi}} \int_{-\infty}^{\infty} dk \ k^2 \ e^{-k^2/\sigma^2} = \frac{\sigma^2}{2}.$$

The bandwidth of the medium is also defined by the variance of its spectral distribution. In this case, we have a rectangle function from $-k_{max}$ to k_{max}. Thus, the normalized spectral distribution of the medium is given by $f(k) = 1/\sqrt{2k_{max}}$ and $\Delta k = k_{max}/\sqrt{3}$ which can be easily shown:

$$(\Delta k)^2 = \int_{-k_{max}}^{k_{max}} dk \ k^2 \ |f(k)|^2 = \frac{1}{2k_{max}} \int_{-k_{max}}^{k_{max}} dk \ k^2 = \frac{k_{max}^2}{3}.$$

We shall introduce a dimensionless parameter $\alpha \equiv \Delta k/\sqrt{\langle k^2 \rangle} = \sqrt{2}k_{max}/\sqrt{3}\sigma$, in terms of which Eq. (2.42) can be rewritten as

$$\sqrt{F}e^{i\phi} = \frac{2}{\sqrt{\pi}} \int_0^{\sqrt{3}\alpha} dx \ e^{-x^2} \ \text{erf}(\sqrt{3}\alpha - x) - \frac{4}{\sqrt{3}\alpha} \int_0^{\sqrt{3}\alpha} dx \ e^{-x^2}$$

$$\times \left| \text{erf}\left[\frac{1}{\sqrt{2}}(\sqrt{3}\alpha - x) \right] \right|^2. \qquad (2.43)$$

The parameter α is the ratio of the medium's bandwidth to the rms frequency spread of

the incoming pulse.
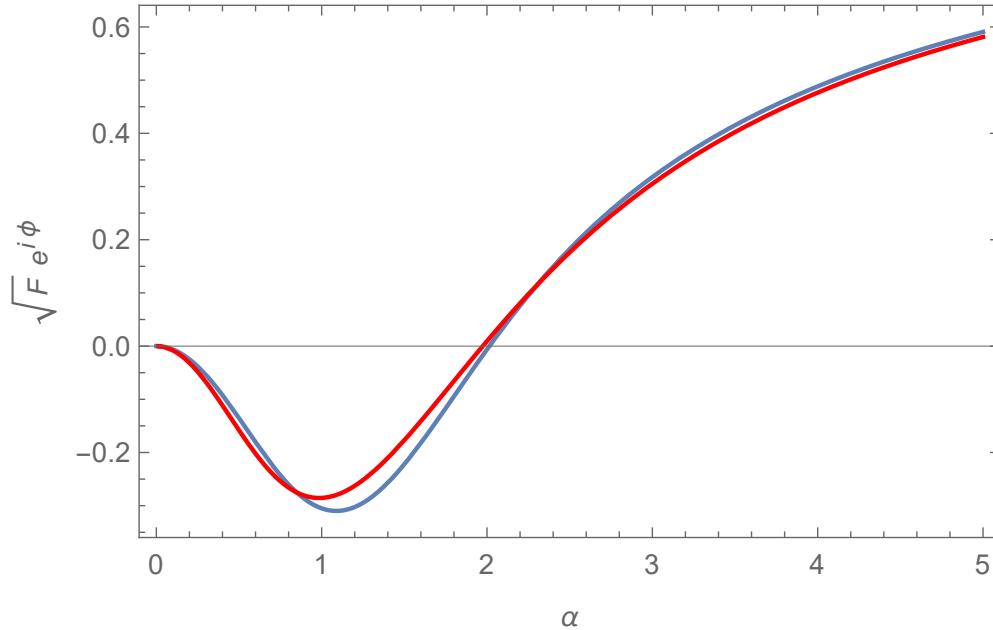
Figure 2.1: Square-root fidelity and phase for hyperbolic secant (red) and Gaussian (blue) pulses, as functions of α. In this figure, ϕ has only two values, zero when the overlap is positive and π when it is negative.

The fidelity of a Gaussian pulse is plotted as the blue curve in figure (2.1), for a range

of values of α and we find that that it is always low, i.e. $F < (-0.3)^2 = 0.09$.

Next we shall calculate the fidelity of a hyperbolic secant pulse of the form

$\xi_b(k_b) = 1/(\sqrt{2\sigma}) \operatorname{sech}(k_b/\sigma)$ and $\xi_c(k_c) = 1/(\sqrt{2\sigma}) \operatorname{sech}(k_c/\sigma)$. In terms of the new

variables η and Λ that we introduced earlier, this pulse can be rewritten as

$\xi_{bc}(\eta, \Lambda, 0) = 1/(2\sigma) \operatorname{sech}[(\eta + \Lambda)/2\sigma] \operatorname{sech}[(\eta - \Lambda)/2\sigma]$.

On inserting the hyperbolic secant pulse in Eq. (2.36), we get

$$\sqrt{F}e^{i\phi} = \frac{1}{8\sigma^2} \int_{-2k_{max}}^{2k_{max}} d\eta \int_{-2k_{max}+|\eta|}^{2k_{max}-|\eta|} d\Lambda \operatorname{sech}^2\left(\frac{\eta + \Lambda}{2\sigma}\right) \operatorname{sech}^2\left(\frac{\eta - \Lambda}{2\sigma}\right)$$

$$- \frac{1}{16k_{max}\sigma^2} \int_{-2k_{max}}^{2k_{max}} d\eta \left| \int_{-2k_{max}+|\eta|}^{2k_{max}-|\eta|} d\Lambda \operatorname{sech}\left(\frac{\eta + \Lambda}{2\sigma}\right) \operatorname{sech}\left(\frac{\eta - \Lambda}{2\sigma}\right) \right|^2. \quad (2.44)$$

Here, it is effective to use the following identity:

$$\operatorname{sech}\left(\frac{\eta + \Lambda}{2\sigma}\right) \operatorname{sech}\left(\frac{\eta - \Lambda}{2\sigma}\right) = \frac{2}{\cosh(\eta/\sigma) + \cosh(\Lambda/\sigma)}.$$

On using this identity in Eq. (2.44), we get

$$\sqrt{F}e^{i\phi} = \underbrace{\frac{1}{2\sigma^2} \int_{-2k_{max}}^{2k_{max}} d\eta \int_{-2k_{max}+|\eta|}^{2k_{max}-|\eta|} d\Lambda \left(\frac{1}{\cosh(\eta/\sigma) + \cosh(\Lambda/\sigma)}\right)^2}_{I_3}$$

$$- \underbrace{\frac{1}{4k_{max}\sigma^2} \int_{-2k_{max}}^{2k_{max}} d\eta \left| \int_{-2k_{max}+|\eta|}^{2k_{max}-|\eta|} d\Lambda \frac{1}{\cosh(\eta/\sigma) + \cosh(\Lambda/\sigma)} \right|^2}_{I_4}. \quad (2.45)$$

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The next task, obviously, is to evaluate I_3 and I_4.

$$I_3 = \frac{2}{\sigma^2} \int_0^{2k_{max}} d\eta \underbrace{\int_0^{2k_{max}-\eta} d\Lambda \left(\frac{1}{\cosh(\eta/\sigma) + \cosh(\Lambda/\sigma)} \right)^2}_{I_{31}}, \qquad (2.46)$$

where

$$I_{31} = \frac{\sigma}{2} \operatorname{cosech}^2\left(\frac{\eta}{\sigma}\right) \left[2 \coth\left(\frac{\eta}{\sigma}\right) \left(\log\left[\cosh\left(\frac{k_{max}}{\sigma}\right)\right] - \log\left[\cosh\left(\frac{k_{max}}{\sigma} - \frac{\eta}{\sigma}\right)\right] \right) \right]$$
$$- \frac{\sigma}{2} \operatorname{cosech}^2\left(\frac{\eta}{\sigma}\right) \operatorname{sech}\left(\frac{k_{max}}{\sigma}\right) \operatorname{sech}\left(\frac{k_{max}}{\sigma} - \frac{\eta}{\sigma}\right) \sinh\left(\frac{2k_{max}}{\sigma} - \frac{\eta}{\sigma}\right).$$

In Eq. (2.46), we have once again invoked the property of an even integrand over a symmetric range of integration and set $\Lambda/\sigma \equiv x$ to compute I_{31}. On explicitly substituting for I_{31} in Eq. (2.46) and furthermore, setting $\eta/\sigma \equiv y$, we obtain

$$I_3 = 2 \int_0^{2k_{max}/\sigma} dy \operatorname{cosech}^2(y) \coth(y) \left(\left[\log\left(\frac{k_{max}}{\sigma}\right)\right] - \log\left[\cosh\left(\frac{k_{max}}{\sigma} - y\right)\right] \right)$$
$$- \int_0^{2k_{max}/\sigma} dy \operatorname{cosech}^2(y) \operatorname{sech}\left(\frac{k_{max}}{\sigma}\right) \operatorname{sech}\left(\frac{k_{max}}{\sigma} - y\right) \sinh\left(\frac{2k_{max}}{\sigma} - y\right). \qquad (2.47)$$

This is the best we can get analytically for I_3. We cannot obtain a closed form solution for this integral. Following this, we shall evaluate I_4.

$$I_4 = \frac{2}{k_{max}\sigma^2} \int_0^{2k_{max}} d\eta \left| \underbrace{\int_0^{2k_{max}-\eta} d\Lambda \frac{1}{\cosh(\eta/\sigma) + \cosh(\Lambda/\sigma)}}_{I_{41}} \right|^2, \qquad (2.48)$$

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where

$$I_{41} = \sigma \operatorname{cosech}\left(\frac{\eta}{\sigma}\right)\left(\log\left[\cosh\left(\frac{k_{max}}{\sigma}\right)\right] - \log\left[\cosh\left(\frac{k_{max}}{\sigma} - \frac{\eta}{\sigma}\right)\right]\right).$$

We have followed the same procedure here that we did to evaluate I_3. It is evident from this result that it not possible to get an exact expression for I_4 either. We have to leave the final result as an integral. On explicitly substituting for I_{41} in Eq. (2.48) and once again setting $\eta/\sigma \equiv y$, we obtain

$$I_4 = \frac{2\sigma}{k_{max}}\int_0^{2k_{max}/\sigma} dy \operatorname{cosech}^2(y)\left(\log\left[\cosh\left(\frac{k_{max}}{\sigma}\right)\right] - \log\left[\cosh\left(\frac{k_{max}}{\sigma} - y\right)\right]\right)^2.$$

(2.49)

Now, on directly substituting Eqs. (2.47) and (2.49) in Eq. (2.45), we get the following expression for fidelity of a hyperbolic secant pulse:

$$\sqrt{F}e^{i\phi} = 2\int_0^{2k_{max}/\sigma} dy \operatorname{cosech}^2(y)\coth(y)\left(\left[\log\left(\frac{k_{max}}{\sigma}\right)\right] - \log\left[\cosh\left(\frac{k_{max}}{\sigma} - y\right)\right]\right)$$
$$- \int_0^{2k_{max}/\sigma} dy \operatorname{cosech}^2(y)\operatorname{sech}\left(\frac{k_{max}}{\sigma}\right)\operatorname{sech}\left(\frac{k_{max}}{\sigma} - y\right)\sinh\left(\frac{2k_{max}}{\sigma} - y\right)$$
$$- \frac{2\sigma}{k_{max}}\int_0^{2k_{max}/\sigma} dy \operatorname{cosech}^2(y)\left(\log\left[\cosh\left(\frac{k_{max}}{\sigma}\right)\right] - \log\left[\cosh\left(\frac{k_{max}}{\sigma} - y\right)\right]\right)^2.$$

(2.50)

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For a normalized hyperbolic secant pulse, the variance of k is related to σ by $\langle k^2 \rangle = \pi^2 \sigma^2 / 2$ which can be easily shown:

$$\langle k^2 \rangle = \int_{-\infty}^{\infty} dk \ k \ |\xi(k)|^2 = \frac{1}{2\sigma} \int_{-\infty}^{\infty} dk \ k^2 \ \text{sech}^2(k/\sigma) = \pi^2 \sigma^2 / 12.$$

We have already seen earlier that for the medium, $\Delta k = k_{max}/\sqrt{3}$. Once again, we shall define $\alpha \equiv \Delta k / \sqrt{\langle k \rangle^2}$. For a hyperbolic secant pulse, we have $\alpha = 2k_{max}/\pi\sigma$, in terms of which we can rewrite Eq. (2.50) as

$$\sqrt{F}e^{i\phi} = 2 \int_0^{\pi\alpha} dy \ \text{cosech}^2(y) \ \text{coth}(y) \ \left(\left[\log \left(\frac{\pi}{2}\alpha \right) \right] - \log \left[\cosh \left(\frac{\pi}{2}\alpha - y \right) \right] \right)$$
$$- \int_0^{\pi\alpha} dy \ \text{cosech}^2(y) \ \text{sech} \left(\frac{\pi}{2}\alpha \right) \ \text{sech} \left(\frac{\pi}{2}\alpha - y \right) \ \sinh (\pi\alpha - y)$$
$$- \frac{4}{\pi\alpha} \int_0^{\pi\alpha} dy \ \text{cosech}^2(y) \left(\log \left[\cosh \left(\frac{\pi}{2}\alpha \right) \right] - \log \left[\cosh \left(\frac{\pi}{2}\alpha - y \right) \right] \right)^2 . \qquad (2.51)$$

The fidelity of a hyperbolic secant pulse is plotted as the red curve in figure (2.1). It is very clear from this figure that for both these pulses, the fidelity is always low. The limit $\alpha \to 0$ corresponds to the narrow bandwidth regime. Reducing the medium bandwidth results in less and less of the wavepacket being transmitted through the medium which is why the fidelity (F) goes to zero as $\alpha \to 0$. On the other hand, the opposite limit $\alpha \to \infty$ corresponds to the large bandwidth regime which represents fast nonlinearity. In other words, in this limit the response time of the medium is much shorter compared to the duration of the pulses. Thus, in a long pulse, the probability that two photons could randomly be found within the same narrow time window corresponding to the response

time of the medium, is negligible as a result of which the nonlinear effects vanish in this limit. This simply means that when the bandwidth of the medium is large, the two photons propagate without interacting with the medium which is why $F \to 1$ as $\alpha \to \infty$. In both the cases, the largest overlap with the initial state (with a π phase shift) happens around $\alpha = 1$. In this regime, the medium bandwidth and the pulse bandwidth are evenly matched, in the frequency domain.

Based on the analytical calculation, the conclusion that we get a π phase shift when the medium bandwidth and the pulse bandwidth are of the same order of magnitude invalidates some of the approximations we made in our theoretical analysis, particularly the introduction of delta functions in Eqs. (2.15) and (2.17) under the assumption that the medium's "acceptance bandwidth" is much larger than the rms frequency of the pulse.

So, in order to get a better understanding of the result in this crucial region ($\alpha = 1$), we have carried out a numerical integration of Eq. (2.11) without any further approximation and assumption. For our numerical calculation, we place the pulse and the medium (with a set of discrete modes) in a region of space of length L with periodic boundary conditions, and integrate for one round trip. Changing the bandwidth of the medium is equivalent to changing the number of modes used in the calculations. So the result is given by a set of discrete points in figure (2.2). For each point, i.e. for a given number of modes, we have looked for the value of ϵ that optimizes the fidelity.

Figure (2.2) shows that our analytical calculation underestimates the achievable fidelity around $\alpha \simeq 1$. However, the fidelity still remains small ($F < 0.36$). For an initial Gaussian pulse, two sets of results are plotted in figure (2.2). In the first case, we simply plot the overlap between the final and initial state as given by the numerical calculation and filtered

by the medium. Here, the norm of the final state will most likely be less than 1, reflecting the possibility of absorption of photons in the medium. In the second case which is more favorable, we renormalize the final $b - c$ state before calculating the overlap with the initial state. This means that we are calculating a "conditional" fidelity on the assumption that the b and the c photons do not get absorbed in the medium.
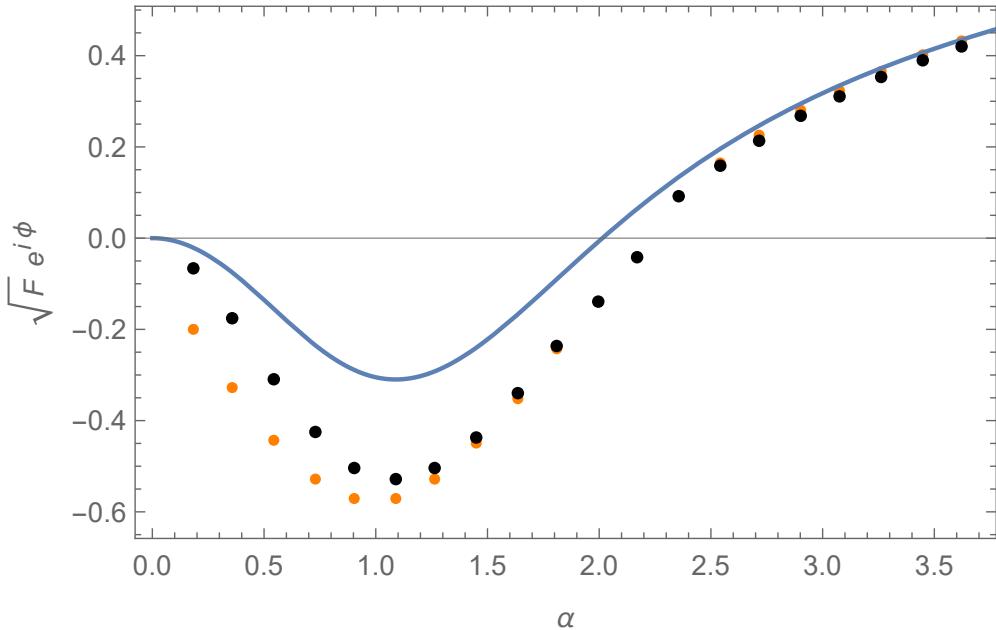
Figure 2.2: Numerically calculated fidelity for Gaussian pulses (dots) compared to the analytical approximation (continuous line). The darker dots (upper trace) show the result for the un-normalized wavepacket, so they implicitly include the effect of medium absorption. The lighter dots (lower trace) use a renormalized wave function, so they give the fidelity conditioned on the photons not being absorbed.

2.3   Cavity configuration

In this section, we place the nonlinear medium inside a one-sided optical cavity. One of the primary reasons to study this system is because cavities can help enhance weak

nonlinearities. Besides, a cavity provides a natural bandwidth for the system through the field's decay rate, and as an added incentive, in the absence of other losses, it provides us with a setup for which an analytical solution can be obtained.

The system comprising of a nonlinear medium in a cavity, and neglecting absorption losses or spontaneous emission into off-axis modes, can be considered to be a closed system that can be treated by the Hamiltonian formalism. Once again, we shall work in the continuous mode description in the interaction picture, but here, for notational convenience, we label the modes by frequency instead of wave vector.

The most general state is written as

$$|\psi(t)\rangle = \int d\omega \, \xi_a(\omega, t) \, \hat{a}_\omega^\dagger |0\rangle_a |0\rangle_b |0\rangle_c + \int d\omega' \int d\omega'' \, \xi_{bc}(\omega'.\omega'', t) \, |0\rangle_a \, \hat{b}_{\omega'}^\dagger |0\rangle_b \, \hat{c}_{\omega''}^\dagger |0\rangle_c \quad (2.52)$$

and the Hamiltonian corresponding to the nonlinear interaction in a cavity is written as

$$\hat{H} = \hbar g[\hat{A}^\dagger(t) \, \hat{B}(t) \, \hat{C}(t) + \hat{A}(t) \, \hat{B}^\dagger(t) \, \hat{C}^\dagger(t)], \quad (2.53)$$

where g is the coupling strength and $\hat{A}(t)$, $\hat{B}(t)$ and $\hat{C}(t)$ are the cavity quasimode operators, which in the continuous mode formalism [14, 15] can be written as

$$\hat{A}(t) = \int d\omega \frac{\sqrt{\kappa/\pi}}{\kappa - i(\Delta_a + \omega)} \hat{a}_\omega e^{-i\omega t},$$

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

$$\hat{C}(t) = \int d\omega \frac{\sqrt{\kappa/\pi}}{\kappa - i(\Delta_c + \omega)} \hat{c}_\omega e^{-i\omega t}. \tag{2.54}$$

In Eq. (2.54), κ is the cavity decay rate and Δ_a, Δ_b and Δ_c are the cavity-field

detunings for the a, b and c photons, respectively. The operators $\hat{a}_\omega, \hat{b}_\omega$ and $\hat{c}_\omega$ obey the

canonical commutation relations: $[\hat{a}_\omega, \hat{a}_{\omega'}^\dagger] = [\hat{b}_\omega, \hat{b}_{\omega'}^\dagger] = [\hat{c}_\omega, \hat{c}_{\omega'}^\dagger] = \delta(\omega - \omega')$.

Assuming a doubly resonant cavity, i.e. $\Delta_a = \Delta_b = \Delta_c = 0$ and on inserting Eqs. (2.52),

(2.53) and (2.54) in the Schrödinger equation: $|\dot{\psi}\rangle = -(i/\hbar)\hat{H}|\psi\rangle$, we get the following pair

of differential equations for the a and the b, c pulses:

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

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

$$\tag{2.55}$$

which can be solved by the method of Laplace transform [16]. The Laplace transform of

the system in Eqs. (2.55) is given by

$$s\,\tilde{\xi}_a(\omega, s) - \xi_a(\omega, 0) = -ig\left(\frac{\kappa}{\pi}\right)^{3/2} \int d\omega' \int d\omega'' \frac{\tilde{\xi}_{bc}(\omega', \omega'', s + i(\omega' + \omega'' - \omega))}{(\kappa + i\omega)(\kappa - i\omega')(\kappa - i\omega'')}, \tag{2.56}$$

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and

$$s\,\tilde{\xi}_{bc}(\omega',\omega'',s) - \xi_{bc}(\omega',\omega'',0) = -ig\left(\frac{\kappa}{\pi}\right)^{3/2}\int d\omega\,\frac{\tilde{\xi}_a(\omega,s+i(\omega-\omega'-\omega''))}{(\kappa-i\omega)(\kappa+i\omega')(\kappa+i\omega'')}, \qquad (2.57)$$

where $\tilde{\xi}_a$ and $\tilde{\xi}_{bc}$ are the Laplace transform of ξ_a and ξ_{bc}, respectively. Our next step is to substitute for $\tilde{\xi}_{bc}(\omega',\omega'',s+i(\omega'+\omega''-\omega))$ in Eq. (2.56) in terms of $\tilde{\xi}_a$ using Eq. (2.57). In Eq. (2.57), on shifting $s \to s+i(\omega'+\omega''-\omega)$, we get

$$\begin{aligned}\tilde{\xi}_{bc}(\omega',\omega'',s+i(\omega'+\omega''-\omega)) =&\frac{\xi_{bc}(\omega',\omega'',0)}{s+i(\omega'+\omega''-\omega)} - ig\left(\frac{\kappa}{\pi}\right)^{3/2}\frac{1}{s+i(\omega'+\omega''-\omega)}\\ &\times\int d\omega'''\,\frac{\tilde{\xi}_a(\omega''',s+i(\omega'''-\omega))}{(\kappa-i\omega''')(\kappa+i\omega')(\kappa+i\omega'')}.\end{aligned} \qquad (2.58)$$

Now, on directly substituting Eq. (2.58) in Eq. (2.56) and furthermore setting $\xi_a(\omega,0)=0$ since there is no a photon at $t=0$, we obtain

$$\begin{aligned}\tilde{\xi}_a(\omega,s) =& -ig\left(\frac{\kappa}{\pi}\right)^{3/2}\frac{1}{s(\kappa+i\omega)}\int d\omega'\int d\omega''\frac{\xi_{bc}(\omega',\omega'',0)}{[s+i(\omega'+\omega''-\omega)](\kappa-i\omega')(\kappa-i\omega'')}\\ &-\left(\frac{\kappa}{\pi}\right)^3 g^2\frac{1}{s(\kappa+i\omega)}\underbrace{\int d\omega'\int d\omega''\frac{1}{[s+i(\omega'+\omega''-\omega)](\kappa^2+\omega'^2)(\kappa^2+\omega''^2)}}_{I}\\ &\times\int d\omega'''\,\frac{\tilde{\xi}_a(\omega''',s+i(\omega'''-\omega))}{\kappa-i\omega'''},\end{aligned}$$

$$(2.59)$$

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where

$$I = \left(\frac{\pi}{\kappa}\right)^2 \frac{1}{s + 2\kappa - i\omega}.$$

On explicitly substituting for I in Eq. (2.59), we get

$$\tilde{\xi}_a(\omega, s) = -ig \left(\frac{\kappa}{\pi}\right)^{3/2} \frac{1}{s(\kappa + i\omega)} \int d\omega' \int d\omega'' \frac{\xi_{bc}(\omega', \omega'', 0)}{[s + i(\omega' + \omega'' - \omega)](\kappa - i\omega')(\kappa - i\omega'')}$$
$$- \left(\frac{\kappa}{\pi}\right) g^2 \frac{1}{s(\kappa + i\omega)} \frac{1}{s + 2\kappa - i\omega} \int d\omega''' \frac{\tilde{\xi}_a(\omega''', s + i(\omega''' - \omega))}{\kappa - i\omega'''}. \tag{2.60}$$

In order to make our calculation more compact, we shall define the following two functions:

$$F(\omega, s) \equiv -ig \left(\frac{\kappa}{\pi}\right)^{3/2} \frac{1}{s(\kappa + i\omega)} \int d\omega' \int d\omega'' \frac{\xi_{bc}(\omega', \omega'', 0)}{[s + i(\omega' + \omega'' - \omega)](\kappa - i\omega')(\kappa - i\omega'')}$$

and

$$E(s - i\omega) \equiv \left(\frac{\kappa}{\pi}\right) g^2 \frac{1}{s + 2\kappa - i\omega}.$$

In terms of these new definitions, Eq. (2.60) can be rewritten as

$$\tilde{\xi}_a(\omega, s) = F(\omega, s) - \frac{1}{s} \frac{E(s - i\omega)}{\kappa + i\omega} \int d\omega''' \frac{\tilde{\xi}_a(\omega''', s + i(\omega''' - \omega))}{\kappa - i\omega'''}. \tag{2.61}$$

Eq. (2.61) can be solved by shifting to dummy arguments in the same equation, viz. $\omega \to \omega'$ and $s \to s + i(\omega' - \omega)$.

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This yields,

$$\tilde{\xi}_a(\omega', s + i(\omega' - \omega)) = F(\omega', s + i(\omega' - \omega)) - \frac{1}{s + i(\omega' - \omega)} \frac{E(s - i\omega)}{\kappa + i\omega'}$$
$$\times \int d\omega''' \frac{\tilde{\xi}_a(\omega''', s + i(\omega''' - \omega))}{\kappa - i\omega'''}. \quad (2.62)$$

Following this, we shall divide both sides of the previous equation by $\kappa - i\omega'$ and integrate over ω' which results in

$$\underbrace{\int d\omega' \frac{\tilde{\xi}_a(\omega', s + i(\omega' - \omega))}{\kappa - i\omega'}}_{\tilde{I}} = \int d\omega' \frac{F(\omega', s + i(\omega' - \omega))}{\kappa - i\omega'} - E(s - i\omega')$$
$$\times \underbrace{\int d\omega' \frac{1}{(s + i(\omega' - \omega))(\kappa^2 + \omega'^2)}}_{I_I}$$
$$\times \underbrace{\int d\omega''' \frac{\tilde{\xi}_a(\omega''', s + i(\omega''' - \omega))}{\kappa - i\omega'''}}_{\tilde{I}}, \quad (2.63)$$

where

$$I_I = \frac{\pi}{\kappa} \frac{1}{s + \kappa - i\omega}.$$

On explicitly substituting for I_I in Eq. (2.63), we can easily solve for $\tilde{I}$ which is given by

$$\tilde{I} = \int d\omega' \frac{F(\omega', s + i(\omega' - \omega))}{\kappa - i\omega'} - \frac{\pi}{\kappa} \frac{E(s - i\omega)}{s + \kappa - i\omega} \tilde{I},$$
$$\tilde{I} = \left(1 + \frac{\pi}{\kappa} \frac{E(s - i\omega)}{s + \kappa - i\omega}\right)^{-1} \int d\omega' \frac{F(\omega', s + i(\omega' - \omega))}{\kappa - i\omega'}. \quad (2.64)$$

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Next, we shall substitute the explicit expression for $\tilde{I}$ in Eq. (2.61), which yields

$$
\tilde{\xi}_a(\omega, s) = F(\omega, s) - \frac{1}{s} \frac{E(s - i\omega)}{\kappa + i\omega} \left(1 + \frac{\pi}{\kappa} \frac{E(s - i\omega)}{s + \kappa - i\omega} \right)^{-1}
$$
$$
\times \int d\omega''' \frac{F(\omega''', s + i(\omega''' - \omega))}{\kappa - i\omega'''}, \qquad (2.65)
$$

where in the previous equation, we have replaced the variable of integration in the last integral to ω'''. In the definition of F, on shifting $\omega \to \omega'''$ and $s \to s + i(\omega''' - \omega)$, we get the following explicit form for $F(\omega''', s + i(\omega''' - \omega))$:

$$
F(\omega''', s + i(\omega''' - \omega)) = -ig \left(\frac{\kappa}{\pi} \right)^{3/2} \frac{1}{[s + i(\omega''' - \omega)] \, (\kappa + i\omega''')}
$$
$$
\times \int d\omega' \int d\omega'' \frac{\xi_{bc}(\omega', \omega'', 0)}{[s + i(\omega' + \omega'' - \omega)](\kappa - i\omega')(\kappa - i\omega'')}. \quad (2.66)
$$

On directly substituting Eq. (2.66) and the explicit form for E [see below Eq. (2.60)] in Eq. (2.65), we obtain

$$
\tilde{\xi}_a(\omega, s) = -ig \left(\frac{\kappa}{\pi} \right)^{3/2} \frac{1}{s(\kappa + i\omega)} \int d\omega' \int d\omega'' \frac{\xi_{bc}(\omega', \omega'', 0)}{[s + i(\omega' + \omega'' - \omega)](\kappa - i\omega')(\kappa - i\omega'')}
$$
$$
+ ig^3 \left(\frac{\kappa}{\pi} \right)^{5/2} \frac{1}{s(\kappa + i\omega)} \frac{s + \kappa - i\omega}{g^2 + (s + 2\kappa - i\omega)(s + \kappa - i\omega)} \int d\omega''' \frac{1}{[s + i(\omega''' - \omega)](\kappa^2 + \omega'''^2)}
$$
$$
\times \int d\omega' \int d\omega'' \frac{\xi_{bc}(\omega', \omega'', 0)}{[s + i(\omega' + \omega'' - \omega)](\kappa - i\omega')(\kappa - i\omega'')}. \qquad (2.67)
$$

The next step is to substitute for $\tilde{\xi}_a$ in Eq. (2.57) in terms of $\xi_{bc}(\omega', \omega'', 0)$ from Eq. (2.67). In the previous equation, on shifting to dummy arguments viz. $\omega \to \omega'''$ and $s \to s + i(\omega''' - \omega' - \omega'')$, we get

$$
\begin{aligned}
\tilde{\xi}_a(\omega''', s + i(\omega''' - \omega' - \omega'')) = {} & -ig \left(\frac{\kappa}{\pi}\right)^{3/2} \frac{1}{[s + i(\omega''' - \omega' - \omega'')](\kappa + i\omega''')} \\
& \times \int d\omega_1 \int d\omega_2 \, \frac{\xi_{bc}(\omega_1, \omega_2, 0)}{[s + i(\omega_1 + \omega_2 - \omega' - \omega'')](\kappa - i\omega_1)(\kappa - i\omega_2)} \\
& + ig^3 \left(\frac{\kappa}{\pi}\right)^{5/2} \frac{1}{[s + i(\omega''' - \omega' - \omega'')](\kappa + i\omega''')} \\
& \times \frac{s + \kappa - i(\omega' + \omega'')}{g^2 + [s + 2\kappa - i(\omega' + \omega'')] \, [s + \kappa - i(\omega' + \omega'')]} \\
& \times \int d\omega_3 \, \frac{1}{[s + i(\omega_3 - \omega' - \omega'')] \, (\kappa^2 + \omega_3^2)} \\
& \times \int d\omega_1 \int d\omega_2 \, \frac{\xi_{bc}(\omega_1, \omega_2, 0)}{[s + i(\omega_1 + \omega_2 - \omega' - \omega'')] \, (\kappa - i\omega_1) \, (\kappa - i\omega_2)}.
\end{aligned} \tag{2.68}
$$

This can be directly substituted in Eq. (2.57) which finally yields

$$
\begin{aligned}
\tilde{\xi}_{bc}(\omega', \omega'', s) = {} & \frac{1}{s} \, \xi_{bc}(\omega', \omega'', 0) - g^2 \left(\frac{\kappa}{\pi}\right)^3 \frac{1}{s} \int d\omega''' \frac{1}{[s + i(\omega''' - \omega' - \omega'')](\kappa^2 + \omega'''^2)} \\
& \times \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')} \int d\omega_1 \int d\omega_2 \frac{\xi_{bc}(\omega_1, \omega_2, 0)}{[s + i(\omega_1 + \omega_2 - \omega' - \omega'')](\kappa - i\omega_1)(\kappa - i\omega_2)} \\
& + g^4 \left(\frac{\kappa}{\pi}\right)^4 \frac{1}{s} \int d\omega''' \frac{1}{s + i(\omega''' - \omega' - \omega'')} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')(\kappa^2 + \omega'''^2)} \\
& \times \frac{s + \kappa - i(\omega' + \omega'')}{g^2 + (s + 2\kappa - i(\omega' + \omega''))(s + \kappa - i(\omega' + \omega''))} \\
& \times \int d\omega_3 \, \frac{1}{[s + i(\omega_3 - \omega' - \omega'')](\kappa^2 + \omega_3^2)} \\
& \times \int d\omega_1 \int d\omega_2 \, \frac{\xi_{bc}(\omega_1, \omega_2, 0)}{[s + i(\omega_1 + \omega_2 - \omega' - \omega'')](\kappa - i\omega_1)(\kappa - i\omega_2)}.
\end{aligned} \tag{2.69}
$$

Eq. (2.69) is the formal solution to our problem in the s-domain. In order to get the formal solution as a function of time, we have to invert the previous equation. But full inversion of Eq. (2.69) could be substantially complicated. In many cases, it might not even be possible. However, we are only interested in the pulse long after its interaction with the nonlinear medium inside the cavity. In other words, we only require the asymptotic state, i.e. $\lim_{t \to \infty} \xi_{bc}(\omega', \omega'', t)$. We can then exploit the final value theorem of operational calculus which says that $\lim_{t \to \infty} \xi_{bc}(\omega', \omega'', t) = \lim_{s \to 0} s\, \tilde{\xi}_{bc}(\omega', \omega'', s)$.

This gives us

$$
\lim_{s \to 0} \left[s\, \tilde{\xi}_{bc}(\omega', \omega'', s) \right] = \xi_{bc}(\omega', \omega'', 0) - g^2 \left(\frac{\kappa}{\pi} \right)^3 \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')}
$$

$$
\times \lim_{s \to 0} \underbrace{\int d\omega''' \frac{1}{[s + i(\omega''' - \omega' - \omega'')](\kappa^2 + \omega'''^2)}}_{I_1}
$$

$$
\times \lim_{s \to 0} \underbrace{\int d\omega_1 \int d\omega_2 \frac{\xi_{bc}(\omega_1, \omega_2, 0)}{[s + i(\omega_1 + \omega_2 - \omega' - \omega'')](\kappa - i\omega_1)(\kappa - i\omega_2)}}_{I_2}
$$

$$
+ g^4 \left(\frac{\kappa}{\pi} \right)^4 \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')} \lim_{s \to 0} \underbrace{\int d\omega''' \frac{1}{[s + i(\omega''' - \omega' - \omega'')](\kappa^2 + \omega'''^2)}}_{I_1}
$$

$$
\times \frac{\kappa - i(\omega' + \omega'')}{g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')]}
$$

$$
\times \lim_{s \to 0} \underbrace{\int d\omega_3 \frac{1}{[s + i(\omega_3 - \omega' - \omega'')](\kappa^2 + \omega_3^2)}}_{I_1}
$$

$$
\times \lim_{s \to 0} \underbrace{\int d\omega_1 \int d\omega_2 \frac{\xi_{bc}(\omega_1, \omega_2, 0)}{[s + i(\omega_1 + \omega_2 - \omega' - \omega'')](\kappa - i\omega_1)(\kappa - i\omega_2)}}_{I_2}, \tag{2.70}
$$

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where

$$I_1 = \frac{\pi}{\kappa} \frac{1}{\kappa - i(\omega' + \omega'')},$$

and

$$I_2 = 2\pi \int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega' + \omega'' - \omega_1, 0)}{(\kappa - i\omega_1)[\kappa - i(\omega' + \omega'' - \omega_1)]}.$$

The integral I_2 is evaluated from the calculus of residues. We have computed this integral over ω_2 and expressed the result in terms of an integral over ω_1. We shall assume that the initial state $\xi_{bc}(\omega_1, \omega_2, 0)$ is factorizable, i.e. $\xi_{bc}(\omega_1, \omega_2, 0) = \xi_b(\omega_1, 0)\ \xi_c(\omega_2, 0)$. It is legitimate to assume that the wavepacket $\xi(\omega)$ vanishes as $\omega \to \infty$ since it must be normalizable. Since we are integrating over ω_2, what we need to compute is

$$\int d\omega_2 \frac{\xi_c(\omega_2)}{[s + i(\omega_1 + \omega_2 - \omega' - \omega'')](\kappa - i\omega_2)}.$$

Note that $\xi_c(\omega_2)$ is the Fourier transform of the incoming pulse, which we we may take to vanish for $t < 0$. Thus, the function $\xi_c(\omega_2)$ ought to be of the form

$$\xi_c(\omega_2) \sim \int_0^\infty \xi_c(t)\ e^{i\omega_2 t}\ dt.$$

Since $t > 0$, we close the contour using a semicircle in the upper half of the complex ω_2 plane [i.e. $\text{Im}(\omega_2) > 0$] and then we shall evaluate the integral using the theorem of residues. Since we are in the upper half plane, only the poles with positive imaginary part

will contribute to the result. In the integral over ω_2, of the two terms in the denominator, only $[s + i(\omega_1 + \omega_2 - \omega' - \omega'')]$ has a pole with a positive imaginary part, i.e. for this term, the pole is at $\omega_2 = \omega' + \omega'' - \omega_1 + is$. So we just need to calculate the residue at this pole which then gives us the final result for I_2.

Next, on explicitly substituting for I_1 and I_2 in Eq. (2.70) and carrying out some simplification, we obtain

$$\lim_{s \to 0} [s \, \tilde{\xi}_{bc}(\omega', \omega'', s)] = \xi_{bc}(\omega', \omega'', 0) - \frac{2g^2 \kappa^2}{\pi} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')}$$
$$\times \frac{2\kappa - i(\omega' + \omega'')}{g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')]}$$
$$\times \int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega' + \omega'' - \omega_1, 0)}{(\kappa - i\omega_1)[\kappa - i(\omega' + \omega'' - \omega_1)]}. \tag{2.71}$$

The integral in Eq. (2.71) can be simplified by making use of a partial fraction decomposition. Note that

$$\frac{1}{(\kappa - i\omega_1)[\kappa - i(\omega' + \omega'' - \omega_1)]} = \frac{1}{2\kappa - i(\omega' + \omega'')} \left[\frac{1}{\kappa - i\omega_1} + \frac{1}{\kappa - i(\omega' + \omega'' - \omega_1)} \right]. \tag{2.72}$$

On substituting Eq. (2.72) in Eq. (2.71), we get

$$\lim_{s \to 0} \left[s\, \tilde{\xi}_{bc}(\omega', \omega'', s) \right] = \xi_{bc}(\omega', \omega'', 0) - \frac{2g^2\kappa^2}{\pi} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')}$$

$$\times \frac{1}{g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')]}$$

$$\times \left(\int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega' + \omega'' - \omega_1, 0)}{\kappa - i\omega_1} + \int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega' + \omega'' - \omega_1, 0)}{\kappa - i(\omega' + \omega'' - \omega_1)} \right) . (2.73)$$

In the second integral on the right hand side of the previous equation, if we set

$\omega' + \omega'' - \omega_1 \equiv \omega$, it becomes identical to the first integral, which then simplifies Eq. (2.74)

to the following form:

$$\lim_{s \to 0} \left[s\, \tilde{\xi}_{bc}(\omega', \omega'', s) \right] = \xi_{bc}(\omega', \omega'', 0) - \frac{4g^2\kappa^2}{\pi} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')}$$

$$\times \frac{1}{g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')]} \int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega' + \omega'' - \omega_1, 0)}{\kappa - i\omega_1}. \quad (2.74)$$

In order to get the actual spectrum of the outgoing field, we should multiply the

expression in Eq. (2.74) by the "empty cavity" factors $(\kappa + i\omega')/(\kappa - i\omega')$ and

$(\kappa + i\omega'')/(\kappa - i\omega'')$.

Thus, the expression for fidelity, i.e. the overlap of the final state with the initial one, is then expressed as

$$
\sqrt{F}e^{i\phi} = \int d\omega' \int d\omega'' \left(\frac{\kappa + i\omega'}{\kappa - i\omega'}\right) \left(\frac{\kappa + i\omega''}{\kappa - i\omega''}\right) \xi_{bc}^*(\omega', \omega'', 0) \lim_{s \to 0} \left[s\, \tilde{\xi}_{bc}(\omega', \omega'', s)\right]
$$

$$
= \int d\omega' \int d\omega'' \left(\frac{\kappa + i\omega'}{\kappa - i\omega'}\right) \left(\frac{\kappa + i\omega''}{\kappa - i\omega''}\right) |\xi_{bc}(\omega', \omega'', 0)|^2 - \frac{4g^2\kappa^2}{\pi}
$$

$$
\times \int d\omega' \int d\omega'' \frac{1}{(\kappa - i\omega')(\kappa - i\omega'')} \frac{\xi_{bc}^*(\omega', \omega'', 0)}{g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')]}
$$

$$
\times \int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega' + \omega'' - \omega_1, 0)}{\kappa - i\omega_1}. \tag{2.75}
$$

We can obtain a particularly simple result in the strong-coupling limit in which g is much greater than both the cavity bandwidth κ and the pulse bandwidth as a result of which we can approximate $g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')] \approx g^2$. In this limit, the expression for square-root fidelity and phase in Eq. (2.75) simplifies to

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

$$
\times \int d\omega' \int d\omega'' \frac{\xi_{bc}^*(\omega', \omega'', 0)}{(\kappa - i\omega')(\kappa - i\omega'')} \int d\omega_1 \frac{\xi_{bc}(\omega_1, \omega' + \omega'' - \omega_1, 0)}{\kappa - i\omega_1}. \tag{2.76}
$$

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

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

On substituting for the initial pulse in Eq. (2.76) in terms of time-domain integrals, we get

$$
\sqrt{F}e^{i\phi} = \frac{1}{4\pi^2} \int d\omega' \int d\omega'' \left(1 - \frac{2\kappa}{\kappa - i\omega'} - \frac{2\kappa}{\kappa - i\omega''} + \frac{4\kappa^2}{(\kappa - i\omega')(\kappa - i\omega'')} \right)
$$

$$
\times \int dt_1\, \xi_b^*(t_1)\, e^{-i\omega' t_1} \int dt_2\, \xi_c^*(t_2)\, e^{-i\omega'' t_2} \int dt_3\, \xi_b(t_3)\, e^{i\omega' t_3}
$$

$$
\times \int dt_4\, \xi_b(t_4)\, e^{i\omega'' t_4} - \left(\frac{\kappa^2}{\pi^3} \right) \int dt_1\, \xi_b^*(t_1) \int dt_2\, \xi_c^*(t_2) \int dt_3\, \xi_b(t_3)
$$

$$
\times \int dt_4\, \xi_b(t_4) \int d\omega'\, \frac{e^{i\omega'(t_4 - t_1)}}{\kappa - i\omega'} \int d\omega''\, \frac{e^{i\omega''(t_4 - t_2)}}{\kappa - i\omega''} \int d\omega_1\, \frac{e^{i\omega_1(t_4 - t_2)}}{\kappa - i\omega_1}. \qquad (2.77)
$$

In the previous equation, we have made use of the following identity in the first term: $(\kappa + i\omega)/(\kappa - i\omega) = -1 + (2\kappa/\kappa - i\omega)$. Note that $\kappa > 0$.

In order to simplify Eq. (2.77) further, we shall repeatedly use the following result:

$$
\int d\omega\, \frac{e^{i\omega(t - \tau)}}{\kappa - i\omega} = \begin{cases} 2\pi\, e^{\kappa(t - \tau)}, & \text{if } t - \tau < 0 \\[2mm] 0, & \text{otherwise.} \end{cases} \qquad (2.78)
$$

In Eq. (2.77), we shall denote the first term as T_1 and the second term as T_2. Thus, on simplifying each term (i.e. T_1 and T_2) in Eq. (2.77) separately using the result in Eq. (2.78), we obtain the following expressions for the two terms:

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$$T_1 \equiv \frac{1}{4\pi^2} \int d\omega' \int d\omega'' \left(1 - \frac{2\kappa}{\kappa - i\omega'} - \frac{2\kappa}{\kappa - i\omega''} + \frac{4\kappa^2}{(\kappa - i\omega')(\kappa - i\omega'')} \right)$$

$$\times \int dt_1 \, \xi_b^*(t_1) \, e^{-i\omega' t_1} \int dt_2 \, \xi_c^*(t_2) \, e^{-i\omega'' t_2} \int dt_3 \, \xi_b(t_3) \, e^{i\omega' t_3}$$

$$\times \int dt_4 \, \xi_b(t_4) \, e^{i\omega'' t_4}$$

$$= 1 - 2\kappa \int_{-\infty}^{\infty} dt_1 \, \xi_b^*(t_1) \, e^{-\kappa t_1} \int_{-\infty}^{t_1} dt_3 \, \xi_b(t_3) \, e^{\kappa t_3} - 2\kappa \int_{-\infty}^{\infty} dt_2 \, \xi_c^*(t_2) \, e^{-\kappa t_2}$$

$$\times \int_{-\infty}^{t_2} dt_4 \, \xi_c(t_4) \, e^{\kappa t_4} + 4\kappa^2 \int_{-\infty}^{\infty} dt_1 \, \xi_b^*(t_1) \, e^{-\kappa t_1} \int_{-\infty}^{t_1} dt_3 \, \xi_b(t_3) \, e^{\kappa t_3}$$

$$\times \int_{-\infty}^{\infty} dt_2 \, \xi_c^*(t_2) \, e^{-\kappa t_2} \int_{-\infty}^{t_2} dt_4 \, \xi_c(t_4) \, e^{\kappa t_4}, \tag{2.79}$$

and

$$T_2 \equiv \left(\frac{\kappa^2}{\pi^3} \right) \int dt_1 \, \xi_b^*(t_1) \int dt_2 \, \xi_c^*(t_2) \int dt_3 \, \xi_b(t_3)$$

$$\times \int dt_4 \, \xi_b(t_4) \int d\omega' \, \frac{e^{i\omega'(t_4 - t_1)}}{\kappa - i\omega'} \int d\omega'' \, \frac{e^{i\omega''(t_4 - t_2)}}{\kappa - i\omega''} \int d\omega_1 \, \frac{e^{i\omega_1(t_4 - t_2)}}{\kappa - i\omega_1}$$

$$= 8\kappa^2 \int_{-\infty}^{\infty} dt_4 \, \xi_c(t_4) \, e^{\kappa t_4} \int_{t_4}^{\infty} dt_1 \, \xi_b^*(t_1) \, e^{-\kappa t_1} \int_{t_4}^{\infty} dt_2 \, e^{-\kappa t_2}$$

$$\times \int_{-\infty}^{t_4} dt_3 \, \xi_b(t_3) \, e^{\kappa t_3}. \tag{2.80}$$

Next, on substituting the results from Eqs. (2.79) and (2.80) in Eq. (2.77) and furthermore, assuming that $\xi_b(t)$ and $\xi_c(t)$ are identical and real, we finally obtain, in the strong coupling limit, the following expression for fidelity, in terms of time-domain integrals:

$$\sqrt{F}e^{i\phi} = T_1 - T_2$$

$$= 1 - 4\kappa \int_{-\infty}^{\infty} dt\ \xi(t)\ e^{-\kappa t} \int_{-\infty}^{t} dt'\ \xi(t')\ e^{\kappa t'}$$

$$+ 4\kappa^2 \left[\int_{-\infty}^{\infty} dt\ \xi(t)\ e^{-\kappa t} \int_{-\infty}^{t} dt'\ \xi(t')\ e^{\kappa t'}\right]^2$$

$$- 8\kappa^2 \int_{-\infty}^{\infty} dt\ \xi(t)\ e^{\kappa t} \left[\int_{t}^{\infty} dt'\ \xi(t')\ e^{-\kappa t'}\right]^2$$

$$\times \int_{-\infty}^{t} dt''\ \xi(t'')\ e^{\kappa t''}. \tag{2.81}$$

Now we shall consider some specific examples to calculate the combined fidelity-phase. First, we shall consider the case in which the initial pulse is a Gaussian of duration T. As mentioned earlier, we shall assume that the initial state is factorizable, i.e. $\xi_{bc}(\omega', \omega'', 0) = \xi_b(\omega', 0)\ \xi_c(\omega'', 0)$, where ξ_b and ξ_c are both given by $\xi(\omega, 0) = \sqrt{T/\sqrt{\pi}}\ \exp[-\omega^2\ T^2/2]$. We shall use Eq. (2.75) to calculate the fidelity. On substituting the explicit form for a Gaussian wavepacket in Eq. (2.75), we get

$$\sqrt{F}e^{i\phi} = \frac{T^2}{\pi} \int d\omega' \int d\omega''\ \frac{(\kappa + i\omega')(\kappa + i\omega'')}{(\kappa - i\omega')(\kappa - i\omega'')}\ e^{-(\omega'^2 + \omega''^2)T^2} - \frac{4g^2\kappa^2}{\pi}\ \frac{T^2}{\pi}$$

$$\times \int d\omega' \int d\omega''\ e^{-(\omega'^2 + \omega''^2)T^2/2}\ \frac{1}{(\kappa - i\omega')(\kappa - i\omega'')}$$

$$\times \frac{1}{g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')]}$$

$$\times \underbrace{\int d\omega_1\ \frac{e^{-\omega_1^2 T^2/2}\ e^{-(\omega' + \omega'' - \omega_1)^2 T^2/2}}{(\kappa - i\omega_1)}}_{I_{II}}, \tag{2.82}$$

where

$$I_{II} = \pi \; e^{-T^2(\omega'+\omega'')^2/4} \; e^{T^2[\kappa - i(\omega'+\omega'')/2]^2} \; \mathrm{erfc}\left[\kappa T - iT\left(\frac{\omega'+\omega''}{2}\right)\right].$$

On explicitly substituting for I_{II} in Eq. (2.82), we obtain

$$\begin{aligned}
\sqrt{F}e^{i\phi} &= \frac{T^2}{\pi} \int d\omega' \int d\omega'' \; \frac{(\kappa + i\omega')(\kappa + i\omega'')}{(\kappa - i\omega')(\kappa - i\omega'')} \; e^{-(\omega'^2+\omega''^2)T^2} - \frac{4g^2\kappa^2 T^2}{\pi} \\
&\quad \times \int d\omega' \int d\omega'' \; \frac{e^{-(\omega'^2+\omega''^2)T^2/2}}{(\kappa - i\omega')(\kappa - i\omega'')} \; e^{-T^2(\omega'+\omega'')^2/4} \; e^{T^2[\kappa - i(\omega'+\omega'')/2]^2} \\
&\quad \times \frac{1}{g^2 + [2\kappa - i(\omega'+\omega'')][\kappa - i(\omega'+\omega'')]} \\
&\quad \times \mathrm{erfc}\left[\kappa T - iT\left(\frac{\omega'+\omega''}{2}\right)\right].
\end{aligned} \tag{2.83}$$

In order to save space and make the subsequent calculation easier to follow, we shall compute each term in Eq. (2.83) separately. We shall label the two terms in the previous equation T_I and T_{II} where,

$$T_I \equiv \frac{T^2}{\pi} \int d\omega' \int d\omega'' \; \frac{(\kappa + i\omega')(\kappa + i\omega'')}{(\kappa - i\omega')(\kappa - i\omega'')} \; e^{-(\omega'^2+\omega''^2)T^2},$$

and

$$\begin{aligned}
T_{II} &\equiv \frac{4g^2\kappa^2 T^2}{\pi} \int d\omega' \int d\omega'' \; \frac{e^{-(\omega'^2+\omega''^2)T^2/2}}{(\kappa - i\omega')(\kappa - i\omega'')} \; e^{-T^2(\omega'+\omega'')^2/4} \; e^{-T^2(\omega'+\omega'')^2/4} \; e^{T^2[\kappa - i(\omega'+\omega'')/2]^2} \\
&\quad \times \frac{1}{g^2 + [2\kappa - i(\omega'+\omega'')][\kappa - i(\omega'+\omega'')]} \; \mathrm{erfc}\left[\kappa T - iT\left(\frac{\omega'+\omega''}{2}\right)\right].
\end{aligned}$$

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First we shall work on T_I. Here, we shall once again make use of the identity: $(\kappa + i\omega)/(\kappa - i\omega) = -1 + (2\kappa/\kappa - i\omega)$ and expand the product, after which we shall make use of the following result:

$$\int_{-\infty}^{\infty} d\omega \; \frac{e^{-\omega^2 T^2}}{\kappa - i\omega} = \pi \; e^{\kappa^2 T^2} \; \text{erfc}(\kappa T),$$

to compute T_I. This procedure yields

$$T_I = \left[2\sqrt{\pi} \; \alpha \; e^{\alpha^2} \text{erfc}(\alpha) - 1\right]^2, \tag{2.84}$$

where $\alpha \equiv \kappa T$ is a dimensionless parameter. Next we shall calculate T_{II}. Here, we shall introduce two new variables η and Λ such that $\eta = (\omega' + \omega'')/2$ and $\Lambda = (\omega' - \omega'')/2$. In terms of these new variables, T_{II} can be rewritten as

$$T_{II} = \frac{8g^2 \kappa^2 T^2}{\pi} \int d\eta \; e^{-2\eta^2 T^2} \; \frac{e^{T^2(\kappa - i\eta)^2} \; \text{erfc}\left[T(\kappa - i\eta)\right]}{g^2 + 2(\kappa - i\eta)(\kappa - 2i\eta)}$$
$$\times \int d\Lambda \; \frac{e^{-\Lambda^2 T^2}}{[\kappa - i(\eta + \Lambda)][\kappa - i(\eta - \Lambda)]}.$$

We can now make a partial fraction decomposition of the denominator in the last integral (over Λ). This gives us

$$\frac{1}{[\kappa - i(\eta + \Lambda)][\kappa - i(\eta - \Lambda)]} = \frac{1}{2(\kappa - i\eta)} \left[\frac{1}{\kappa - i(\eta + \Lambda)} + \frac{1}{\kappa - i(\eta - \Lambda)}\right].$$

On explicitly substituting this decomposition in T_{II}, we get

$$T_{II} = \frac{8g^2\kappa^2 T^2}{\pi} \int d\eta \; \frac{e^{-2\eta^2 T^2}}{2(\kappa - i\eta)} \; \frac{e^{T^2(\kappa - i\eta)^2} \; \mathrm{erfc}\left[T(\kappa - i\eta)\right]}{g^2 + 2(\kappa - i\eta)(\kappa - 2i\eta)}$$

$$\times \left(\underbrace{\int d\Lambda \; \frac{e^{-\Lambda^2 T^2}}{\kappa - i(\eta + \Lambda)}}_{I_+} + \underbrace{\int d\Lambda \; \frac{e^{-\Lambda^2 T^2}}{\kappa - i(\eta - \Lambda)}}_{I_-} \right),$$

where

$$I_+ = I_- = \pi \; e^{T^2(\kappa - i\eta)^2} \; \mathrm{erfc}\left[T(\kappa - i\eta)\right].$$

On explicitly substituting for I_+ and I_- in T_{II}, we obtain

$$T_{II} = 8(gT)^2 \alpha^2 \int d\eta \; \frac{e^{-2\eta^2}}{\alpha - i\eta} \; \frac{e^{2(\alpha - i\eta)^2} \left[\mathrm{erfc}(\alpha - i\eta)\right]^2}{(gT)^2 + 2(\alpha - i\eta)(\alpha - 2i\eta)}, \tag{2.85}$$

where once again, $\alpha \equiv \kappa T$. Now on substituting Eqs. (2.84) and (2.85) in Eq. (2.83), we finally obtain the following explicit expression for the fidelity of a Gaussian wavepacket:

$$\sqrt{F} e^{i\phi} = \left[2\sqrt{\pi} \; \alpha \; e^{\alpha^2} \mathrm{erfc}(\alpha) - 1\right]^2 - 8 \; (gT)^2 \alpha^2$$

$$\times \int d\eta \; \frac{e^{-2\eta^2}}{\alpha - i\eta} \; \frac{e^{2(\alpha - i\eta)^2} \left[\mathrm{erfc}(\alpha - i\eta)\right]^2}{(gT)^2 + 2(\alpha - i\eta)(\alpha - 2i\eta)}. \tag{2.86}$$

In the strong-coupling limit, $(gT)^2 + 2(\alpha - i\eta)(\alpha - 2i\eta) \approx (gT)^2$. Thus, in this limit, the expression for the fidelity in Eq. (2.86) reduces to

$$\sqrt{F}e^{i\phi} = \left[2\sqrt{\pi}\;\alpha\;e^{\alpha^2}\mathrm{erfc}(\alpha) - 1\right]^2 - 8\;\alpha^2 \int d\eta\; \frac{e^{-2\eta^2}}{\alpha - i\eta}\; e^{2(\alpha-i\eta)^2}\left[\mathrm{erfc}(\alpha - i\eta)\right]^2. \qquad (2.87)$$
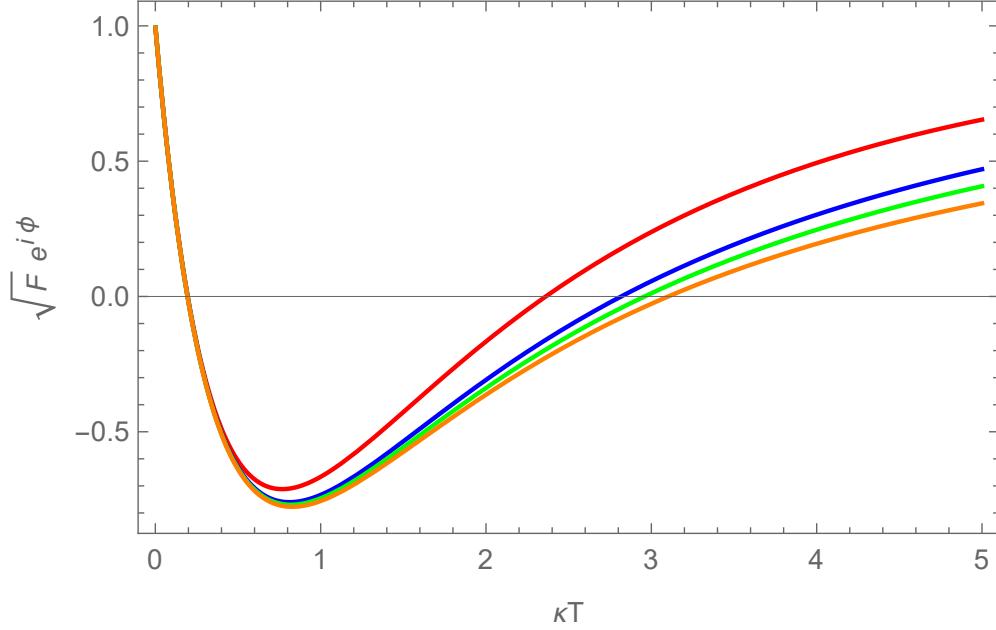
Figure 2.3: Square-root fidelity and phase for a Gaussian pulse, as a function of κT, for different values of the coupling strength g. From top to bottom: $gT = 7$; $gT = 14$; $gT = 21$; and the strong-coupling limit $(gT \to \infty)$, Eq. (2.89).

Figure (2.3) shows a plot of the square-root fidelity of a Gaussian wavepacket for different values of the coupling strength.

For a hyperbolic secant pulse of duration T, whose profile in the time domain is given by $\xi(t) = (1/\sqrt{2T})\,\mathrm{sech}(t/T)$, an explicit expression for the fidelity using Eq. (2.75) is extremely complicated to evaluate analytically. Instead we have only done this calculation numerically in the strong-coupling limit. Figure (2.4) shows this result.
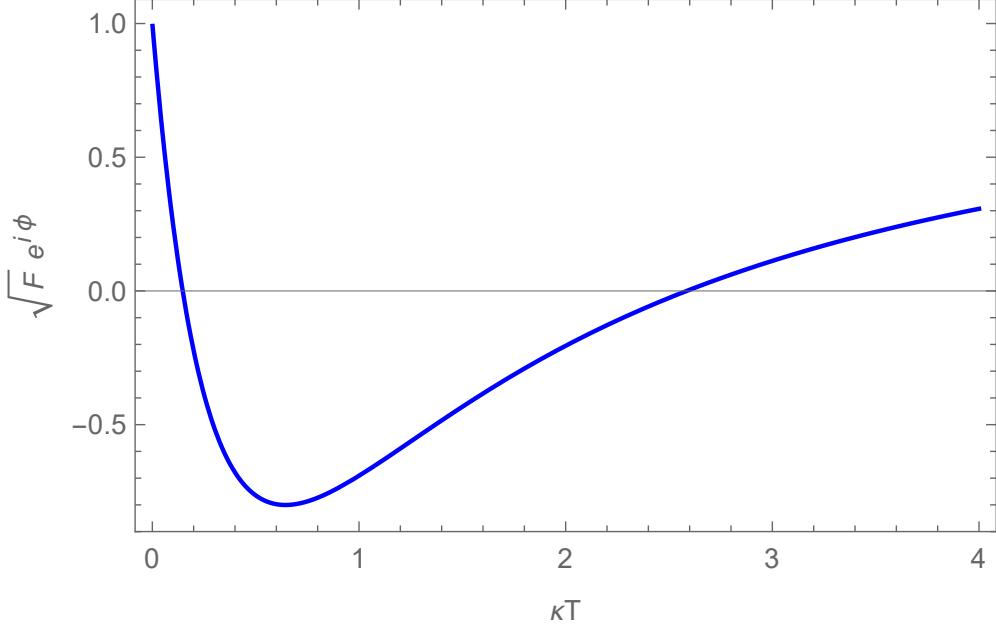
Figure 2.4: Square-root fidelity and phase for a hyperbolic secant pulse, as a function of κT, in the strong-coupling limit.

As these figures show, we obtain the optimal results when the bandwidth of the cavity and the pulse are more or less evenly matched, i.e. they are of the same order of magnitude ($\kappa T \sim 1$), similar to what we have seen in the free space scenario. However, we get a slightly higher fidelity (F) in the cavity configuration. For a moderately large coupling, $gT \sim 7$, we get $F \simeq 0.5$, while in the strong coupling limit, $F \simeq 0.6$.

In both the figures, (2.3) and (2.4), we see that as $\kappa T \to 0$, the fidelity (F) approaches 1. This is because in this limit, the pulse is simply reflected off of the mirror at the entrance of the cavity. On the other hand, we see that in the opposite limit, i.e. as $\kappa T \to \infty$, $F \to 1$. In this limit, the cavity empties itself very fast (over a time scale of the order of $1/\kappa \ll T$) as a result of which the probability for two photons to be present at the same time is negligible. This means that the two photons in all likelihood do not interact with the medium. Furthermore, when the bandwidth of the cavity is very large, the

spectral distortion of the wavepackets is negligible, which is why the fidelity again approaches 1 in this limit.

2.4 Kerr medium inside a one-sided cavity

The possibility of obtaining an analytical solution in a cavity configuration as we have seen in detail, in the previous section, motivated us to explore the case where we replace the $\chi^{(2)}$ medium in a one-sided cavity with a Kerr $[\chi^{(3)}]$ medium. We were driven by a curiosity to see if the results for this case would be any different from the second-order one.

For this problem, we only have two types of photons viz. a and b, in a general state of the form

$$|\psi(t)\rangle = \int d\omega' \int d\omega'' \ \xi_{ab}(\omega', \omega'', t) \ \hat{a}_{\omega'}^{\dagger} |0\rangle_a \ \hat{b}_{\omega''}^{\dagger} |0\rangle_b \tag{2.88}$$

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

$$\hat{H} = \hbar g \ [\hat{A}^{\dagger}(t)\hat{A}(t)\hat{B}^{\dagger}(t)\hat{B}(t)], \tag{2.89}$$

with the operators $\hat{A}^{\dagger}(t)$, $\hat{A}(t)$, $\hat{B}^{\dagger}(t)$ and $\hat{B}(t)$ given by Eq. (2.54).

On substituting Eqs. (2.88), (2.89) and (2.54) in the Schrödinger equation, we obtain under perfect resonance (i.e. $\Delta_a = \Delta_b = 0$), the following equation of motion:

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

$$\times \, \xi_{ab}(\omega_1, \omega_2, t), \qquad (2.90)$$

which can indeed be solved by the method of Laplace transform. The Laplace transform of Eq. (2.90) is given by

$$\tilde{\xi}_{ab}(\omega', \omega'', s) = \frac{1}{s} \, \xi_{ab}(\omega', \omega'', 0) - ig \left(\frac{\kappa}{\pi}\right)^2 \frac{1}{s} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')}$$

$$\times \int d\omega_1 \int d\omega_2 \, \frac{\tilde{\xi}_{ab}[\omega_1, \omega_2, s + i(\omega_1 + \omega_2 - \omega' - \omega'')]}{(\kappa - i\omega_1)(\kappa - i\omega_2)}. \qquad (2.91)$$

The recipe to proceed further is the same as what we had done in the previous section.

We shall define

$$\frac{1}{s} \, \xi_{ab}(\omega', \omega'', 0) \equiv F(\omega', \omega'', s),$$

following which, we shall shift to dummy arguments in Eq. (2.91) viz. $\omega' \to \omega_3$, $\omega'' \to \omega_4$ and $s \to s + i(\omega_3 + \omega_4 - \omega' - \omega'')$. This procedure yields

$$\tilde{\xi}_{ab}[\omega_3, \omega_4, s + i(\omega_3 + \omega_4 - \omega' - \omega'')] = F[\omega_3, \omega_4, s + i(\omega_3 + \omega_4 - \omega' - \omega'')]$$

$$- \, ig \left(\frac{\kappa}{\pi}\right)^2 \frac{1}{s + i(\omega_3 + \omega_4 - \omega' - \omega'')} \frac{1}{(\kappa + i\omega_3)(\kappa + i\omega_4)}$$

$$\times \int d\omega_1 \int d\omega_2 \, \frac{\tilde{\xi}_{ab}[\omega_1, \omega_2, s + i(\omega_1 + \omega_2 - \omega' - \omega'')]}{(\kappa - i\omega_1)(\kappa - i\omega_2)}. \qquad (2.92)$$

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Next, we shall divide both sides of the previous equation by $(\kappa - i\omega_3)(\kappa - i\omega_4)$ and integrate over ω_3 and ω_4, which then gives us

$$
\begin{aligned}
I' \equiv \int d\omega_3 \int d\omega_4 \; & \frac{\tilde{\xi}_{ab}[\omega_3, \omega_4, s + i(\omega_3 + \omega_4 - \omega' - \omega'')]}{(\kappa - i\omega_3)(\kappa - i\omega_4)} \\
= & \int d\omega_3 \int d\omega_4 \; \frac{F[\omega_3, \omega_4, s + i(\omega_3 + \omega_4 - \omega' - \omega'')]}{(\kappa - i\omega_3)(\kappa - i\omega_4)} \\
& - ig \left(\frac{\kappa}{\pi}\right)^2 \underbrace{\int d\omega_3 \int d\omega_4 \; \frac{1}{s + i(\omega_3 + \omega_4 - \omega' - \omega'')} \frac{1}{(\kappa^2 + \omega_3^2)(\kappa^2 + \omega_4^2)}}_{I''} \\
& \times \underbrace{\int d\omega_1 \int d\omega_2 \; \frac{\tilde{\xi}_{ab}[\omega_1, \omega_2, s + i(\omega_1 + \omega_2 - \omega' - \omega'')]}{(\kappa - i\omega_1)(\kappa - i\omega_2)}}_{I'},
\end{aligned}
\tag{2.93}
$$

where

$$
I'' = \left(\frac{\pi}{\kappa}\right)^2 \frac{1}{s + 2\kappa - i(\omega' + \omega'')}.
$$

On explicitly substituting for I'' in Eq. (2.93), we get

$$
I' = \int d\omega_3 \int d\omega_4 \; \frac{F[\omega_3, \omega_4, s + i(\omega_3 + \omega_4 - \omega' - \omega'')]}{(\kappa - i\omega_3)(\kappa - i\omega_4)} - \left(\frac{ig}{s + 2\kappa - i(\omega' + \omega'')}\right) I', \tag{2.94}
$$

from which it is straightforward to obtain an expression for I'. In the previous equation, we just need to move the term involving I' on the right hand side to the left following which we can express I' in terms of F.

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Thus, we obtain

$$
\begin{aligned}
I' &\equiv \int d\omega_1 \int d\omega_2 \, \frac{\tilde{\xi}_{ab}[\omega_1, \omega_2, s + i(\omega_1 + \omega_2 - \omega' - \omega'')]}{(\kappa - i\omega_1)(\kappa - i\omega_2)} \\
&= \left(1 + \frac{ig}{s + 2\kappa - i(\omega' + \omega'')}\right)^{-1} \int d\omega_3 \int d\omega_4 \, \frac{F[\omega_3, \omega_4, s + i(\omega_3 + \omega_4 - \omega' - \omega'')]}{(\kappa - i\omega_3)(\kappa - i\omega_4)}.
\end{aligned}
\tag{2.95}
$$

On substituting Eq. (2.95) in Eq. (2.91), we obtain

$$
\begin{aligned}
\tilde{\xi}_{ab}(\omega', \omega'', s) &= F(\omega', \omega'', s) - ig \left(\frac{\kappa}{\pi}\right)^2 \frac{1}{s} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')} \left(1 + \frac{ig}{s + 2\kappa - i(\omega' + \omega'')}\right)^{-1} \\
&\quad \times \int d\omega_3 \int d\omega_4 \, \frac{F[\omega_3, \omega_4, s + i(\omega_3 + \omega_4 - \omega' - \omega'')]}{(\kappa - i\omega_3)(\kappa - i\omega_4)}
\end{aligned}
$$

$$
\tag{2.96}
$$

In the definition of F [see below Eq. (2.91)], on shifting $\omega' \to \omega_3$, $\omega'' \to \omega_4$ and

$s \to s + i(\omega_3 + \omega_4 - \omega' - \omega'')$, we get the following explicit form for

$F[\omega_3, \omega_4, s + i(\omega_3 + \omega_4 - \omega' - \omega'')]$:

$$
F[\omega_3, \omega_4, s + i(\omega_3 + \omega_4 - \omega' - \omega'')] = \frac{\xi_{ab}(\omega_3, \omega_4, 0)}{s + i(\omega_3 + \omega_4 - \omega' - \omega'')}.
\tag{2.97}
$$

Now, on directly substituting Eq. (2.97) in Eq. (2.96) and expressing $F(\omega', \omega'', s)$ in

terms of $\xi_{ab}(\omega', \omega'', 0)$ from the definition of F, we obtain

$$\tilde{\xi}_{ab}(\omega', \omega'', s) = \frac{1}{s} \, \xi_{ab}(\omega', \omega'', 0) - ig \left(\frac{\kappa}{\pi}\right)^2 \frac{1}{s} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')}$$

$$\times \left(1 + \frac{ig}{s + 2\kappa - i(\omega' + \omega'')}\right)^{-1}$$

$$\times \int d\omega_3 \int d\omega_4 \frac{\xi_{ab}(\omega_3, \omega_4, 0)}{[s + i(\omega_3 + \omega_4 - \omega' - \omega'')](\kappa - i\omega_3)(\kappa - i\omega_4)}. \qquad (2.98)$$

Eq.(2.98) is the formal solution to our problem in the s-domain. We can clearly see that the full inversion of this equation is extremely hard. So once again, we shall only compute the asymptotic final state by using the final value theorem of operational calculus:

$\lim_{t\to\infty} \xi_{bc}(\omega', \omega'', t) = \lim_{s\to 0} s \, \tilde{\xi}_{bc}(\omega', \omega'', s)$.

This yields

$$\lim_{s\to 0} \left[s \, \tilde{\xi}_{ab}(\omega', \omega'', s)\right] = \xi_{ab}(\omega', \omega'', 0) - ig \left(\frac{\kappa}{\pi}\right)^2 \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')}$$

$$\times \left(\frac{2\kappa - i(\omega' + \omega'')}{ig + 2\kappa - i(\omega' + \omega'')}\right)$$

$$\times \underbrace{\lim_{s\to 0} \int d\omega_3 \int d\omega_4 \frac{\xi_{ab}(\omega_3, \omega_4, 0)}{[s + i(\omega_3 + \omega_4 - \omega' - \omega'')](\kappa - i\omega_3)(\kappa - i\omega_4)}}_{I_f}. \qquad (2.99)$$

The integral I_f in Eq. (2.99) is identical to the integral I_2 in Eq.(2.70) which can then be evaluated to give us the following result [see the discussion below Eq. (2.72) for justification]:

$$I_f = 2\pi \int d\omega_3 \, \frac{\xi_{ab}(\omega_3, \omega' + \omega'' - \omega_3, 0)}{(\kappa - i\omega_3)[\kappa - i(\omega' + \omega'' - \omega_3)]}.$$

On explicitly substituting the result for I_f in Eq. (2.99), we get

$$\lim_{s \to 0} \left[s \, \tilde{\xi}_{ab}(\omega', \omega'', s) \right] = \xi_{ab}(\omega', \omega'', 0) - \frac{2ig\kappa^2}{\pi} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')}$$
$$\times \left(\frac{2\kappa - i(\omega' + \omega'')}{ig + 2\kappa - i(\omega' + \omega'')} \right) \int d\omega \, \frac{\xi_{ab}(\omega, \omega' + \omega'' - \omega, 0)}{(\kappa - i\omega)[\kappa - i(\omega' + \omega'' - \omega)]}, \qquad (2.100)$$

where in the last integral, the variable of integration is replaced by ω. The integral in Eq. (2.100) is identical to the integral in Eq. (2.71). So, the partial fraction decomposition in Eq. (2.72) can be used here to simplify the result. Thus, substituting Eq. (2.72) in Eq. (2.100) leaves us with

$$\lim_{s \to 0} \left[s \, \tilde{\xi}_{ab}(\omega', \omega'', s) \right] = \xi_{ab}(\omega', \omega'', 0) - \frac{2ig\kappa^2}{\pi} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')[ig + 2\kappa - i(\omega' + \omega'')]}$$
$$\times \left[\int d\omega \, \frac{\xi_{ab}(\omega, \omega' + \omega'' - \omega, 0)}{\kappa - i\omega} + \int d\omega \, \frac{\xi_{ab}(\omega, \omega' + \omega'' - \omega, 0)}{\kappa - i(\omega' + \omega'' - \omega)} \right].$$

$$(2.101)$$

In the previous equation, if we set $\omega' + \omega'' - \omega \equiv \omega_1$ in the second integral, it becomes identical to the first integral, which then simplifies Eq. (2.101) to

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$$\lim_{s \to 0} \left[s \, \tilde{\xi}_{ab}(\omega', \omega'', s) \right] = \xi_{ab}(\omega', \omega'', 0) - \frac{4ig\kappa^2}{\pi} \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')[ig + 2\kappa - i(\omega' + \omega'')]}$$
$$\times \int d\omega \, \frac{\xi_{ab}(\omega, \omega' + \omega'' - \omega, 0)}{\kappa - i\omega}.$$

$$(2.102)$$

Next, we must multiply Eq. (2.102) by the "empty cavity" factors $(\kappa + i\omega')/(\kappa - i\omega')$

and $(\kappa + i\omega'')/(\kappa - i\omega'')$ to get the outgoing field, following which we can compute the

fidelity that is given by

$$\sqrt{F} e^{i\phi} = \int d\omega' \int d\omega'' \left(\frac{\kappa + i\omega'}{\kappa - i\omega'} \right) \left(\frac{\kappa + i\omega''}{\kappa - i\omega''} \right) \xi_{ab}^*(\omega', \omega'', 0) \lim_{s \to 0} \left[s \, \tilde{\xi}_{ab}(\omega', \omega'', s) \right]$$
$$= \int d\omega' \int d\omega'' \left(\frac{\kappa + i\omega'}{\kappa - i\omega'} \right) \left(\frac{\kappa + i\omega''}{\kappa - i\omega''} \right) |\xi_{ab}(\omega', \omega'', 0)|^2$$
$$- \frac{4ig\kappa^2}{\pi} \int d\omega' \int d\omega'' \frac{1}{(\kappa - i\omega')(\kappa - i\omega'')} \frac{\xi_{ab}^*(\omega', \omega'', 0)}{ig + [2\kappa - i(\omega' + \omega'')]}$$
$$\times \int d\omega \, \frac{\xi_{ab}(\omega, \omega' + \omega'' - \omega, 0)}{\kappa - i\omega}.$$

$$(2.103)$$

From the expression in Eq. (2.103), it is clear that the Kerr medium placed inside an

optical cavity is not likely to give us any better result than the second-order case. In the

strong-coupling limit, i.e. $ig + [2\kappa - i(\omega' + \omega'')] \simeq ig$, Eq. (2.103) reduces to

$$\sqrt{F} e^{i\phi} = \int d\omega' \int d\omega'' \left(\frac{\kappa + i\omega'}{\kappa - i\omega'} \right) \left(\frac{\kappa + i\omega''}{\kappa - i\omega''} \right) |\xi_{ab}(\omega', \omega'', 0)|^2$$
$$- \frac{4\kappa^2}{\pi} \int d\omega' \int d\omega'' \frac{\xi_{ab}^*(\omega', \omega'', 0)}{(\kappa - i\omega')(\kappa - i\omega'')} \int d\omega \, \frac{\xi_{ab}(\omega, \omega' + \omega'' - \omega, 0)}{\kappa - i\omega}, \quad (2.104)$$

which is identical to Eq. (2.76). Interestingly, we find that in the strong-coupling limit which is the most favorable case for the cavity configuration, the second- and the third-order nonlinearities are completely equivalent.

2.5 Drawbacks of an optical cavity

The major shortcoming of using an optical cavity for single-photon quantum logic is that even an empty cavity will, in general, considerably distort an incident pulse. Hence, we should essentially expect low fidelities for such cavity systems. However, the results shown in figures (2.3) and (2.4) for the two-photon gate may be called surprisingly high.

In order to appreciate this point better, we shall consider the situation when only one photon is incident on the cavity. This would correspond to either one of the states $|01\rangle$ or $|10\rangle$ in Eq. (2.1), which on a random basis, may be expected to happen half the time. It is important to note that when we do quantum logic, the initial state has be considered unknown by definition. The goal in quantum logic, in the most ideal case, is to realize the transformation in Eq. (2.1) with unit fidelity for all the four input states. This is the reason why we resort to nonlinear optical schemes to build a phase gate.

In the case of a single-photon incident on the cavity, the spectrum of the outgoing field is

$$\tilde{f}_{out}(\omega) = \frac{\kappa + i\omega}{\kappa - i\omega} \; \tilde{f}_{in}(\omega), \tag{2.105}$$

where $\tilde{f}_{in}(\omega)$ could stand for either $\xi_b(\omega, 0)$ or $\xi_c(\omega, 0)$, and the overlap with the initial state is simply given by

$$\sqrt{F}e^{i\phi} = \int_{-\infty}^{\infty} d\omega \; \tilde{f}_{in}^*(\omega) \; \tilde{f}_{out}(\omega)$$

$$= \int_{-\infty}^{\infty} d\omega \; \left(\frac{\kappa + i\omega}{\kappa - i\omega}\right) \; |\tilde{f}_{in}(\omega)|^2. \tag{2.106}$$

For a Gaussian pulse $\tilde{f}_{in}(\omega) = \sqrt{T/\sqrt{\pi}} \exp(-\omega^2 T^2/2)$, the square-root fidelity given by Eq. (2.106) can be easily computed as shown below.

$$\sqrt{F}e^{i\phi} = \frac{T}{\sqrt{\pi}} \int_{-\infty}^{\infty} d\omega \; \left(-1 + \frac{2\kappa}{\kappa - i\omega}\right) \; e^{-\omega^2 T^2}$$

$$= -\frac{T}{\sqrt{\pi}} \int_{-\infty}^{\infty} d\omega \; e^{-\omega^2 T^2} + \frac{2\kappa T}{\sqrt{\pi}} \int_{-\infty}^{\infty} d\omega \; \frac{e^{-\omega^2 T^2}}{\kappa - i\omega}$$

$$= -1 + 2\sqrt{\pi}\kappa T \; e^{(\kappa T)^2} \; \text{erfc}(\kappa T), \tag{2.107}$$

which makes

$$F = \left[2\sqrt{\pi}\kappa T \; e^{(\kappa T)^2} \; \text{erfc}(\kappa T) - 1\right]^2.$$

We can see that F is equal to 1 in the limits $\kappa T = 0$ and $\kappa T \to \infty$. In particular, around $\kappa T = 0.8$, where the two-photon gate performs best, the single-photon fidelity is very low, i.e. $F \simeq 0.15$.

The two limits where the cavity does not distort the incident pulse are, as we have just seen, the small bandwidth limit $\kappa T \to 0$ (where the pulse is simply reflected off of the entrance mirror) and the adiabatic limit $\kappa T \to \infty$. Most of the feasible cavity based schemes for single-photon quantum logic operate in the adiabatic limit, such as the

Duan-Kimble gate [17], or the Koshino-Ishizaka-Nakamura gate [18]. However, in these two limits, as figures (2.3) and (2.4) reveal, the gate operation involving two photons incident on the cavity fails to produce the desired phase shift for the state $|1, 1\rangle$.

Finally, we shall discuss a special case where the distortion of the incoming pulse by an empty cavity is, in principle, reversible. This happens when the pulse has the form of a rising exponential with a time constant κ. This is in fact, the time reversal of the pulse leaking out of the cavity, at least in the absence of a nonlinear medium inside. The use of time-reversed pulses for the transmission of quantum information between two optical cavities was first proposed by Cirac *et al.* in [19]. However, it is essential to note that this is a very different scheme, and in a very distinct context from what we have considered here. It is important to keep in mind that in quantum computation, the two qubits must be identical. So in this case, either we must choose a rising exponential or a decaying exponential for the qubits. If we settle on, say the rising exponential as our default pulse, then the time reversal operation has to be arranged to happen automatically for all the four states in Eq. (2.1) since the initial state is unknown by definition when we do quantum logic. The time reversal operation is accomplished in practice through optical phase conjugation, which is a nonlinear process in itself, and which, as far as we know, has not yet been demonstrated for single-photo pulses [20]. In our theoretical analysis, phase conjugation is formally just the complex conjugation of the spectrum.

If we send in a single-photon as a rising exponential pulse with the spectrum

$$\tilde{f}_{in}(\omega) = \sqrt{\frac{\kappa}{\pi}} \frac{1}{(\kappa + i\omega)},$$

then from Eq. (2.105), the pulse coming out of the cavity after phase conjugation is

$$\tilde{f}_{out}(\omega) = \sqrt{\frac{\kappa}{\pi}} \left(\frac{\kappa + i\omega}{\kappa - i\omega} \times \frac{1}{\kappa + i\omega} \right)^* = \sqrt{\frac{\kappa}{\pi}} \frac{1}{(\kappa + i\omega)}.$$

From here, the calculation of the fidelity is straightforward.

$$\sqrt{F}e^{i\phi} = \int_{-\infty}^{\infty} d\omega \ \tilde{f}_{in}^*(\omega) \ \tilde{f}_{out}(\omega)$$

$$= \frac{\kappa}{\pi} \underbrace{\int_{-\infty}^{\infty} d\omega \ \frac{1}{\kappa^2 + \omega^2}}_{\pi/\kappa},$$

$$= 1. \hspace{6cm} (2.108)$$

We see that for the single-photon case, the fidelity is of course, 1. Next we shall see what happens if we send in two photons as rising exponential pulses with the spectrum of the form

$$\xi_{bc}(\omega', \omega'', 0) = \left(\frac{\kappa}{\pi} \right) \frac{1}{(\kappa + i\omega')(\kappa + i\omega'')}.$$

We shall compute the outgoing field for the two-photon case in the strong-coupling limit since this turned out the be the most favorable situation in the cavity configuration. We can write down the general expression for the outgoing spectrum in the asymptotic limit by reducing Eq. (2.74) to the case in which the nonlinear coupling strength is strong (i.e. $g^2 + [2\kappa - i(\omega' + \omega'')][\kappa - i(\omega' + \omega'')] \simeq g^2$) and multiplying the resulting expression by the "empty cavity" factors $(\kappa + i\omega')/(\kappa - i\omega')$ and $(\kappa + i\omega'')/(\kappa - i\omega'')$. This yields

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$$\tilde{\xi}_{out}(\omega',\omega'',t\to\infty) = \left(\frac{\kappa+i\omega'}{\kappa-i\omega'}\right)\left(\frac{\kappa+i\omega''}{\kappa-i\omega''}\right)\xi_{bc}(\omega',\omega'',0)$$
$$-\frac{4\kappa^2}{\pi}\frac{1}{(\kappa-i\omega')(\kappa-i\omega'')}\int d\omega_1 \frac{\xi_{bc}(\omega_1,\omega'+\omega''-\omega_1,0)}{\kappa-i\omega_1}. \qquad (2.109)$$

On explicitly substituting for the rising exponential in Eq. (2.109), we get

$$\tilde{\xi}_{out}(\omega',\omega'',t\to\infty) = \left(\frac{\kappa}{\pi}\right)\frac{1}{(\kappa-i\omega')(\kappa-i\omega'')} - \frac{4\kappa^3}{\pi^2}\frac{1}{(\kappa-i\omega')(\kappa-i\omega'')}$$
$$\times \underbrace{\int d\omega_1 \frac{1}{(\kappa^2+\omega_1^2)}\frac{1}{\kappa+i(\omega'+\omega''-\omega_1)}}_{I}, \qquad (2.110)$$

where

$$I = \frac{\pi}{\kappa}\frac{1}{2\kappa+i(\omega'+\omega'')}.$$

On explicitly substituting for I in Eq. (2.110), we obtain

$$\tilde{\xi}_{out} = \left(\frac{\kappa}{\pi}\right)\frac{1}{(\kappa-i\omega')(\kappa-i\omega'')} - \frac{4\kappa^2}{\pi}\frac{1}{(\kappa-i\omega')(\kappa-i\omega'')}\frac{1}{2\kappa+i(\omega'+\omega'')}. \qquad (2.111)$$

Next, we shall perform the phase conjugation on $\tilde{\xi}_{out}$ which then gives us

$$\tilde{\xi}_{out}^* = \left(\frac{\kappa}{\pi}\right)\frac{1}{(\kappa+i\omega')(\kappa+i\omega'')} - \frac{4\kappa^2}{\pi}\frac{1}{(\kappa+i\omega')(\kappa+i\omega'')}\frac{1}{2\kappa-i(\omega'+\omega'')}. \qquad (2.112)$$

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Finally, we shall compute the fidelity for this case which is given by

$$
\sqrt{F}\,e^{i\phi} = \int d\omega' \int d\omega'' \; \xi_{in}^*(\omega',\omega'',0) \; \xi_{out}^*(\omega',\omega'',t\to\infty)
$$

$$
= \left(\frac{\kappa}{\pi}\right)^2 \underbrace{\int d\omega' \; \frac{1}{\kappa^2+\omega'^2}}_{\pi/\kappa} \; \underbrace{\int d\omega'' \; \frac{1}{\kappa^2+\omega''^2}}_{\pi/\kappa}
$$

$$
-\frac{4\kappa^3}{\pi^2} \underbrace{\int d\omega' \int d\omega'' \; \frac{1}{(\kappa^2+\omega'^2)(\kappa^2+\omega''^2)} \; \frac{1}{2\kappa - i(\omega'+\omega'')}}_{\pi^2/4\kappa^3},
$$

$$
= 1 - 1 = 0. \tag{2.113}
$$

We can clearly see that in the case of two incoming photons, in the strong-coupling limit, after the interaction with the nonlinear medium and phase conjugation, the fidelity turns out be exactly zero. So these pulses would not be useful at all for quantum logic even in the most ideal case.

2.6   Conclusion

We have carried out a thorough multimode quantized field analysis of the proposal to use a second-order optical nonlinearity to perform a conditional phase shift between two co-propagating single-photon pulses traveling with equal speeds. Our study has revealed that in the "free-space scenario" where the pulses travel through a nonlinear medium with a finite transmission bandwidth, the maximum fidelity that we could achieve is less than 0.4. Following this, we have extended our analysis to a situation in which the nonlinear medium is placed inside a one-sided cavity and found that here fidelities as large as 0.6 are

theoretically possible. In both these cases, we obtained optimal results when the medium bandwidth and the pulse bandwidth are more or less evenly matched in the frequency space.

In both, the "free-space" and the cavity configuration, we conclude based on our analysis that the spectral entanglement of the final state is indeed an important fidelity degrading mechanism. In other words, once the two incident photons are destroyed in the medium, the two "re-created" photons are constrained only by the energy and momentum conservation (which is the same for both of them here since the pulses travel with the same speed) and the spectral properties of the medium, and indeed they need not resemble the initial state very much. We have seen that the final state is in general, spectrally entangled in momentum even though we assumed the initial state to be factorizable.

Perhaps most intriguingly, we have found that if the second-order nonlinear medium is replaced by a third-order medium in the cavity, the fidelity in the strong-coupling limit is given by the same mathematical expression.

We thus, conclude that for schemes involving two co-propagating single-photon pulses with equal velocities, the second-order nonlinearities suffer from the same limitations as the third-order ones and do not have any apparent advantage over "Kerr" media for conditional single-photon quantum logic [21].

Chapter 3

Conditional phase shift in a nonlocal nonlinear medium

3.1 Introduction

In the previous chapter, we carried out a multimode quantized field analysis of a conditional phase gate based on $\chi^{(2)}$ nonlinearity using two co-propagating single photon wavepackets traveling with equal speeds, both in free space and a one-sided optical cavity. We concluded that spectral entanglement of the final state is an important fidelity degrading mechanism, and this scheme suffers from the same limitations as the third-order ones. The conventional wisdom until now which is the culmination of all these studies is that it is impossible to achieve unit fidelity with a π phase shift. This was indeed the original claim made by Shapiro [5] and further strengthened by Gea-Banacloche [12] and a few others [22, 23, 24]. Nevertheless, in recent times there have been a number of claims that appear to challenge this view [25, 26, 27, 28, 29, 30] and present a strong case for high fidelity conditional phase shift. The other reassuring theoretical results have been presented in [31, 32]. In particular, we were strongly motivated by couple of theoretical papers viz. one by Xia *et al.* [28] and the other one by Brod *et al.* [30], which showed that it is indeed possible to achieve unit fidelity with a π phase shift.

In this chapter, we shall develop an analytical model for the system studied numerically by Xia *et al.* Our problem here is to study the conditional phase shift between two co-propagating single-photon wavepackets traveling with different speeds in a $\chi^{(2)}$ medium. When the b and the c photons travel with different speeds, it means that they are no longer identical as would be required for qubits in quantum computation. One possible way to

circumvent this difficulty, in principle, is to use different polarizations for the two photons in a birefringent medium, assuming that the qubit is not encoded in the polarization state. Following Xia *et al.*, we treat the nonlinear medium as being spatially nonlocal, as a result of which we can derive the Hamiltonian without the need for any noise operators. Assuming three modes a, b and c, we envisage a region of interaction that consists of a $\chi^{(2)}$ medium upon which the b and the c photons are incident. The pulses travel with different speeds, so they pass through each other. As they do, the b and the c photons annihilate to create an a photon. Still later, the a photon annihilates to create a new $b - c$ pair. This scheme in a simplified, single-mode picture could be described by the Hamiltonian $\hat{H} = \hbar\epsilon(\hat{a}^\dagger \hat{b} \hat{c} + \hat{a} \hat{b}^\dagger \hat{c}^\dagger)$, where ϵ is the strength of nonlinear coupling. We consider here the most general (still one dimensional) multimode version of this problem and generalize the results of Xia *et al.* to deal with an arbitrary response function, initial state and pulse velocity. In our model, we assume that the width of the pulse (corresponding to the a, b and c photons) remains constant during the entire process.

3.2 Analytical model for a spatially nonlocal $\chi^{(2)}$ medium

The system that we wish to study is described by the following Hamiltonian in the continuous mode representation:

$$\hat{H} = \hat{H}_0 + \hat{H}_{int},$$

$$\hat{H}_0 = \hbar v_a \int dk \, k \, \hat{a}_k^\dagger \, \hat{a}_k + \hbar v_b \int dk \, k \, \hat{b}_k^\dagger \, \hat{b}_k + \hbar v_c \int dk \, k \, \hat{c}_k^\dagger \, \hat{c}_k \qquad (3.1)$$

$$\hat{H}_{int} = \hbar\epsilon \int_0^L dz_a \int_0^L dz_b \int_0^L dz_c \, f(z_a, z_b, z_c) \, \hat{A}^\dagger(z_a) \, \hat{B}(z_b) \, \hat{C}(z_c) + H.c,$$

where $\hat{H}_0$ is the Hamiltonian of the free field, $\hat{H}_{int}$ is the Hamiltonian corresponding to the interaction with the nonlinear $(\chi^{(2)})$ medium and $H.c$ stands for hermitian conjugate. In our problem, we consider the most general case, in which the three photons, a, b and c, travel with different speeds viz. v_a, v_b and v_c, respectively. The length of the region of interaction is taken to be L, however, this will not figure in our calculation because we will let one pulse sweep across the other and assume that the interaction starts well after both the photons enter the medium and ends well before they leave. The function $f(z_a, z_b, z_c)$ describes the nonlocal response of the nonlinear medium. We shall assume the response function to have the following general form

$$f(z_a, z_b, z_c) = h(z_a - z_b) \ h(z_a - z_c), \tag{3.2}$$

where $h(z)$ is an appropriate real function. Physically, the two photons b and c don't have to annihilate at the same location in the region of interaction. They can be at different locations within a characteristic length which we will call the length scale of medium nonlocality. In other words, the b and the c photons have to be present in a region of space inside the nonlinear medium characterized by this length scale of medium nonlocality, for them to interact, i.e. to undergo parametric up- and down-conversion processes. Otherwise, these two photons will never interact. So they cannot be at arbitrary locations inside the medium. Similarly, the a photon can be created anywhere within this length scale and later when it annihilates to create a new b, c pair, these two photons can be created anywhere within this characteristic length. Mathematically speaking, in our analysis, we use the coordinate of the center of the wavepacket of the a photon, z_a, as a

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reference to connect the centers of the wavepackets of the b and the c photons, i.e. z_b and z_c, respectively, to incorporate this physical requirement. This is the reason for assuming the form in Eq. (3.2) for the response function of the medium.

The operators $\hat{A}(z_a)$, $\hat{B}(z_b)$ and $\hat{C}(z_c)$ are defined as

$$
\hat{A}(z_a) = \frac{1}{\sqrt{2\pi}} \int dk \; e^{ikz_a} \; \hat{a}_k,
$$

$$
\hat{B}(z_b) = \frac{1}{\sqrt{2\pi}} \int dk \; e^{ikz_b} \; \hat{b}_k,
$$

$$
\hat{C}(z_c) = \frac{1}{\sqrt{2\pi}} \int dk \; e^{ikz_c} \; \hat{c}_k, \tag{3.3}
$$

and they satisfy the canonical commutation relations $[\hat{A}(z), \hat{A}^\dagger(z')] = [\hat{B}(z), \hat{B}^\dagger(z')] = [\hat{C}(z), \hat{C}^\dagger(z')] = \delta(z - z')$.

On explicitly substituting for operators A, B and C from Eq.(3.3) in Eq.(3.1), we get the following expression for $\hat{H}_{int}$:

$$
\hat{H}_{int} = \frac{\hbar\epsilon}{(2\pi)^{3/2}} \int_0^L dz_a \int_0^L dz_b \int_0^L dz_c \; f(z_a, z_b, z_c) \int dk_a \; e^{-ik_a}
$$

$$
\times \int dk_b \; e^{ik_b} \int dk_c \; e^{ik_c} \; \hat{a}_{k_a}^\dagger \; \hat{b}_{k_b} \; \hat{c}_{k_c} + H.c. \tag{3.4}
$$

We shall work out this problem in the Schrödinger picture and in the momentum representation, where the most general state of the field is written as

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

$$(3.5)$$

The equations of motion for the a and the $b - c$ pulses can be derived from the

Schrödinger equation: $|\dot{\psi}\rangle = -(i/\hbar)\hat{H}|\psi\rangle$, using Eqs.(3.1), (3.4) and (3.5). On doing so, we

get

$$\left(\frac{\partial}{\partial t} + i \, v_a \, k_a\right) \xi_a(k_a, t) = -\frac{i\epsilon}{(2\pi)^{3/2}} \int dz_a \, e^{-ik_a z_a} \int dz_b \int dz_c \, f(z_a, z_b, z_c)$$

$$\times \int dk_b \, e^{ik_b z_b} \int dk_c \, e^{ik_c z_c} \, \xi_{bc}(k_b, k_c, t),$$

$$(3.6)$$

$$\left(\frac{\partial}{\partial t} + i \, v_b \, k_b + i \, v_c \, k_c\right) \xi_{bc}(k_b, k_c, t) = -\frac{i\epsilon}{(2\pi)^{3/2}} \int dz_a \int dz_b \int dz_c \, f(z_a, z_b, z_c)$$

$$\times \, e^{-ik_b z_b} \, e^{-ik_c z_c} \int dk_a \, e^{ik_a z_a} \, \xi_a(k_a, t).$$

$$(3.7)$$

Our next step is to insert the response function from Eq. (3.2) in Eqs. (3.6) and (3.7) to

get the exact equations of motion for our problem. In all the subsequent steps, we shall let

the limits of the integrals over z_a, z_b and z_c to extend from $-\infty$ to ∞ because as we

discussed earlier, in this scheme, we let the pulses pass through each other and assume that

the interaction starts well after both of them enter the medium and ends well before they

leave. So, we can harmlessly extend the limits of these integrals (over spatial coordinates)

to infinity without affecting the result. We shall first work on the equation of motion for

the a pulse. On substituting the response function in Eq. (3.6), we get

$$
\left(\frac{\partial}{\partial t} + i \, v_a \, k_a\right) \xi_a(k_a, t) = -\frac{i\epsilon}{(2\pi)^{3/2}} \int dk_b \int dk_c \, \xi_{bc}(k_b, k_c, t) \int dz_a \, e^{-ik_a z_a}
$$

$$
\times \underbrace{\int dz_b \, e^{ik_b z_b} \, h(z_a - z_b)}_{I_I} \underbrace{\int dz_c \, e^{ik_c z_c} \, h(z_a - z_c)}_{I_{II}} . \tag{3.8}
$$

Next, we have to evaluate the two integrals viz. I_I and I_{II}. To begin with, let us set $z_a - z_b = x$ in I_I. In terms of x, we can rewrite I_I as

$$
I_I = \sqrt{2\pi} \, e^{ik_b z_a} \underbrace{\frac{1}{\sqrt{2\pi}} \int dx \, e^{-ik_b x} \, h(x)}_{\tilde{h}(k_b)},
$$

$$
= \sqrt{2\pi} \, e^{ik_b z_a} \, \tilde{h}(k_b), \tag{3.9}
$$

where $\tilde{h}(k)$ is the Fourier transform of $h(z)$.

Similarly,

$$
I_{II} = \sqrt{2\pi} \, e^{ik_c z_a} \, \tilde{h}(k_c). \tag{3.10}
$$

On inserting Eqs. (3.9) and (3.10) in Eq.(3.8), we obtain

$$\left(\frac{\partial}{\partial t} + i\ v_a\ k_a\right) \xi_a(k_a, t) = -\frac{i\epsilon}{\sqrt{2\pi}} \int dk_b \int dk_c\ \tilde{h}(k_b)\ \tilde{h}(k_c)\ \xi_{bc}(k_b, k_c, t)$$

$$\times \underbrace{\int dz_a\ e^{i(k_b+k_c-k_a)z_a}}_{2\pi\ \delta(k_b+k_c-k_a)} . \tag{3.11}$$

On enforcing the δ function, we get the following equation of motion for the a pulse,

$$\left(\frac{\partial}{\partial t} + i\ v_a\ k_a\right) \xi_a(k_a, t) = -i\epsilon\sqrt{2\pi} \int dk_b\ \tilde{h}(k_b)\ \tilde{h}(k_a - k_b)\ \xi_{bc}(k_b, k_a - k_b, t). \tag{3.12}$$

Following this, we shall work on the equation of motion for the b, c pulses. On substituting the response function in Eq.(3.7), we get

$$\left(\frac{\partial}{\partial t} + i\ v_b\ k_b + i\ v_c\ k_c\right) \xi_{bc}(k_b, k_c, t) = -\frac{i\epsilon}{(2\pi)^{3/2}} \int dk_a\ \xi_a(k_a, t)$$

$$\times \underbrace{\int dz_b\ e^{-ik_b z_b}\ h(z_a - z_b)}_{I_{III}} \underbrace{\int dz_c\ e^{-ik_c z_c}\ h(z_a - z_c)}_{I_{IV}} . \tag{3.13}$$

The procedure from here is similar to what we did for the a pulse. We have to evaluate the integrals, I_{III} and I_{IV} along the same line. We shall set $z_a - z_b = y$ in I_{III} and rewrite this integral in terms of y.

This yields

$$
\begin{aligned}
I_{III} &= \sqrt{2\pi}\ e^{-ik_b z_a}\ \frac{1}{\sqrt{2\pi}}\ \int dy\ e^{ik_b y}\ h(y), \\
&= \sqrt{2\pi}\ e^{-ik_b z_a}\ \underbrace{\left(\frac{1}{\sqrt{2\pi}}\ \int dy\ e^{-ik_b y}\ h(y)\right)^{*}}_{\tilde{h}^{*}(k_b)}, \\
&= \sqrt{2\pi}\ e^{-ik_b z_a}\ \tilde{h}^{*}(k_b),
\end{aligned}
\tag{3.14}
$$

where $\tilde{h}^{*}(k)$ is the complex conjugated Fourier transform of $h(z)$.

Similarly,

$$
I_{IV} = \sqrt{2\pi}\ e^{-ik_c z_a}\ \tilde{h}^{*}(k_c).
\tag{3.15}
$$

On inserting Eqs. (3.14) and (3.15) in Eq. (3.13), we obtain

$$
\begin{aligned}
\left(\frac{\partial}{\partial t} + i\ v_b\ k_b + i\ v_c\ k_c\right)\xi_{bc}(k_b, k_c, t) = &-\frac{i\epsilon}{\sqrt{2\pi}}\ \tilde{h}^{*}(k_b)\ \tilde{h}^{*}(k_c)\ \int dk_a\ \xi_a(k_a, t) \\
&\times \underbrace{\int dz_a\ e^{i(k_a - k_b - k_c)z_a}}_{2\pi\ \delta(k_a - k_b - k_c)}.
\end{aligned}
\tag{3.16}
$$

On enforcing the δ function, we get the following equation for the b, c pulses,

$$
\left(\frac{\partial}{\partial t} + i\ v_b\ k_b + i\ v_c\ k_c\right)\xi_{bc}(k_b, k_c, t) = -i\epsilon\sqrt{2\pi}\ \tilde{h}^{*}(k_b)\ \tilde{h}^{*}(k_c)\ \xi_a(k_b + k_c, t).
\tag{3.17}
$$

Thus, we have obtained the equations of motion for the system with a Hamiltonian formalism involving only field operators satisfying canonical commutation relations. Having obtained the equations of motion, we now have solve this system of equations. Before we embark on this, we shall once again write down the two equations of motion in one place together for convenience so that we don't have to go back for any reference.

$$\left(\frac{\partial}{\partial t} + ik_a v_a\right) \xi_a(k_a, t) = -i\epsilon\sqrt{2\pi} \int dk_b \ \tilde{h}(k_b) \ \tilde{h}(k_a - k_b) \ \xi_{bc}(k_b, k_a - k_b, t),$$

$$\left(\frac{\partial}{\partial t} + ik_b v_b + ik_c v_c\right) \xi_{bc}(k_b, k_c, t) = -i\epsilon\sqrt{2\pi} \ \tilde{h}^*(k_b) \ \tilde{h}^*(k_c) \ \xi_a(k_b + k_c, t). \qquad (3.18)$$

We shall solve this system of differential equations by the method of Laplace transform. The Laplace transform of Eqs. (3.18) with respect to time is written as

$$(s + ik_a v_a) \ \tilde{\xi}_a(k_a, s) - \xi_a(k_a, 0) = -i\epsilon\sqrt{2\pi} \int dk_b \ \tilde{h}(k_b) \ \tilde{h}(k_a - k_b) \ \tilde{\xi}_{bc}(k_b, k_a - k_b, s), \qquad (3.19)$$

$$(s + ik_b v_b + ik_c v_c) \ \tilde{\xi}_{bc}(k_b, k_c, s) - \xi_{bc}(k_b, k_c, 0) = -i\epsilon\sqrt{2\pi} \ \tilde{h}^*(k_b) \ \tilde{h}^*(k_c) \ \tilde{\xi}_a(k_b + k_c, s), \qquad (3.20)$$

where $\tilde{\xi}_a$ and $\tilde{\xi}_{bc}$ are the Laplace transform of ξ_a and ξ_{bc}, respectively.

On substituting for $\tilde{\xi}_a(k_b + k_c, s)$ in terms of $\tilde{\xi}_{bc}$ from Eq. (3.19) in Eq. (3.20), and furthermore, setting $\xi_a(k_a, 0) = 0$ since there is no a photon at $t = 0$, we obtain

$$\tilde{\xi}_{bc}(k_b, k_c, s) = \frac{\xi_{bc}(k_b, k_c, 0)}{s + ik_b v_c + ik_c v_c} - \frac{2\pi\epsilon^2}{s + i(k_b + k_c)v_a} \frac{\tilde{h}^*(k_b)\tilde{h}^*(k_c)}{s + ik_b v_b + ik_c v_c}$$

$$\times \int dk\, \tilde{h}(k)\, \tilde{h}(k_b + k_c - k)\, \tilde{\xi}_{bc}(k, k_b + k_c - k, s). \qquad (3.21)$$

The next step is to evaluate the integral on the right hand side of Eq.(3.21). This can

be accomplished by shifting to dummy arguments in the same equation, i.e. $k_b \to k'$ and

$k_c \to k_b + k_c - k'$. Following this, we shall multiply both sides of Eq. (3.21) by

$\tilde{h}(k')\, \tilde{h}(k_b + k_c - k')$ and finally integrate both sides of the equation over k'. This yields,

$$\tilde{I} \equiv \int dk'\, \tilde{h}(k')\, \tilde{h}(k_b + k_c - k')\, \tilde{\xi}_{bc}(k', k_b + k_c - k', s) = \int dk' \frac{\tilde{h}(k')\, \tilde{h}(k_b + k_c - k')}{s + i\, v_b\, k' + i\, v_c\, (k_b + k_c - k')}$$

$$\times \xi_{bc}(k', k_b + k_c - k', 0) - \frac{2\pi\epsilon^2}{s + i\, v_a\, (k_b + k_c)}$$

$$\times \int dk' \frac{|\tilde{h}(k')|^2\, |\tilde{h}(k_b + k_c - k')|^2}{s + i\, v_b\, k' + i\, v_c\, (k_b + k_c - k')}$$

$$\times \underbrace{\int dk\, \tilde{h}(k)\, \tilde{h}(k_b + k_c - k)\, \tilde{\xi}_{bc}(k, k_b + k_c - k, s)}_{\tilde{I}}.$$

$$(3.22)$$

From the previous equation, it is straightforward to get an expression for $\tilde{I}$ which can

be done by moving the term involving $\tilde{I}$ on the right hand side to the left and expressing $\tilde{I}$

in terms of the initial state, $\xi_{bc}(k, k_b + k_c - k, 0)$ and the response function of the medium.

This gives us

$$\tilde{I} \equiv \int dk\ \tilde{h}(k)\ \tilde{h}(k_b + k_c - k)\ \xi_{bc}(k, k_b + k_c - k, s)$$

$$= \left(1 + \frac{2\pi\epsilon^2}{s + i\ v_a\ (k_b + k_c)} \int dk \frac{|\tilde{h}(k)|^2\ |\tilde{h}(k_b + k_c - k)|^2}{s + i\ v_b\ k + i\ v_c\ (k_b + k_c - k)}\right)^{-1}$$

$$\times \int dk\ \frac{\tilde{h}(k)\ \tilde{h}(k_b + k_c - k)}{s + i\ v_b\ k + i\ v_c\ (k_b + k_c - k)}\ \xi_{bc}(k, k_b + k_c - k, 0). \tag{3.23}$$

Now, on substituting Eq. (3.23) in Eq. (3.21), we obtain the following expression for $\tilde{\xi}_{bc}(k_b, k_c, s)$:

$$\tilde{\xi}_{bc}(k_b, k_c, s) = \frac{\xi_{bc}(k_b, k_c, 0)}{s + i\ k_b\ v_b + i\ k_c\ v_c} - \frac{2\pi\epsilon^2}{s + i\ v_a\ (k_b + k_c)}\ \frac{\tilde{h}^*(k_b)\ \tilde{h}^*(k_c)}{s + i\ v_b\ k_b + i\ v_c\ k_c}$$

$$\times \left(1 + \frac{2\pi\epsilon^2}{s + i\ v_a\ (k_b + k_c)} \int dk \frac{|\tilde{h}(k)|^2\ |\tilde{h}(k_b + k_c - k)|^2}{s + i\ v_b\ k + i\ v_c\ (k_b + k_c - k)}\right)^{-1}$$

$$\times \int dk\ \frac{\tilde{h}(k)\ \tilde{h}(k_b + k_c - k)}{s + i\ v_b\ k + i\ v_c\ (k_b + k_c - k)}\ \xi_{bc}(k, k_b + k_c - k, 0). \tag{3.24}$$

Eq. (3.24) is the formal solution to our problem in the s- domain. The next step should naturally be inverting the Laplace transform in order to obtain the expression for the final state, ξ_{bc} as a function of time. However, full inversion of the Laplace transform is in general not possible. Moreover, we are not interested in the detailed time evolution of the final state, but only in the asymptotic state of the b, c wavepacket long after the interaction is over (i.e. as $t \to \infty$). In such a scenario, our first thought would be to exploit the final value theorem of operational calculus that says:

$$\lim_{t \to \infty} \xi_{bc}(k_b, k_c, t) = \lim_{s \to 0} s \; \tilde{\xi}_{bc}(k_b, k_c, s).$$

However, this result is not applicable here because in the absence of interaction (i.e. when $\epsilon = 0$), the system of equations in Eqs.(3.18) does not evolve toward a constant value, but rather we have

$$\xi_{bc}(k_b, k_c, t) = e^{-i(k_b v_b + k_c v_c)t} \; \xi_{bc}(k_b, k_c, 0).$$

This can be easily seen by setting $\epsilon = 0$ in Eqs. (3.18). When we turn on the interaction, we expect that we should be able to separate the changing phase factor from the slowly varying spectral amplitude as follows:

$$\lim_{t \to \infty} [e^{i(k_b v_b + k_c v_c)t} \; \xi_{bc}(k_b, k_c, t)] = \lim_{s \to 0} s \; \tilde{\xi}_{bc}(k_b, k_c, s - ik_b v_b - ik_c v_c). \tag{3.25}$$

Consequently, we make the substitution $s \to s - ik_b v_b - ik_c v_c$ in Eq. (3.24) and take the limit in Eq. (3.25) to obtain

$$\lim_{t \to \infty} [e^{i(k_b v_b + k_c v_c)t} \; \xi_{bc}(k_b, k_c, t)] = \xi_{bc}(k_b, k_c, 0)$$
$$- \frac{2\pi\epsilon^2 \; \tilde{h}^*(k_b) \; \tilde{h}^*(k_c) \; I_1(k_b, k_c)}{ik_b(v_a - v_b) + ik_c(v_a - v_c) + 2\pi\epsilon^2 I_2(k_b, k_c)}, \tag{3.26}$$

where we have defined

$$I_1 \equiv \lim_{s \to 0} \int dk \; \frac{\tilde{h}(k) \; \tilde{h}(k_b + k_c - k)}{s + i(k - k_b)(v_b - v_c)} \; \xi_{bc}(k, k_b + k_c - k, 0), \qquad (3.27)$$

and

$$I_2 \equiv \lim_{s \to 0} \int dk \; \frac{|\tilde{h}(k)|^2 \; |\tilde{h}(k_b + k_c - k)|^2}{s + i(k - k_b)(v_b - v_c)}. \qquad (3.28)$$

We can in fact simplify Eq.(3.27) with some straightforward assumptions. First we shall assume that in this scheme, the b photon starts far behind the c photon and travels with a higher speed than the c pulse such that both of them meet only in the region of interaction. We shall denote the initial position of the center of the b wavepacket by $-z_0$ and the center of the c wavepacket is taken to be $z_c = 0$. Furthermore, we shall also assume that the initial state is factorizable. Thus, we can write

$$\xi_{bc}(k, k_b + k_c - k, 0) = e^{ikz_0} \; \xi_b(k, 0) \; \xi_c(k_b + k_c - k, 0), \qquad (3.29)$$

where $\xi_b(k_b, 0)$ and $\xi_c(k_c, 0)$ are the Fourier transforms of wavepackets centered around $z_b = 0$ and $z_c = 0$, respectively. The exponential factor e^{ikz_0} arises in Eq. (3.29) because as we have stated above, the initial position of the b wavepacket is $z_b = -z_0$. So a displacement in spatial coordinate introduces a phase shift in the Fourier space (or here, the momentum space). Following this, we shall make use of the following identity

$$\frac{1}{s + i(k - k_b)(v_b - v_c)} = \int_0^\infty dt \ e^{-[s + i(k - k_b)(v_b - v_c)]t}, \tag{3.30}$$

to rewrite I_1 as

$$I_1 = \lim_{s \to 0} \int_0^\infty dt \ e^{-st} \ e^{ik_b v_{bc} t} \int_{-\infty}^\infty dk \ e^{ik(z_0 - v_{bc})t}$$

$$\times \ \tilde{h}(k) \ \tilde{h}(k_b + k_c - k) \ \xi_b(k, 0) \ \xi_c(k_b + k_c - k, 0), \tag{3.31}$$

where $v_{bc} \equiv v_b - v_c$. In the previous equation, the integral over k when integrated gives a function that depends only on t. Since, physically, we want z_0 to be much larger than the width of the wavepackets and the length scale of medium nonlocality [i.e. the width of the function $h(z)$], it is reasonable to assume that the integral over k represents a function of t that peaks around $t = t_0 \equiv z_0/v_{bc}$ and decays sufficiently rapidly for both $t \gg t_0$ and $t \ll t_0$. We can also assume that this decay is exponential or faster. So we could first take the limit $s \to 0$ and then formally extend the lower limit of the integral over t to $-\infty$. This procedure yields

$$I_1 = \int_{-\infty}^\infty dk \ e^{ikz_0} \ \tilde{h}(k) \ \tilde{h}(k_b + k_c - k) \ \xi_b(k, 0)\xi_c(k_b + k_c - k, 0)$$

$$\times \underbrace{\int_{-\infty}^\infty dt \ e^{i(k_b - k)v_{bc}t}}_{(2\pi/v_{bc}) \ \delta[(k_b - k)v_{bc}]}. \tag{3.32}$$

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On enforcing the δ function, Eq. (3.32) gets simplified to the following compact expression

$$I_1(k_b, k_c) = \frac{2\pi}{v_{bc}} \, \tilde{h}(k_b) \, \tilde{h}(k_c) \, \underbrace{e^{ik_b z_0} \, \xi_b(k_b, 0) \, \xi_c(k_c, 0)}_{\xi_{bc}(k_b, k_c, 0)},$$

$$= \frac{2\pi}{v_{bc}} \, \tilde{h}(k_b) \, \tilde{h}(k_c) \, \xi_{bc}(k_b, k_c, 0). \tag{3.33}$$

The second integral, I_2 [Eq. (3.28)] can be partially simplified by using a well-known result from the theory of analytic functions, involving Cauchy's principal value:

$$I_2(k_b, k_c) = \frac{\pi}{v_{bc}} \int dk \; \delta[(k - k_b)v_{bc}] \, |\tilde{h}(k)|^2 \, |\tilde{h}(k_b + k_c - k)|^2$$

$$- \frac{i}{v_{bc}} \, \underbrace{P \int dk \frac{|\tilde{h}(k)|^2 \, |\tilde{h}(k_b + k_c - k)|^2}{k - k_b}}_{I_p},$$

$$= \frac{\pi}{v_{bc}} \, |\tilde{h}(k_b)|^2 \, |\tilde{h}(k_c)|^2 - \frac{i}{v_{bc}} I_p, \tag{3.34}$$

where P stands for the principal value and I_p is the notation to denote this integral for brevity. An added advantage of Eq. (3.34) is that it explicitly shows the real and imaginary part of the result.

On substituting Eqs. (3.33) and (3.34) in Eq. (3.26) and setting $(k_b v_{ab} + k_c v_{ac})v_{bc} - 2\pi\epsilon^2 I_p \equiv x$ and $2\pi^2\epsilon^2 \, |\tilde{h}(k_b)|^2 \, |\tilde{h}(k_c)|^2 \equiv y$, to save space and make the intermediate steps easier to follow, where $v_{ab} \equiv v_a - v_b$ and $v_{ac} \equiv v_a - v_c$, we obtain

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$$\xi_{bc}(k_b, k_c, t \to \infty) = e^{i(k_b v_b + k_c v_c)t} \; \xi_{bc}(k_b, k_c, 0) \left(1 - \frac{2y}{ix + y} \right),$$

$$= e^{i(k_b v_b + k_c v_c)t} \; \xi_{bc}(k_b, k_c, 0) \left(\frac{ix - y}{ix + y} \right),$$

$$= e^{i(k_b v_b + k_c v_c)t} \; \xi_{bc}(k_b, k_c, 0) \left(\frac{x + iy}{x - iy} \right). \tag{3.35}$$

Eq.(3.35) can be reduced to a more compact form and thus, the final state can be written as:

$$\xi_{bc}(k_b, k_c, t \to \infty) = e^{-i(k_b v_b + k_c v_c)t} \; \xi_{bc}(k_b, k_c, 0) \; e^{2i\theta(k_b, k_c)}, \tag{3.36}$$

where the first phase factor is just the free evolution and the second one is the phase arising from the interaction with the nonlinear medium:

$$\theta(k_b, k_c) = \tan^{-1} \left(\frac{y}{x} \right) \equiv \tan^{-1} \left(\frac{2\pi^2 \epsilon^2 \; |\tilde{h}(k_b)|^2 \; |\tilde{h}(k_c)|^2}{(k_b v_{ab} + k_c v_{ac}) v_{bc} - 2\pi\epsilon^2 I_p} \right). \tag{3.37}$$

In order to get a π phase shift with unit fidelity, we want $\theta \approx \pi/2$ in the previous expression, to a reasonably good approximation, for all the relevant values of k_b and k_c. It would be very illustrative to see how this can be accomplished by considering a specific example in the following section.

3.3 Specific Example: Gaussian pulses and medium response

We shall now consider a specific case where the response function of the medium is Gaussian and the initial state is also a Gaussian pulse. For this example, the response function in the real space is written as

$$f(z_a, z_b, z_c) = h(z_a - z_b) \, h(z_a - z_c)$$

$$= \frac{1}{\sqrt{\pi\sigma^3}} \, e^{-(z_a - z_b)^2/2\sigma^2} \, e^{-(z_a - z_c)^2/2\sigma^2}, \tag{3.38}$$

where σ is the length scale of medium nonlocality. In the momentum space, we have

$$\tilde{h}(k) = \left(\frac{\sigma}{\pi}\right)^{1/4} \, e^{-k^2\sigma^2/2}. \tag{3.39}$$

The initial state is written as

$$\xi_{bc}(k_b, k_c, 0) = \frac{\sigma_0}{\sqrt{\pi}} \, e^{ik_b z_0} \, e^{-k_b^2\sigma_0^2/2} \, e^{-k_c^2\sigma_0^2/2}, \tag{3.40}$$

where σ_0 is the width of the wavepacket.

We shall first evaluate $I_2(k_b, k_c)$ for this specific example. We shall henceforth denote this integral by simply I_2 for brevity. For a Gaussian response function [see Eq.(3.39)], Eq.(3.28) becomes

$$I_2 = \frac{\sigma}{\pi} \lim_{s \to 0} \int_{-\infty}^{\infty} dk \, \frac{e^{-\sigma^2 k^2} e^{-\sigma^2(k_b + k_c - k)^2}}{s + i(k - k_b)v_{bc}}, \tag{3.41}$$

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where $v_{bc} \equiv v_b - v_c$. We shall make use of Eq.(3.30) to evaluate this integral and interchange the order of integration, i.e. we shall first integrate over k and then integrate over t. This yields

$$I_2 = \frac{\sigma}{\pi} \lim_{s \to 0} \int_0^\infty dt \ e^{-st} \ e^{ik_b v_{bc} t} \underbrace{\int_{-\infty}^\infty dk \ e^{-\sigma^2 k^2} \ e^{-\sigma^2 (k_b + k_c - k)^2} \ e^{ikv_{bc}t}}_{I'}, \qquad (3.42)$$

where

$$I' = \frac{1}{\sigma} \sqrt{\frac{\pi}{2}} \ e^{-itv_{bc}(k_b + k_c)/2} \ e^{-t^2 v^2 / 8\sigma^2} \ e^{-\sigma^2 (k_b + k_c)^2 / 2}.$$

On substituting I' in Eq.(3.42) and integrating over t, we get

$$I_2 = \frac{1}{\sqrt{2\pi}} \ e^{-\sigma^2 (k_b + k_c)^2 / 2} \lim_{s \to 0} \underbrace{\int_0^\infty dt \ e^{-st} \ e^{ik_b v_{bc} t} \ e^{-t^2 v_{bc}^2 / 8\sigma^2} \ e^{-itv_{bc}(k_b + k_c)/2}}_{I''}, \qquad (3.43)$$

where

$$I'' = \frac{\sigma\sqrt{2\pi}}{v_{bc}} \ e^{-\sigma^2 (k_b - k_c)^2 / 2} \left(1 + i \ \text{erfi} \left[\frac{\sigma(k_b - k_c)}{\sqrt{2}} \right] \right).$$

On substituting I'' in Eq. (3.43), we obtain

$$I_2(k_b, k_c) = \frac{\sigma}{v_{bc}} \ e^{-\sigma^2 (k_b^2 + k_c^2)} \left(1 + i \ \text{erfi} \left[\frac{\sigma(k_b - k_c)}{\sqrt{2}} \right] \right). \qquad (3.44)$$

On comparing Eq.(3.44) with Eq.(3.34), we can easily see that the principal value

integral can be evaluated as

$$I_p = -\sigma \ e^{-\sigma^2(k_b^2+k_c^2)} \left(1 + i \ \text{erfi} \left[\frac{\sigma(k_b - k_c)}{\sqrt{2}} \right] \right), \qquad (3.45)$$

which is proportional to the Hilbert transform of a Gaussian.

It is very straightforward to evaluate $I_1(k_b, k_c)$ for this specific example. All we have to

do is to directly substitute Eqs. (3.39) and (3.40) in Eq.(3.33) to get I_1 for a Gaussian

response and a Gaussian initial state.

Once we have calculated I_1 and I_2, we shall substitute these two expressions in

Eq.(3.26) and furthermore, set

$$\frac{(k_b v_{ab} + k_c v_{ac})v_{bc}}{2\pi\epsilon^2\sigma} \ e^{\sigma^2(k_b^2+k_c^2)} \equiv r$$

and

$$\text{erfi} \left[\frac{\sigma(k_b - k_c)}{\sqrt{2}} \right] \equiv q,$$

to save space and make the intermediate steps easier to follow. We thus, obtain the

following expression for the final state, in the same form as Eq.(3.36), for this special case,

$$\xi_{bc}(k_b, k_c, t \to \infty) = e^{-i(k_b v_b + k_c v_c)t} \xi_{bc}(k_b, k_c, 0) \left(\frac{i(r+q) - 1}{i(r+q) + 1} \right),$$

$$= e^{-i(k_b v_b + k_c v_c)t} \frac{\sigma_0}{\sqrt{\pi}} e^{ik_b z_0} e^{-\sigma_0^2(k_b^2 + k_c^2)/2} \left(\frac{(r+q) + i}{(r+q) - i} \right),$$

$$= e^{-i(k_b v_b + k_c v_c)t} \underbrace{\frac{\sigma_0}{\sqrt{\pi}} e^{ik_b z_0} e^{-\sigma_0^2(k_b^2 + k_c^2)/2}}_{\xi_{bc}(k_b, k_c, 0)} e^{2i\theta(k_b, k_c)}, \qquad (3.46)$$

where

$$\theta(k_b, k_c) = \cot^{-1}(r + q)$$

$$= \cot^{-1} \left[\frac{(k_b v_{ab} + k_c v_{ac}) v_{bc}}{2\pi \epsilon^2 \sigma} e^{\sigma^2(k_b^2 + k_c^2)} + \mathrm{erfi} \left(\frac{\sigma(k_b - k_c)}{\sqrt{2}} \right) \right]. \qquad (3.47)$$

In order to get a π phase shift, we want the argument of the inverse cot function to be very close to zero for all the relevant k_b and k_c. We can see that the initial state in Eq. (3.40) restricts $|k_b|$, $|k_c|$ to be of the order of $1/\sigma_0$ as a result of which the argument of the erfi function goes as σ/σ_0. So the condition $\sigma_0 \gg \sigma$ makes the erfi function negligible and the exponential $\exp[\sigma^2(k_b^2 + k_c^2)] \simeq 1$, in Eq.(3.47). We still need to make the first term in the same equation small enough, to make the argument of the inverse cot function close to zero. To accomplish this, we require $|\Delta v|^2 \ll 2\pi \epsilon^2 \sigma \sigma_0$, where Δv is the characteristic velocity difference (i.e. v_{ab}, v_{ac} or v_{bc}). However, note that we cannot let $v_b = v_c$ because this would invalidate the whole analysis; in such a case, the b photon would never catch up with, and interact with, the c photon. Similarly, we cannot completely remove the nonlocality (i.e. let $\sigma \to 0$) because then it would not be possible to keep the first term in

Eq. (3.47) small.

As in the previous chapter, we define the fidelity F and the phase shift ϕ as the overlap between the initial and final state, i.e. $\sqrt{F}e^{i\phi} \equiv \langle\psi(0)|\psi(t)\rangle$. In order to remove the phase factor $\exp[-i(k_b v_b + k_c v_c)t]$, we substitute this phase factor (due to free evolution) for $|\psi(0)\rangle$ in the calculation of fidelity. We thus have,

$$\sqrt{F}e^{i\phi} = \int_{-\infty}^{\infty} dk_b \int_{-\infty}^{\infty} dk_c \, \xi_{bc}^*(k_b, k_c, 0) \, e^{i(k_b v_b + k_c v_c)t} \, \xi_{bc}(k_b, k_c, t \to \infty). \tag{3.48}$$

We can clearly see from both Eqs. (3.36) and (3.46) that when we substitute the expression for the final state in the previous equation, the phase factors due to free evolution cancel each other. The next step is to explicitly calculate the fidelity for our specific problem, i.e. for a Gaussian response and a Gaussian state. On substituting Eqs. (3.33), (3.39), (3.40) and (3.44) in Eq.(3.26) and using Eq.(3.48) to calculate the fidelity, we have

$$\sqrt{F}e^{i\phi} = 1 - 4\epsilon^2\sigma \int_{-\infty}^{\infty} dk_b \int_{-\infty}^{\infty} dk_c \frac{e^{-(\sigma^2+\sigma_0^2)(k_b^2+k_c^2)}}{iv_{bc}(v_{ab}k_b + v_{ac}k_c) + 2\pi\epsilon^2\sigma e^{-\sigma^2(k_b^2+k_c^2)}\text{erfc}[-i\sigma(k_b - k_c)/\sqrt{2}]}, \tag{3.49}$$

where we have used the identity $1 + i\,\text{erfi}[\sigma(k_b - k_c)/\sqrt{2}] = \text{erfc}[-i\sigma(k_b - k_c)/\sqrt{2}]$ in the previous equation. At this stage, we shall switch to dimensionless variables of integration, viz. $\tilde{k}_b \equiv k_b\sigma_0$ and $\tilde{k}_c \equiv k_c\sigma_0$. Furthermore, we shall introduce two more dimensionless parameters $\alpha \equiv v_{ac}v_{bc}/\epsilon^2\sigma^2$ and $\tau \equiv \sigma/\sigma_0$. This scaling simplifies the numerical calculations

to follow and makes it much clearer the limit in which we may expect $\sqrt{F}e^{i\phi} \simeq -1$.

In terms of these new dimensionless variables and parameters, we obtain the following expression for the fidelity:

$$\sqrt{F}e^{i\phi} = 1 - 4\int_{-\infty}^{\infty} d\tilde{k}_b \int_{-\infty}^{\infty} d\tilde{k}_c \frac{e^{-(\tau^2+1)(\tilde{k}_b^2+\tilde{k}_c^2)}}{i\tau\alpha(\tilde{k}_b v_{ab}/v_{ac} + \tilde{k}_c) + 2\pi e^{-\tau^2(\tilde{k}_b^2+\tilde{k}_c^2)}\text{erfc}[-i\tau(\tilde{k}_b - \tilde{k}_c)/\sqrt{2}]}.$$
(3.50)

In the special case considered by Xia *et al.*, the a and the b photons are assumed to travel with the same speed, i.e. $v_a = v_b$. In this case, the term containing v_{ab} would vanish in Eq. (3.50) and thus, we get a more simplified expression for the fidelity that is given by

$$\sqrt{F}e^{i\phi} = 1 - 4\int_{-\infty}^{\infty} d\tilde{k}_b \int_{-\infty}^{\infty} d\tilde{k}_c \frac{e^{-(\tau^2+1)(\tilde{k}_b^2+\tilde{k}_c^2)}}{i\tau\alpha\tilde{k}_c + 2\pi e^{-\tau^2(\tilde{k}_b^2+\tilde{k}_c^2)}\text{erfc}[-i\tau(\tilde{k}_b - \tilde{k}_c)/\sqrt{2}]}, \qquad (3.51)$$

where now $\alpha \equiv v_{bc}^2/\epsilon^2\sigma^2$. We can clearly see from both Eqs. (3.50) and (3.51) that when $\tau \ll 1$ and $\alpha\tau \ll 1$, we get $\sqrt{F}e^{i\phi} \simeq -1$. In this limit, the first term in the denominator of the two previous equations is negligible and the complimentary error function approaches 1. Moreover, the exponential term in the denominator $\exp[-\tau^2(\tilde{k}_b^2 + \tilde{k}_c^2)]$ approaches 1, and the numerator reduces to simply $\exp[-(\tilde{k}_b^2 + \tilde{k}_c^2)]$.

So when $\alpha\tau \ll 1$ and $\tau \ll 1$, we obtain

$$\sqrt{F}e^{i\phi} \simeq 1 - \frac{2}{\pi} \underbrace{\int_{-\infty}^{\infty} d\tilde{k}_b\ e^{-\tilde{k}_b^2}}_{\sqrt{\pi}} \underbrace{\int_{-\infty}^{\infty} d\tilde{k}_c\ e^{-\tilde{k}_c^2}}_{\sqrt{\pi}},$$

$$= -1. \tag{3.52}$$

Figures (3.1) and (3.2) show the result for $\sqrt{F}e^{i\phi}$ as a function of α and τ, respectively, for the case where the three photons travel with different velocities. Here ϕ is limited to take on the values 0 and π, since the quantity being evaluated is real. These figures show that it is more essential to have a small τ than a small α to get unit fidelity with π phase shift, and indeed it does not matter how large α is, we can always achieve the desired result by making τ small enough. This can be clearly seen from figure (3.2). Note that α essentially contains only the medium parameters such as nonlinear coupling strength, pulse speeds and the characteristic length of medium nonlocality, whereas τ depends on the "initial condition", namely the spatial extent of the initial pulse σ_0. So, what this seems to suggest is that irrespective of the properties of the medium, we can always "in principle" get this scheme to work by making the pulse long enough.

The condition on velocity $v_a = v_b \neq v_c$ of Xia et $al.$ seems quite unnatural in a true $\chi^{(2)}$ medium since in that case we would expect the b and the c photons to be much closer in frequency to each other than they are to the a photon in order to satisfy the condition on the conservation of energy: $\omega_a = \omega_b + \omega_c$. However, the scheme conceived by Xia et $al.$ is in fact a four-wave mixing process with a classical pump, so we actually have

$\omega_p + \omega_a = \omega_b + \omega_c$, and here all the three photons a, b and c could be very close to each other in frequency. Nevertheless, we can clearly see from figures (3.3) and (3.4) that this condition is not really necessary and it does not affect the fidelity in any serious way. Both the cases viz. $v_a = v_b$ and when all the photons have different velocities yield similar results.
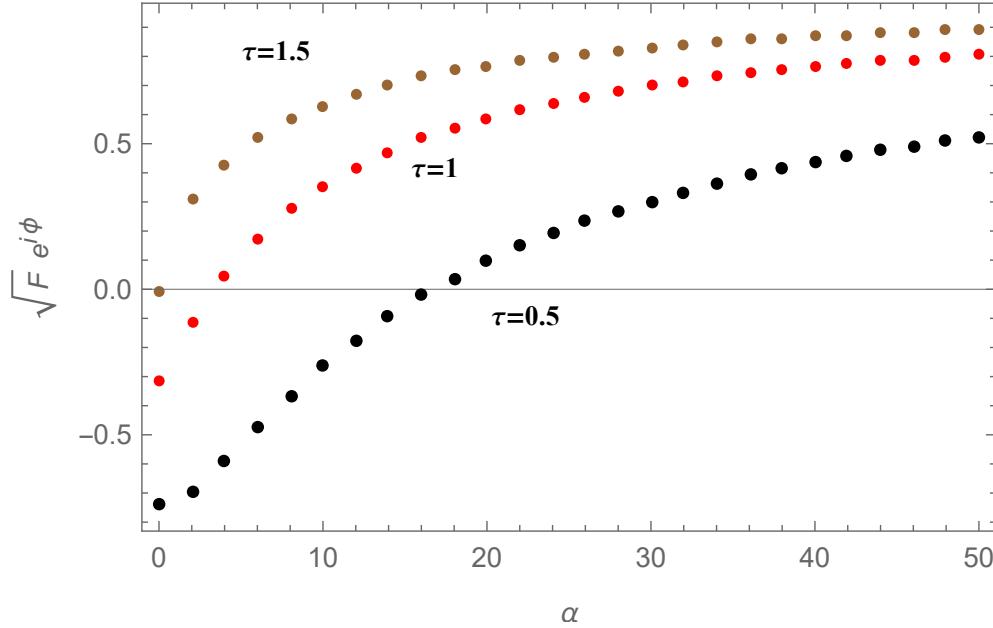
Figure 3.1: Fidelity and phase shift as a function of α for different values of τ when the three photons have different velocities. Here $v_a = 2v_b$ and $v_b = 1.1v_c$.

3.4 Removal of spectral entanglement

In the previous section, we developed a rigorous analytical model for the scheme in which two co-propagating photons travel with different speeds in a nonlocal $\chi^{(2)}$ medium. In the end, we obtained a solution which tells us that we can "in principle" get unit fidelity with a π phase shift when the pulse is very long compared to the characteristic length of the

medium's nonlocality (i.e. $\tau \ll 1$) and $\alpha\tau \ll 1$. Furthermore, we can see from Eq. (3.52) that in this limit, the spectral entanglement of the final state disappears. In this section, we shall discuss in detail the underlying physical mechanism that makes this result possible.
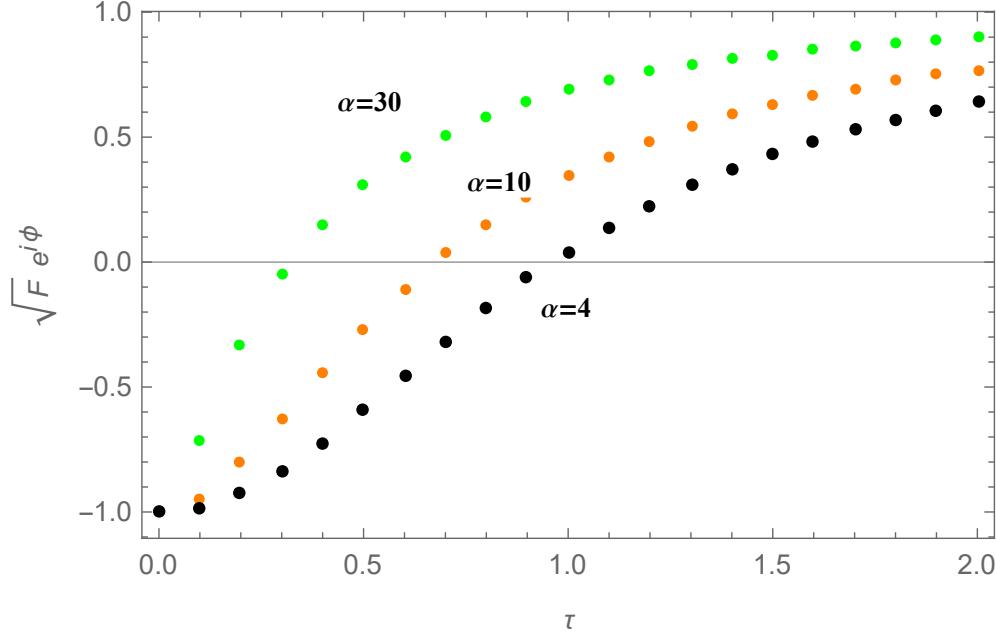
Figure 3.2: Fidelity and phase shift as a function of τ for different values of α for the case where the three photons have different velocities. Here $v_a = 2v_b$ and $v_b = 1.1v_c$.

We have assumed an effectively infinite medium (i.e. we have formally let $L \to \infty$) in our calculation. The interaction Hamiltonian [see Eq. (3.4)] is invariant under translation because the response function $f(z_a, z_b, z_c)$, due to the form we have assumed for it in Eq. (3.2), remains invariant under a displacement of the spatial coordinates z_a, z_b and z_c by a constant amount. This ensures that linear momentum is conserved. We can see that momentum conservation is already enforced in Eqs. (3.18). From the first of Eqs. (3.18), it is clear that any momentum component k_a of the a photon will grow from any two

momentum components k_b and k_c of the b and the c photons, respectively, that add up to k_a. The second of Eqs. (3.18) expresses the same fact in reverse, i.e. k_a splits into two components k_b and k_c such that they once again up to k_a. The mathematical condition for momentum conservation is $k_a' + k_b' = k_a + k_b$. In fact, this is the origin of spectral entanglement in nonlinear processes.
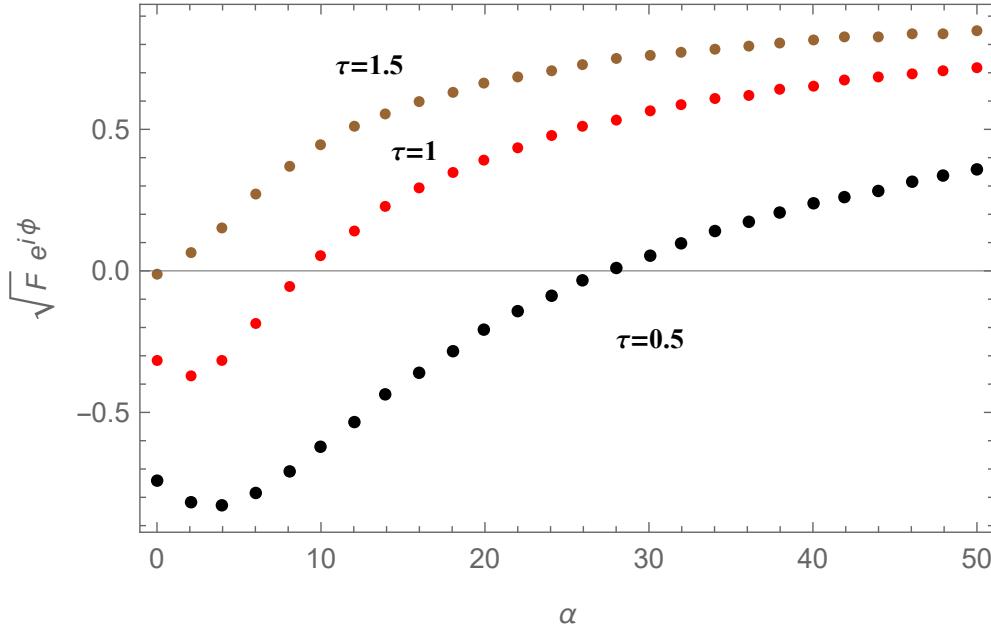
Figure 3.3: Fidelity and phase shift as a function of α for different values of τ when $v_a = v_b$.

Following momentum conservation, the next ingredient, conservation of energy, comes into action when dealing with the integrals in Eq. (3.24), especially the last one on the right hand side. The denominator of this integral through the pole of the Laplace transform gives us the long-time dependence of ξ_{bc}, in the form of a phase factor $\exp[-i(k_b'v_b + k_c'v_c)t]$, where $k_b' \equiv k$ and $k_c' \equiv k_b + k_c - k$. On comparing this with the free-evolution phase factor $\exp[-i(k_b v_b + k_c v_c)t]$ from Eq. (3.36), we get the requirement for

the conservation of energy: $k'_b v_b + k'_c v_c = k_b v_b + k_c v_c$. The treatment of $I_1(k_b, k_c)$ [Eq.(3.27)] in the limit $s \to 0$ or the long-time limit as discussed below Eq.(3.31) yields a δ function $\delta(k_b - k'_b)$. When we enforce this δ function, we get $k_b = k'_b$ and $k_c = k'_c$. We thus see that the simultaneous enforcement of momentum and energy conservation [30] removes the main source of spectral entanglement in the final state. In the previous chapter we just had one condition for both momentum and energy conservation since the two photons traveled with the same speed. So over there, we obtained just one δ function $\delta(k_a + k_b - k'_a - k'_b)$, which when enforced resulted in the spectral entanglement of the final state.
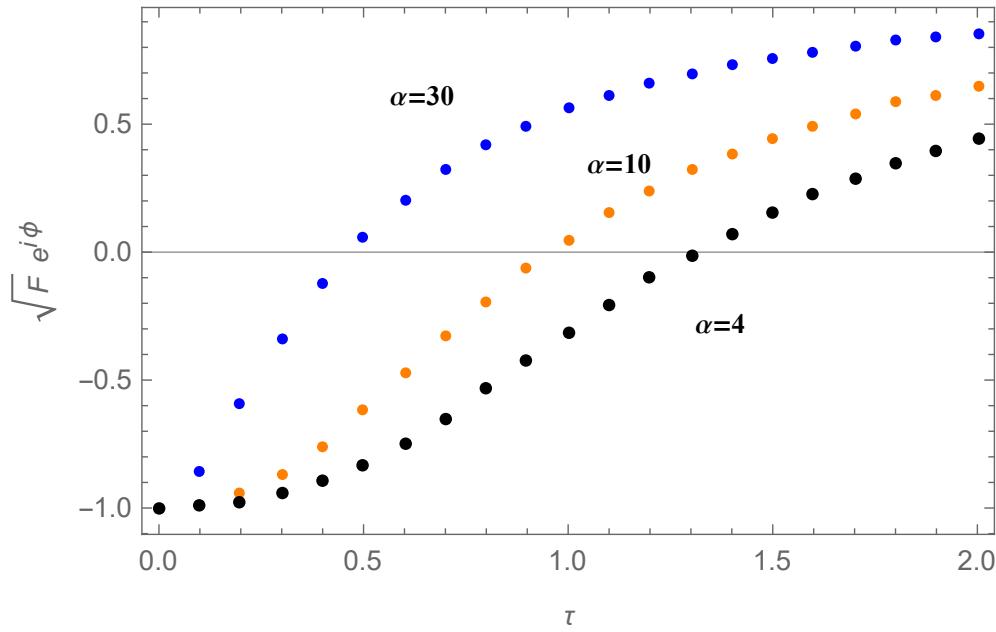
Figure 3.4: Fidelity and phase shift as a function of τ for different values of α when $v_a = v_b$.

We can indeed see the disappearance of spectral entanglement for our problem in the long-time limit, in the approximate time-domain solution to the equations of motion, Eqs. (3.18).

In the limit that we have determined as leading to unit fidelity with a π phase shift ($\alpha\tau \ll 1$), it is possible to obtain an approximate solution to Eqs. (3.18) in the time-domain. We approach this problem in the following way.

First we shall formally integrate the equation of motion for ξ_{bc}, i.e. the second of Eqs. (3.18). This gives us

$$\xi_{bc}(k_b, k_c, t) = \xi_{bc}(k_b, k_c, 0) \, e^{-i(k_b v_b + k_c v_c)t} - i\epsilon\sqrt{2\pi} \, \tilde{h}^*(k_b) \, \tilde{h}^*(k_c)$$
$$\times \int_0^t dt' e^{-i(k_b v_b + k_c v_c)(t-t')} \, \xi_a(k_b + k_c, t'). \tag{3.53}$$

Next, on substituting this in the equation for ξ_a, i.e. the first of Eqs. (3.18), we obtain

$$\left(\frac{\partial}{\partial t} + iv_a k_a\right)\xi_a(k_a, t) = -i\epsilon\sqrt{2\pi} \int dk_b \, \tilde{h}(k_b) \, \tilde{h}(k_a - k_b) \, e^{-i[k_b v_b + (k_a - k_b)v_c]t}$$
$$\times \xi_{bc}(k_b, k_a - k_b, 0) - 2\pi\epsilon^2 \int_0^t dt' \int dk_b \, |\tilde{h}(k_b)|^2 \, |\tilde{h}(k_a - k_b)|^2$$
$$\times e^{-i[k_b v_b + (k_a - k_b)v_c](t-t')} \, \xi_a(k_a, t'). \tag{3.54}$$

The integral over k_b in the previous equation can be evaluated for a specific form of the function $\tilde{h}$. We shall compute this integral in the second term for Gaussian functions that we have assumed for $\tilde{h}$ in Eq. (3.39). On substituting for $\tilde{h}(k)$ from Eq. (3.39) in the second term of Eq. (3.54), we get the following result for the integral over k_b,

$$I_{\tilde{h}} \equiv \frac{\sigma}{\pi} \int_{-\infty}^{\infty} dk_b \; e^{-\sigma^2 k_b^2} \; e^{-\sigma^2 (k_a - k_b)^2} \; e^{-i[k_b v_b + (k_a - k_c) v_c](t - t')},$$

$$= \frac{1}{\sqrt{2\pi}} \; e^{-k_a^2 \sigma^2 / 2} \; e^{-v_{bc}^2 (t-t')^2 / 8\sigma^2} \; e^{-ik_a(v_b + v_c)(t-t')/2}. \tag{3.55}$$

Now, we shall substitute this result in Eq. (3.54), which gives us

$$\left(\frac{\partial}{\partial t} + i v_a k_a \right) \xi_a(k_a, t) = -i\epsilon \sqrt{2\pi} \int dk_b \; \tilde{h}(k_b) \; \tilde{h}(k_a - k_b) \; e^{-i[k_b v_b + (k_a - k_b) v_c] t}$$

$$\times \; \xi_{bc}(k_b, k_a - k_b, 0) - \epsilon^2 \sqrt{2\pi} \; e^{-k_a^2 \sigma^2 / 2} \int_0^t dt' \; \xi_a(k_a, t')$$

$$\times \; e^{-ik_a(v_b + v_c)(t-t')/2} \; e^{-v_{bc}^2 (t-t')^2 / 8\sigma^2}. \tag{3.56}$$

We will now make an approximation that ξ_a is slowly varying compared to $\exp[-v_{bc}^2(t-t')^2/8\sigma^2]$ which essentially only requires σ to be small enough. So pulling $\xi_a(k_a, t)$ out of the integral and extending the lower limit of integration over t' to $-\infty$, and finally completing the squares of the exponential terms, Eq. (3.56) becomes

$$\left(\frac{\partial}{\partial t} + i v_a k_a \right) \xi_a(k_a, t) = -i\epsilon \sqrt{2\pi} \int dk_b \; \tilde{h}(k_b) \; \tilde{h}(k_a - k_b) \; e^{-i[k_b v_b + (k_a - k_b) v_c] t}$$

$$\times \; \xi_{bc}(k_b, k_a - k_b, 0) - \epsilon^2 \sqrt{2\pi} \; e^{-k_a^2 \sigma^2 (v_b^2 + v_c^2)/v_{bc}^2} \; \xi_a(k_a, t)$$

$$\times \; \underbrace{\int_{-\infty}^{t} dt' \; e^{-v_{bc}^2 [(t-t') + 2ik_a \sigma^2 (v_b + v_c)/v_{bc}^2]^2 / 8\sigma^2}}_{I_{t'}}, \tag{3.57}$$

where

$$I_{t'} = \frac{\sigma\sqrt{2\pi}}{v_{bc}} \left(1 - i \operatorname{erfi}\left[\frac{k_a\sigma(v_b + v_c)}{\sqrt{2}v_{bc}}\right]\right).$$

We now have the following equation for ξ_a,

$$\left(\frac{\partial}{\partial t} + iv_a k_a\right)\xi_a(k_a, t) = -i\epsilon\sqrt{2\pi}\int dk_b\ \tilde{h}(k_b)\ \tilde{h}(k_a - k_b)\ e^{-i[k_b v_b + (k_a - k_b)v_c]t}$$

$$\times\ \xi_{bc}(k_b, k_a - k_b, 0) - \frac{2\pi\epsilon^2\sigma}{v_{bc}}\ e^{-k_a^2\sigma^2(v_b^2 + v_c^2)/v_{bc}^2}\ \xi_a(k_a, t)$$

$$\times\left(1 - i \operatorname{erfi}\left[\frac{k_a\sigma(v_b + v_c)}{\sqrt{2}v_{bc}}\right]\right). \tag{3.58}$$

We are working in the limit where $\tau \ll 1$. In other words, $\sigma \ll \sigma_0$. Furthermore, we shall assume that $k_a \sim 1/\sigma_0$. So under these conditions, we set the arguments of both the exponential (in the second term of the previous equation) and the error function to $\simeq 0$. This approximates the exponential to 1 and the error function to zero. Thus, we end up with a simpler equation for ξ_a:

$$\left(\frac{\partial}{\partial t} + iv_a k_a + \gamma\right)\xi_a(k_a, t) \simeq -i\epsilon\sqrt{2\pi}\int dk\ \tilde{h}(k)\ \tilde{h}(k_a - k)$$

$$\times\ e^{-i(kv_{bc} + k_a v_c)t}\ \xi_{bc}(k, k_a - k, 0), \tag{3.59}$$

where $\gamma \equiv 2\pi\epsilon^2\sigma/v_{bc}$, is the rate at which the a photon decays.

This equation can be formally integrated to yield

$$\xi_a(k_a, t) = -i\epsilon\sqrt{2\pi} \; e^{-(\gamma + ik_a v_a)t} \int dk \; \tilde{h}(k) \; \tilde{h}(k_a - k) \; \xi_{bc}(k, k_a - k, 0)$$

$$\times \underbrace{\int_{-\infty}^{t} dt' e^{[\gamma + i(k_a v_a - k v_{bc} - k_a v_c)]t'}}_{I}, \tag{3.60}$$

where

$$I = \frac{e^{[\gamma + i(k_a v_a - k v_{bc} - k_a v_c)]t}}{\gamma + i(k_a v_{ac} - k v_{bc})}.$$

The factor $\exp[-(\gamma + ik_a v_a)t]$ cancels with its complex conjugate from I. Hence, we obtain the following result:

$$\xi_a(k_a, t) = -i\epsilon\sqrt{2\pi} \; e^{-ik_a v_c t} \int dk \; \tilde{h}(k) \; \tilde{h}(k_a - k) \; e^{-ik v_{bc} t} \; \frac{\xi_{bc}(k, k_a - k, 0)}{\gamma + i(k_a v_{ac} - k v_{bc})}. \tag{3.61}$$

We can make one final simplification of Eq. (3.61). We can assume that $\gamma \gg |k_a v_{ac} - k v_{bc}|$. So we can approximate the denominator in Eq. (3.61) to simply γ. Noting that we should expect $k, k_a \sim 1/\sigma_0$, we observe that this is essentially the same condition as $\alpha\tau \ll 1$. This can be easily seen. Note that $\gamma = 2\pi\epsilon^2\sigma/v_{bc}$ and $\alpha\tau = v_{ac} v_{bc}/\epsilon^2 \sigma\sigma_0$. Now, γ can be rewritten as $(2\pi)(\epsilon^2\sigma\sigma_0/v_{bc}v_{ac})(v_{ac}/\sigma_0)$. This is same as $\gamma = 2\pi(1/\alpha\tau)(k_a v_{ac})$. Since $\alpha\tau \ll 1$, this implies that $\gamma \gg 1$. This justifies the final simplification that we intend to make and it now gives us the following simplified expression for ξ_a:

$$\xi_a(k_a, t) \simeq -i\frac{v_{bc}}{\sqrt{2\pi\epsilon\sigma}} \, e^{-ik_a v_c t} \int dk \, \tilde{h}(k) \, \tilde{h}(k_a - k) \, e^{-ikv_{bc}t} \, \xi_{bc}(k, k_a - k, 0), \qquad (3.62)$$

where we have explicitly substituted for γ in the previous equation. If we substitute this into the second of Eqs. (3.18), we obtain

$$\left(\frac{\partial}{\partial t} + ik_b v_b + ik_c v_c\right) \xi_{bc}(k_b, k_c, t) = -\frac{v_{bc}}{\sigma} \, e^{-i(k_b + k_c)v_c t} \, \tilde{h}^*(k_b) \, \tilde{h}^*(k_c) \int dk \, e^{-ikv_{bc}t}$$

$$\times \tilde{h}(k) \, \tilde{h}(k_b + k_c - k) \, \xi_{bc}(k, k_b + k_c - k, 0). \qquad (3.63)$$

In the previous equation, k_a has been replaced by $k_b + k_c$, which is due to momentum conservation. Next, formally integrating this equation yields

$$\xi_{bc}(k_b, k_c, t) \simeq e^{-i(k_b v_b + k_c v_c)t} \, \xi_{bc}(k_b, k_c, 0) - \frac{v_{bc}}{\sigma} \, e^{-i(k_b v_b + k_c v_c)t} \, \tilde{h}^*(k_b) \, \tilde{h}^*(k_c)$$

$$\times \int dk \, \tilde{h}(k) \, \tilde{h}(k_b + k_c - k) \, \xi_{bc}(k, k_b + k_c - k, 0)$$

$$\times \int_0^t dt' \, e^{-i(k-k_b)v_{bc}t'}. \qquad (3.64)$$

Here again, we can take the lower limit of the integral over t' to $-\infty$ since the term in question is negligible before $t = 0$ and extend the upper limit to ∞ to get the long-time limit. When this is done, we observe that the integral over t' gives us a δ function $2\pi\delta[(k - k_b)v_{bc}]$ which is the same as what we obtained while treating the integral I_1 in the limit $s \to 0$. As discussed earlier, this is indeed the condition for energy conservation. On

enforcing this δ function and with the choice of Gaussian function for $\tilde{h}(k)$ [Eq.(3.39)] in the previous equation, we get the following expression for ξ_{bc} in the long-time limit

$$\xi_{bc}(k_b, k_c, t) \simeq e^{-i(k_b v_b + k_c v_c)t} \left[1 - 2e^{\sigma^2(k_b^2 + k_c^2)}\right] \xi_{bc}(k_b, k_c, 0). \qquad (3.65)$$

Noting that $k_b, k_c \sim 1/\sigma_0$ in the limit $\sigma \ll \sigma_0$ or $\tau \ll 1$, the exponential term in Eq. (3.65) approaches 1 which then yields the desired result,

$$\xi_{bc}(k_b, k_c, t) = -e^{-i(k_b v_b + k_c v_c)t} \ \xi_{bc}(k_b, k_c, t). \qquad (3.66)$$

We have quantitatively seen that in the limit $\alpha\tau \ll 1$, the spectral entanglement of the final state is removed in the long-time limit due to the simultaneous enforcement of momentum and energy-conservation.

3.5   Advantage of a long pulse

Whenever we talk about a single-photon pulse, we have to keep in mind that the photon can be anywhere in the pulse. In fact, the photon appears everywhere in the pulse with different probabilities. In the case of co-propagating pulses traveling at the same speed through a nonlinear medium (the problem studied in the previous chapter), increasing the pulse length in fact tends to eliminate the phase shift altogether, because the probability that both the photons would be found in the same narrow time window (determined by the response time of the medium) becomes negligible. This means that in such a case, the two photons may not even interact in the medium in which case no phase is built. However, the

situation considered here is different. Since one pulse sweeps across the other, it is certain that the two photons would meet and interact in the medium, no matter "where in the pulse" each photon is initially.

We can coherently split the wavepackets corresponding to the b and the c photons into roughly $N \simeq \sigma_0/\sigma$ slices of width σ (the characteristic length of medium nonlocality), and the photons will meet in one of those N slices in the region of interaction. Once the two photons meet, they typically have a time $t_{slip} \sim \sigma/v_{bc}$ to interact before the b pulse slips past the c pulse beyond the range of the characteristic length of the medium.

The $b + c \rightarrow a$ conversion process can be be understood semiquantitatively as follows. It is perhaps easiest to visualize this process in the reference frame of the c photon. We shall assume for simplicity that both the pulses (corresponding to the b and the c photons) have the same width σ_0, and divide each of them into N slices.

The state of the b, c pair before the interaction can be symbolically written as

$$|\psi\rangle_{initial} = \frac{1}{\sqrt{N}} \sum_{n=1}^{N} |z_n\rangle_b \otimes \frac{1}{\sqrt{N}} \sum_{m=1}^{N} |z'_m\rangle_c, \tag{3.67}$$

where $|z_n\rangle$ represents a state in which the b photon is found in the slice centered at $z = z_n$ and likewise, $|z'_m\rangle$ represents a state in which the c photon is found in the slice centered at $z = z'_m$, and $1/\sqrt{N}$ is the normalization factor. Note that there are N^2 states in the superposition in Eq. (3.67). Since the two photons are guaranteed to meet and interact in the medium for a time t_{slip}, all the N^2 states in the superposition will be converted into a state that has an a photon in some slice with the probability amplitude ϵt_{slip}. It would be erroneous to assume that the width of the a pulse is also σ_0. This can be easily determined.

Since we are working in the reference frame of the c photon, it means that the c pulse is at rest. Now the b pulse sweeps across the c pulse and in the course of this process, the a photon is created. The time available for the b pulse to pass through the c pulse is $t_{int} = \sigma_0/v_{bc}$, where v_{bc} is the velocity of the b photon in the reference frame of the c photon. In order to compute the width of the a pulse, all we have to do is to simply multiply this time by the velocity of the a photon in the reference frame of the c photon which is v_{ac}. Thus, the width of the a pulse is clearly $\sigma_0 v_{ac}/v_{bc}$ and it contains $N' = N v_{ac}/v_{bc}$ slices of width σ. All the N^2 terms from the superposition in Eq. (3.67) will be distributed among the N' slices of the a pulse. Thus, each slice of the a pulse will contain N^2/N' terms, all with the same amplitude ϵt_{slip}. So the state of the a pulse can be symbolically written as

$$|\phi\rangle_a = \frac{1}{N} \sum_{n=1}^{N'} \frac{N^2}{N'} \, \epsilon t_{slip} \, |z_n\rangle_a. \tag{3.68}$$

The probability for $b + c \to a$ conversion is proportional to the norm which is

$$P \sim \left(\frac{N^2}{N'}\right) (\epsilon^2 t_{slip}^2) = \frac{N \epsilon^2 \sigma^2}{v_{ac} v_{bc}} = \frac{\epsilon^2 \sigma \sigma_0}{v_{bc} v_{ac}} = \frac{1}{\alpha \tau}. \tag{3.69}$$

This explains in a semiquantitative manner why the scheme works in the limit $\alpha\tau \ll 1$.

We should, however, note that the conversion $b + c \to a$ is really only half the process, since what we ultimately want is a new b, c pair. The eventual decay of the a photon is assured as long as it stays in the nonlinear medium for a sufficiently long time, which is automatically guaranteed in our formalism since we are only interested in the $t \to \infty$ limit. In the previous section, we defined $\gamma \equiv 2\pi\epsilon^2\sigma/v_{bc}$ as the rate of decay of the a photon. So,

the lifetime of the a photon is $\tau_a = 1/\gamma$. If we consider a medium of length L, the time spent by the a photon inside the medium is $t_a = L/v_a$. The condition that needs to be satisfied to ensure that the a photon decays into a new b, c pair before it leaves the medium is $t_a/\tau_a = \gamma L/v_a \gg 1$. In other words, we want the time available for the a photon inside the medium to be much larger than its lifetime. Otherwise, the a photon will leave the medium even before the down-conversion process which would ruin the whole scheme. So, what we have to see now is whether the limit in which we operate, i.e. $\alpha\tau \ll 1$ ensures the condition stated above. As we have discussed earlier, the time available for the b photon to interact with the c photon is $t_{int} = \sigma_0/v_{bc}$. The time that the b photon spends inside the nonlinear medium is L/v_b. Clearly, we require $L/v_b > \sigma_0/v_{bc}$, otherwise the two photons may not even interact with each other. This means that $L > \sigma_0 v_b/v_{bc}$. If we multiply both sides of this inequality by γ/v_a and explicitly substitute for γ on the right hand side, we see that

$$
\begin{aligned}
\frac{\gamma L}{v_a} &> \frac{2\pi\epsilon^2\sigma}{v_{bc}} \frac{\sigma_0}{v_{bc}} \frac{v_b}{v_a} \\
&> 2\pi \underbrace{\frac{\epsilon^2\sigma\sigma_0}{v_{bc}v_{ac}}}_{1/\alpha\tau} \frac{v_b}{v_a} \frac{v_{ac}}{v_{bc}} = \frac{2\pi}{\alpha\tau} \frac{v_b}{v_a} \frac{v_{ac}}{v_{bc}}.
\end{aligned}
\tag{3.70}
$$

We can clearly see from Eq.(3.70) that as long as v_b and v_a are not too dissimilar, $\gamma L/v_a \gg 1$ since $\alpha\tau \ll 1$. In other words, the same condition that ensures that $b + c \to a$ conversion happens will also guarantee that $a \to b + c$ conversion too happens in the medium.

We can physically understand the high fidelity shown in the plots in figures (3.2) and

(3.4) in the following way. When $\tau \ll 1$, we see that we obtain unit fidelity with a π phase shift. We can make τ extremely small by either making σ very small or σ_0 very large. In any case, the width of the pulse will be much larger than the length scale of the medium nonlocality. From the Fourier relation, the bandwidth of the medium goes as $1/\sigma$ and the width of the wavepacket goes as $1/\sigma_0$. So, the limit $\tau \ll 1$ means that the bandwidth of the medium is much larger than the width of the wavepackets (in k- space). In this large bandwidth limit, the nonlinearity of the medium interacts with all the spectral components of the wavepackets, or the medium does not filter out any component of the wavepackets, which is why they do not get distorted when they leave the medium. Since the wavepackets travel with different speeds, one sweeps across the other ensuring that they interact in the medium and the three-wave mixing generates a large phase shift. These two features together give us unit fidelity with a π phase shift in this limit.

As τ gets larger, the nonlinearity of the medium does not interact with all the spectral components of the wavepackets because in this case, the width of the wavepackets is larger than the bandwidth of the medium. Here, the medium filters out some of the components of the incoming wavepackets. This results in spectral distortion of the outgoing wavepackets and a low phase shift since the interaction between the two photons is not strong enough. One way to visualize why the interaction is not very strong in this case, is to split the wavepackets into a number of slices or "bins" (in the momentum space). Noting that the photon can be in any one of the slices, there would be no interaction between the two wavepackets if the photons happen to be in the "bins" that get filtered out by the medium in which case the phase that gets built will be low.

Finally, when $\tau \gg 1$ we see from figures (3.2) and (3.4) that the fidelity approaches 1

and the phase shift is zero. This is because, this limit corresponds to the case where the bandwidth of the medium is very much narrower than the width of the wavepackets. So the nonlinearity of the medium does not interact with the wavepackets at all. In other words, the wavepackets will travel as if there is no nonlinear medium, as a result of which there is no spectral distortion and the phase shift is zero since they do not interact with each other.

3.6 Role of nonlocality

We have clearly seen in our discussion until now that we can indeed get the desired high fidelity and large phase shift for any value of the nonlocality parameter σ, as long as it is not exactly zero and provided we make the pulse very long. We might be inclined to contend that this scheme should be physically realizable since we could expect any real world optical medium or a material to exhibit certain degree of spatial nonlocality. In fact, on this specific point, Xia *et al.* [28] cite quite a few references that talk about possible nonlocal effects in four-wave mixing materials due to different physical processes such as charge transport in photorefractive crystal [33] and optical rectification in noncentrosymmetric material [34]. However, it should be mentioned that there is no real physical justification in either these sources or the other available references on nonlinear optical materials, for choosing Gaussian functions for the nonlocal response of the medium, in Eq. (3.38). In this chapter, we have chosen this form for the response function to compare our results with the conclusions of Xia *et al* [28].

It is perhaps best to think of spatial nonlocality as a mathematical artifice to restrict the system's bandwidth in the momentum space to make the theory well behaved. This can be easily seen since the bandwidth $\Delta k \sim 1/\sigma$ which explains why we cannot let $\sigma \to 0$.

This would make the bandwidth infinite which is not physical. It is due to this spatial

nonlocal response in our model that we were able to extend the limits of all the integrals

over k to infinity and still avoid divergence. The nonlocality ensures a finite bandwidth for

the nonlinear medium. This is, in effect, equivalent to the truncation of the system's

bandwidth by hand which we will explicitly verify in the following section.

3.7 "Ad hoc" truncation of interaction bandwidth

In this section, we will assume that the nonlinear interaction involves only a finite range of

frequencies around the pulse's central frequency, and all the frequencies outside this "pass"

bandwidth are unaffected by the nonlinearity of the medium. The goal here is to verify in

an unambiguous manner that we can still get results identical to a nonlocal medium, by

truncating the bandwidth by hand (by introducing "cut-offs" in the Hamiltonian).

The Hamiltonian of the free field is still the same as in Eq. (3.1). However, the

Hamiltonian corresponding to the $\chi^{(2)}$ interaction in this case, is written as

$$\hat{H}_{int} = \hbar\epsilon \int_{z_0}^{z_0+L} dz \; [\hat{A}(z) \; \hat{B}^\dagger(z) \; \hat{C}^\dagger(z) + \hat{A}^\dagger(z) \; \hat{B}(z) \; \hat{C}(z)], \tag{3.71}$$

where once again, we have considered an interaction region of length L.

The operators $\hat{A}$, $\hat{B}$ and $\hat{C}$ are defined as

$$\hat{A}(z) = \frac{1}{\sqrt{2\pi}} \int_{-\Delta k/2}^{\Delta k/2} dk \ e^{ikz} \ \hat{a}(k),$$

$$\hat{B}(z) = \frac{1}{\sqrt{2\pi}} \int_{-\Delta k/2}^{\Delta k/2} dk \ e^{ikz} \ \hat{b}(k),$$

$$\hat{C}(z) = \frac{1}{\sqrt{2\pi}} \int_{-\Delta k/2}^{\Delta k/2} dk \ e^{ikz} \ \hat{c}(k), \tag{3.72}$$

and satisfy the following commutation relations:

$[\hat{A}(z), \hat{A}^\dagger(z')] = [\hat{B}(z), \hat{B}^\dagger(z')] = [\hat{C}(z), \hat{C}^\dagger(z')] = (\Delta k/2\pi) \ \mathrm{sinc}[\Delta k(z - z')/2]$. On

substituting for $\hat{A}$, $\hat{B}$ and $\hat{C}$ from Eq. (3.72) in Eq. (3.71), we get the following expression

for $\hat{H}_{int}$ in the Schrödinger picture,

$$\hat{H}_{int} = \frac{\hbar\epsilon}{(2\pi)^{3/2}} \int_{-\Delta k/2}^{\Delta k/2} dk_a \ \int_{-\Delta k/2}^{\Delta k/2} dk_b \ \int_{-\Delta k/2}^{\Delta k/2} dk_c \ \hat{a}(k_a) \ \hat{b}^\dagger(k_b) \ \hat{c}^\dagger(k_c)$$

$$\times \int_{z_0}^{z_0+L} dz \ e^{i(k_a - k_b - k_c)z} + H.c. \tag{3.73}$$

We shall solve this problem in the interaction picture. The unitary transformation that

takes $\hat{H}_{int}$ from the Schrödinger picture to the interaction picture is

$$\hat{H}_{int}^I = e^{i\hat{H}_0 t/\hbar} \ \hat{H}_{int} \ e^{-i\hat{H}_0 t/\hbar},$$

where the superscript I symbolically denotes the interaction picture. This transformation

gives us the following expression for $\hat{H}_{int}$ in the interaction picture,

$$\hat{H}^I_{int} = \frac{\hbar\epsilon}{(2\pi)^{3/2}} \int_{-\Delta k/2}^{\Delta k/2} dk_a \int_{-\Delta k/2}^{\Delta k/2} dk_b \int_{-\Delta k/2}^{\Delta k/2} dk_c \int_{z_0}^{z_0+L} dz\ e^{i(k_a-k_b-k_c)z}$$

$$\times\ e^{iv_a t \int dk'\ k'\ \hat{a}^\dagger(k')\hat{a}(k')}\ \hat{a}(k_a)\ e^{-iv_a t \int dk'\ k'\ \hat{a}^\dagger(k')\hat{a}(k')}$$

$$\times\ e^{iv_b t \int dk'\ k'\ \hat{b}^\dagger(k')\hat{b}(k')}\ \hat{b}^\dagger(k_b)\ e^{-iv_b t \int dk'\ k'\ \hat{b}^\dagger(k')\hat{b}(k')}$$

$$\times\ e^{iv_c t \int dk'\ k'\ \hat{c}^\dagger(k')\hat{c}(k')}\ \hat{c}^\dagger(k_c)\ e^{-iv_c t \int dk'\ k'\ \hat{c}^\dagger(k')\hat{c}(k')} + H.c.,$$

$$\hat{H}^I_{int} = \frac{\hbar\epsilon}{(2\pi)^{3/2}} \int_{-\Delta k/2}^{\Delta k/2} dk_a \int_{-\Delta k/2}^{\Delta k/2} dk_b \int_{-\Delta k/2}^{\Delta k/2} dk_c\ e^{-i(k_a v_a - k_b v_b - k_c v_c)t}$$

$$\times\ \hat{a}(k_a)\ \hat{b}^\dagger(k_b)\ \hat{c}^\dagger(k_c) \int_{z_0}^{z_0+L} dz\ e^{i(k_a-k_b-k_c)z} + H.c., \tag{3.74}$$

where

$$e^{iv_a t \int dk'\ k'\ \hat{a}^\dagger(k')\hat{a}(k')}\ \hat{a}(k_a)\ e^{-iv_a t \int dk'\ k'\ \hat{a}^\dagger(k')\hat{a}(k')} = \hat{a}(k_a)\ e^{-iv_a k_a t},$$

$$e^{iv_b t \int dk'\ k'\ \hat{b}^\dagger(k')\hat{b}(k')}\ \hat{b}^\dagger(k_b)\ e^{-iv_b t \int dk'\ k'\ \hat{b}^\dagger(k')\hat{b}(k')} = \hat{b}(k_b)\ e^{-iv_b k_b t},$$

and

$$e^{iv_c t \int dk'\ k'\ \hat{c}^\dagger(k')\hat{c}(k')}\ \hat{c}^\dagger(k_c)\ e^{-iv_c t \int dk'\ k'\ \hat{c}^\dagger(k')\hat{c}(k')} = \hat{c}(k_c)\ e^{-iv_c k_c t}.$$

The general state of the system for this problem is same as what we have in Eq. (3.5).

Thus, on substituting Eqs. (3.5) and (3.74) in the Schrödinger equation:

$|\dot{\psi}\rangle = -(i/\hbar)\hat{H}^I_{int}|\psi\rangle$, we get the following equations for the a and the b, c pulses:

$$\frac{\partial}{\partial t}\xi_a(k_a, t) = -\frac{i\epsilon}{(2\pi)^{3/2}} \int_{-\Delta k/2}^{\Delta k/2} dk_b \int_{-\Delta k/2}^{\Delta k/2} dk_c\ e^{i(k_a v_a - k_b v_b - k_c v_c)t}\ \xi_{bc}(k_b, k_c, t)$$

$$\times \int_{z_0}^{z_0+L} dz\ e^{-i(k_a-k_b-k_c)z}, \tag{3.75}$$

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and

$$\frac{\partial}{\partial t}\xi_{bc}(k_b, k_c, t) = -\frac{i\epsilon}{(2\pi)^{3/2}} \int_{-\Delta k/2}^{\Delta k/2} dk_a \ e^{-i(k_a v_a - k_b v_b - k_c v_c)t} \ \xi_a(k_a, t)$$

$$\times \int_{z_0}^{z_0+L} dz \ e^{i(k_a - k_b - k_c)z}. \tag{3.76}$$

In both the equations above, i.e. Eqs. (3.75) and (3.76), we shall let the limits of the integral over z to extend from $-\infty$ to ∞ because as we have already discussed in section 3.2, we let the pulses pass through each other and assume that the interaction starts well after both of them enter the medium and ends well before they leave. This yields us a δ function $2\pi\delta(k_a - k_b - k_c)$, whose argument is the condition for momentum conservation (see section 3.4). On enforcing the δ function in both of these equations, we obtain the following pair of dynamical equations:

$$\frac{\partial}{\partial t}\xi_a(k_a, t) = -\frac{i\epsilon}{\sqrt{2\pi}} \int_{-\Delta k/2}^{\Delta k/2} dk_b \ e^{i(k_a v_{ac} - k_b v_{bc})t} \ \xi_{bc}(k_b, k_a - k_b, t),$$

$$\frac{\partial}{\partial t}\xi_{bc}(k_b, k_c, t) = -\frac{i\epsilon}{\sqrt{2\pi}} \ e^{-i(k_b v_{ab} + k_c v_{ac})t} \ \xi_a(k_b + k_c, t), \tag{3.77}$$

where $v_{ac} \equiv v_a - v_c$ and $v_{bc} \equiv v_b - v_c$.

We shall once again use the method of Laplace transform to solve this system of differential equations. The Laplace transform of the system of equations in Eq. (3.77) can be written as

$$s\,\tilde{\xi}_a(k_a, s) - \xi_a(k_a, 0) = -\frac{i\epsilon}{\sqrt{2\pi}} \int_{-\Delta k/2}^{\Delta k/2} dk_b\, \tilde{\xi}_{bc}(k_b, k_a - k_b, s - ik_a v_{ac} + ik_b v_{bc}), \qquad (3.78)$$

$$s\,\tilde{\xi}_{bc}(k_b, k_c, s) - \xi_{bc}(k_b, k_c, 0) = -\frac{i\epsilon}{\sqrt{2\pi}} \tilde{\xi}_a(k_b + k_c, s + ik_b v_{ab} + ik_c v_{ac}), \qquad (3.79)$$

where $\tilde{\xi}_a$ and $\tilde{\xi}_{bc}$ are the Laplace transforms of ξ_a and ξ_{bc} (with respect to t),

respectively. Next, we shall substitute for $\tilde{\xi}_a$ in Eq. (3.79) in terms of $\tilde{\xi}_{bc}$ from Eq. (3.78).

However, before we do this, we shall shift the arguments of $\tilde{\xi}_a$ in Eq. (3.78), viz.

$k_a \to k_b + k_c$ and $s \to s + ik_b v_{ab} + ik_c v_{ac}$ and set $\xi_a(k_a, 0) = 0$, since there is no a photon at

$t = 0$. This gives us

$$\begin{aligned}
\tilde{\xi}_a(k_b + k_c, s + ik_b v_{ab} + ik_c v_{ac}) = &-\frac{i\epsilon}{\sqrt{2\pi}} \frac{1}{s + ik_b v_{ab} + ik_c v_{ac}} \\
&\times \int_{-\Delta k/2}^{\Delta k/2} dk\, \tilde{\xi}_{bc}(k, k_b + k_c - k, s + i(k - k_b)v_{bc}). \quad (3.80)
\end{aligned}$$

On substituting Eq. (3.80) in Eq.(3.79), we obtain

$$\begin{aligned}
\tilde{\xi}_{bc}(k_b, k_c, s) = &\frac{\xi_{bc}(k_b, k_c, s)}{s} - \frac{\epsilon^2}{2\pi} \frac{1}{s(s + ik_b v_{ab} + ik_c v_{ac})} \\
&\times \int_{-\Delta k/2}^{\Delta k/2} dk\, \tilde{\xi}_{bc}(k, k_b + k_c - k, s + i(k - k_b)v_{bc}). \quad (3.81)
\end{aligned}$$

The next step is to evaluate the integral on the right hand side of Eq. (3.81). This can be accomplished by shifting to dummy arguments in the same equation, i.e. $k_b \to k$, $k_c \to k_b + k_c - k$ and $s \to s + i(k - k_b)v_{bc}$. Following this, we shall integrate both sides of this equation over k. This yields,

$$
\begin{aligned}
I' \equiv \int_{-\Delta k/2}^{\Delta k/2} dk \; \tilde{\xi}_{bc}(k, k_b + k_c - k, s + i(k - k_b)v_{bc}) &= \int_{-\Delta k/2}^{\Delta k/2} dk \; \frac{\xi_{bc}(k, k_b + k_c - k, 0)}{s + i(k - k_b)v_{bc}} - \frac{\epsilon^2}{2\pi} \\
&\times \frac{1}{s + ik_b v_{ab} + ik_c v_{ac}} \underbrace{\int_{-\Delta k/2}^{\Delta k/2} dk \; \frac{1}{s + i(k - k_b)v_{bc}}}_{I''} \\
&\times \underbrace{\int_{-\Delta k/2}^{\Delta k/2} dk' \; \tilde{\xi}_{bc}(k', k_b + k_c - k', s + i(k' - k_b)v_{bc})}_{I'},
\end{aligned}
$$

$$(3.82)$$

where

$$
I'' \equiv \int_{-\Delta k/2}^{\Delta k/2} dk \; \frac{1}{s + i(k - k_b)v_{bc}} = \frac{1}{iv_{bc}} \ln\left[\frac{s + iv_{bc}(\Delta k/2 - k_b)}{s - iv_{bc}(\Delta k/2 + k_b)}\right].
$$

On substituting explicitly for I'' in Eq. (3.82), we get

$$
\begin{aligned}
I' = \int_{-\Delta k/2}^{\Delta k/2} dk \; \frac{\xi_{bc}(k, k_b + k_c - k, 0)}{s + i(k - k_b)v_{bc}} - I' \; \frac{\epsilon^2}{2\pi} \frac{1}{s + ik_b v_{ab} + ik_c v_{ac}} \\
\times \frac{1}{iv_{bc}} \ln\left[\frac{s + iv_{bc}(\Delta k/2 - k_b)}{s - iv_{bc}(\Delta k/2 + k_b)}\right]. \qquad (3.83)
\end{aligned}
$$

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From the previous equation, it is straightforward to get an expression for I' which can be done by moving the term involving I' on the right hand side to the left, and expressing I' in terms of the initial state, $\xi_{bc}(k, k_b + k_c - k, 0)$. This procedure gives us

$$
\begin{aligned}
I' &\equiv \int_{-\Delta k/2}^{\Delta k/2} dk \; \tilde{\xi}_{bc}(k, k_b + k_c - k, s + i(k - k_b)v_{bc}) \\
&= \left(1 + \frac{\epsilon^2}{2\pi i v_{bc}} \frac{1}{s + i k_b v_{ab} + i k_c v_{ac}} \ln\left[\frac{s + i v_{bc}(\Delta k/2 - k_b)}{s - i v_{bc}(\Delta k/2 + k_b)}\right]\right)^{-1} \\
&\quad \times \int_{-\Delta k/2}^{\Delta k/2} dk \; \frac{\xi_{bc}(k, k_b + k_c - k, 0)}{s + i(k - k_b)v_{bc}}.
\end{aligned} \tag{3.84}
$$

On substituting Eq. (3.84) in Eq. (3.81), we obtain the following expression for $\tilde{\xi}_{bc}(k_b, k_c, s)$:

$$
\begin{aligned}
\tilde{\xi}_{bc}(k_b, k_c, s) &= \frac{\xi_{bc}(k_b, k_c, s)}{s} - \frac{i\epsilon^2 v_{bc}}{s} \\
&\quad \times \left(2\pi i v_{bc}(s + i k_b v_{ab} + i k_c v_{ac}) + \epsilon^2 \ln\left[\frac{s + i v_{bc}(\Delta k/2 - k_b)}{s - i v_{bc}(\Delta k/2 + k_b)}\right]\right)^{-1} \\
&\quad \times \int_{-\Delta k/2}^{\Delta k/2} dk \; \frac{\xi_{bc}(k, k_b + k_c - k, 0)}{s + i(k - k_b)v_{bc}}.
\end{aligned} \tag{3.85}
$$

Eq.(3.85) is the formal solution to our problem in the s-domain. Of course, full inversion of this equation is not possible. As we have already seen in section 3.2, we are only interested in the asymptotic state of the wavepacket, long after the interaction is over (i.e. as $t \to \infty$). Here, we can certainly exploit the final value theorem in the form:

$$
\lim_{t \to \infty} \xi_{bc}(k_b, k_c, t) = \lim_{s \to 0} s \; \tilde{\xi}_{bc}(k_b, k_c, s).
$$

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Unlike the situation in section 3.2, this result is applicable here because in the absence of interaction, i.e. when $\epsilon = 0$, ξ_{bc} evolves toward a constant value. This can be readily seen from the system in Eq. (3.77). Thus, the final state in the long-time limit is given by

$$\xi_{bc}(k_b, k_c, t \to \infty) = \xi_{bc}(k_b, k_c, 0) - \left(\frac{2\pi i}{\epsilon^2}(k_b v_{ab} + k_c v_{ac}) - \frac{i}{v_{bc}} \ln \left[\frac{-(\Delta k/2 - k_b)}{(\Delta k/2 + k_b)} \right] \right)^{-1}$$
$$\times \lim_{s \to 0} \int_{-\Delta k/2}^{\Delta k/2} dk \, \frac{\xi_{bc}(k, k_b + k_c - k, 0)}{s + i(k - k_b)v_{bc}}. \tag{3.86}$$

In the model that we developed with spatial nonlocality (see sections 3.2 and 3.3), we identified the limit $\tau << 1$ as leading to the largest fidelity with a π phase shift (see section 3.5 for details). This limit corresponds to the case in which the bandwidth of the medium is much larger compared to the width of the wavepacktets. We shall work in the same regime here. Hence, we can approximate $\Delta k/2 + k_b \simeq \Delta k/2$. In this limit, $\ln[-(\Delta k/2 - k_b)/(\Delta k/2 + k_b)] \simeq \ln(-1) = i\pi$. The integral on the right-hand side of Eq. (3.86) is same as Eq. (3.27) except for the fact that here k is confined to vary only within a finite bandwidth Δk. In the large bandwidth limit, Δk can be treated as a " regularizing factor" which makes the theory formally finite but can be harmlessly taken to infinity in the final result. As discussed in detail below Eq. (3.28) in section 3.2, this integral in the large bandwidth limit yields a δ function $2\pi \, \delta[(k - k_b)v_{bc}]$ which is the condition for energy conservation (see section 3.4). On enforcing the δ function and the large bandwidth limit in Eq. (3.86), we obtain

$$\xi_{bc}(k_b, k_c, t \to \infty) = \xi_{bc}(k_b, k_c, 0) \left(1 - \frac{2\pi\epsilon^2}{\pi\epsilon^2 + 2\pi i v_{bc}(k_b v_{ab} + k_c v_{ac})}\right),$$

$$= \xi_{bc}(k_b, k_c, 0) \left(\frac{2\pi i v_{bc}(k_b v_{ab} + k_c v_{ac}) - \pi\epsilon^2}{2\pi i v_{bc}(k_b v_{ab} + k_c v_{ac}) + \pi\epsilon^2}\right),$$

$$= \xi_{bc}(k_b, k_c, 0) \left(\frac{2\pi v_{bc}(k_b v_{ab} + k_c v_{ac}) + i\pi\epsilon^2}{2\pi v_{bc}(k_b v_{ab} + k_c v_{ac}) - i\pi\epsilon^2}\right). \tag{3.87}$$

The final state in the previous equation can be expressed in a more compact form:

$$\xi_{bc}(k_b, k_c, t \to \infty) = \xi_{bc}(k_b, k_c, 0) \, e^{2i\theta(k_b, k_c)}, \tag{3.88}$$

where

$$\theta(k_b, k_c) = \tan^{-1}\left[\frac{\epsilon^2}{2 v_{bc}(k_b v_{ab} + k_c v_{ac})}\right]. \tag{3.89}$$

We can evidently see from Eq. (3.89) that when we make the nonlinear coupling strength ϵ very large and ensure that v_a, v_b and v_c are not too dissimilar, we can indeed get a π phase shift.

In this section, we have explicitly demonstrated with our analysis that a finite bandwidth medium yields similar results as a spatially nonlocal medium as long as we get two separate mathematical conditions for the conservation of energy and momentum, and we make the bandwidth of the medium much larger than the width of the wavepackets.

3.8 Conclusion

In this chapter, we have carried out a thorough analytical study of the scheme proposed by Xia *et al.* [28] that confirms their claim that a π phase shift with unit fidelity is very much possible. Our analysis also expounds the underlying physical mechanisms that make this outcome feasible.

We have shown that by considering a setup in which the interacting pulses travel with different velocities, the requirements for the conservation of energy and momentum lead to non-equivalent algebraic conditions on the wavevectors and frequencies of the interacting photons which when enforced simultaneously remove the spectral entanglement of the final state.

Our study has also revealed that we can generate a large phase shift by making the pulse very long. This is because when we make the length of the pulse much larger than the characteristic length of medium nonlocality, we essentially make the bandwidth of the medium very large compared to the width of the wavepackets. Thus, in this limit, all the spectral components of the interacting photons contribute to three-wave mixing, as a result of which a large phase shift is generated.

Finally, the nonlocal response is just an artifice to restrict the system's bandwidth to make the theory well behaved. In order words, in effect, this is equivalent to the truncation of the bandwidth by hand to make it finite which is what we expect for any real nonlinear medium. The assumed nonlocality may not even be required for the eventual realization of a conditional phase gate. All that is really necessary is that whatever physical mechanism restricts the system's bandwidth should not degrade the pulse's coherence [35].

Chapter 4

An atomic model for a phase gate with the "giant Kerr" effect

4.1 Introduction

In the last couple of chapters, we had developed a macroscopic model for a phase gate at the single-photon level involving only the interacting field operators. Over there, we had ignored the composition of the nonlinear medium and instead, we characterized the medium by an appropriate Hamiltonian in the multimode framework. We had concluded that as long as we can ensure a finite bandwidth for the medium, we can in principle obtain unit fidelity with a π phase shift. In our theoretical analysis, this was facilitated by the simultaneous enforcement of two separate mathematical conditions for the conservation of energy and momentum.

However, it is to be noted that the models developed in the previous chapters cannot be directly tested in a laboratory. In the current chapter, our goal is to search for a realistic atomic system where the bandwidth-limiting process occurs naturally and develop a consistent, well behaved theory of the interaction of wavepackets with such a medium. Our efforts in this chapter will be to find a plausible scheme that could be physically realized in a laboratory and used for the construction of a conditional phase gate. We are looking at the giant Kerr effect in electromagnetically induced transparency (EIT) as a starting point [36, 37, 38]. We will develop a model to study the interaction of two co-propagating single-photon wavepackets with an ensemble of five-level atoms. In particular, we shall try to understand how the bandwidth of the medium gets restricted here and whether this restriction can be realized without introducing phase noise in the system. Furthermore, in

this model, we would like to see if we once again obtain two separate algebraic conditions for the conservation of energy and momentum and whether their simultaneous enforcement removes the spectral entanglement of the final state.

4.2 A microscopic model via the giant Kerr effect in EIT

We shall first develop a model for a single five-level atomic system and then generalize it to an ensemble by introducing the atomic density in the equations of motion.
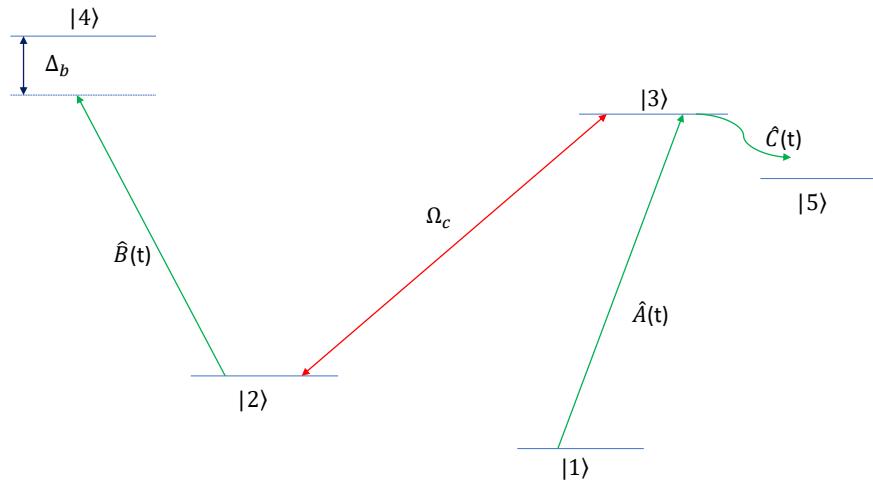

Figure 4.1: Level scheme for the giant Kerr effect. $\hat{A}(t)$ and $\hat{B}(t)$ are weak (single-photon pulses); Ω_c is the EIT "coupling" field.

The Hamiltonian for the atomic system shown in figure (4.1) can be expressed as

$$\hat{H} = \frac{\hbar\Omega_c}{2} \, |2\rangle\langle 3| + \hbar g_{24}^* \, \hat{B}^\dagger(t) \, |2\rangle\langle 4| + \frac{\hbar\Omega_c}{2} \, |3\rangle\langle 2| + \hbar g_{13} \, \hat{A}(t) \, |3\rangle\langle 1| + \hbar\Delta_b \, |4\rangle\langle 4|$$

$$+ \, \hbar g_{24} \, \hat{B}(t) \, |4\rangle\langle 2| + \hbar g_{13}^* \, \hat{A}^\dagger(t) \, |1\rangle\langle 3| + \hbar g_{35} \, \hat{C}(t) \, |3\rangle\langle 5| + \hbar g_{35}^* \, \hat{C}^\dagger(t) \, |5\rangle\langle 3|, \qquad (4.1)$$

where Ω_c is the classical control field that connects levels $|2\rangle \leftrightarrow |3\rangle$ (it is the EIT coupling field) and Δ_b is the detuning from level $|4\rangle$. $\hat{A}(t)$ and $\hat{B}(t)$ represent the single-photon operators that connect the levels $|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |4\rangle$, respectively. In other words, the atom in $|1\rangle$ can absorb the a photon and get excited to $|3\rangle$. Similarly, the atom in $|2\rangle$ can absorb the b photon and get excited to $|4\rangle$. In addition to this, we have another operator $\hat{C}(t)$ that connects $|3\rangle \leftrightarrow |5\rangle$. The purpose of the C photon is to introduce decay naturally in the model. In our scheme, all the fields (i.e. $\hat{A}$, $\hat{C}$ and Ω_c) except $\hat{B}$ are assumed to be resonant with their respective transitions [see figure (4.1)]. In this problem, we are working in the interaction picture right from the beginning which is why the Hamiltonian in Eq. (4.1) only has terms corresponding to the interaction between the atom and the pulses.

The most general atom-field state can be written as

$$|\Psi\rangle = |\psi_1\rangle \, |1\rangle + |\psi_2\rangle \, |2\rangle + |\psi_3\rangle \, |3\rangle + C_4 \, |4\rangle \, |0\rangle + |\psi_5\rangle \, |5\rangle. \qquad (4.2)$$

In the previous equation, $|\psi_1\rangle$, $|\psi_2\rangle$, $|\psi_3\rangle$ and $|\psi_5\rangle$ are the field states, $|0\rangle$ is the field's vacuum, and $|1\rangle$, $|2\rangle$, $|3\rangle$, $|4\rangle$ and $|5\rangle$ are the atomic states. On inserting Eqs. (4.1) and

(4.2) in the Schrödinger equation $|\dot{\Psi}\rangle = -(i/\hbar)\hat{H}|\Psi\rangle$, we obtain the following equations of motion:

$$|\dot{\psi}_1\rangle = -i\, g_{13}^*\, \hat{A}^\dagger(t)\, |\psi_3\rangle, \tag{4.3}$$

$$|\dot{\psi}_2\rangle = -\frac{i}{2}\, \Omega_c\, |\psi_3\rangle - i\, g_{24}^*\, \hat{B}^\dagger(t)\, C_4\, |0\rangle, \tag{4.4}$$

$$|\dot{\psi}_3\rangle = -\frac{i}{2}\, \Omega_c\, |\psi_2\rangle - i\, g_{13}\, \hat{A}(t)\, |\psi_1\rangle - i\, g_{35}\, \hat{C}(t)\, |\psi_5\rangle, \tag{4.5}$$

$$\dot{C}_4 = -i\, \Delta_b\, C_4 - i\, g_{24}\, \hat{B}(t)\, |\psi_2\rangle, \tag{4.6}$$

and

$$|\dot{\psi}_5\rangle = i\, g_{35}^*\, \hat{C}^\dagger(t)\, |\psi_3\rangle. \tag{4.7}$$

We shall begin solving this system of equations by starting with Eq. (4.7) whose formal solution can be written as

$$|\psi_5(t)\rangle = -i\, g_{35}^* \int_0^t dt'\, \hat{C}^\dagger(t')\, |\psi_3(t')\rangle. \tag{4.8}$$

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Next, substituting Eq. (4.8) in Eq. (4.5) yields

$$|\dot{\psi}_3(t)\rangle = -\frac{i}{2}\,\Omega_c\,|\psi_2(t)\rangle - i\,g_{13}\,\hat{A}(t)\,|\psi_1(t)\rangle - |g_{35}|^2 \int_0^t dt'\,\hat{C}(t)\,\hat{C}^\dagger(t')\,|\psi_3(t')\rangle. \qquad (4.9)$$

Note that $\hat{C}(t)\,\hat{C}^\dagger(t') = \hat{C}^\dagger(t')\,\hat{C}(t) + \delta(t - t')$ and furthermore, $\hat{C}(t)\,|\psi_3(t')\rangle = 0$, since there is no C photon at time t'. Putting all this together in Eq. (4.9) results in

$$
\begin{aligned}
|\dot{\psi}_3(t)\rangle &= -\frac{i}{2}\,\Omega_c\,|\psi_2(t)\rangle - i\,g_{13}\,\hat{A}(t)\,|\psi_1(t)\rangle - |g_{35}|^2 \int_0^t dt'\,\delta(t - t')\,|\psi_3(t')\rangle, \\
&= -\frac{i}{2}\,\Omega_c\,|\psi_2(t)\rangle - i\,g_{13}\,\hat{A}(t)\,|\psi_1(t)\rangle - \frac{|g_{35}|^2}{2}\,|\psi_3(t)\rangle, \\
&= -\frac{i}{2}\,\Omega_c\,|\psi_2(t)\rangle - i\,g_{13}\,\hat{A}(t)\,|\psi_1(t)\rangle - \gamma\,|\psi_3(t)\rangle, \qquad (4.10)
\end{aligned}
$$

where $\gamma \equiv |g_{35}|^2/2$. Note that in the previous equation since we are enforcing the δ function in the upper limit, we get a factor of $1/2$ which is absorbed in the definition of γ.

Next, we shall consider the following simplified form of Eqs. (4.4) and (4.10) [ignoring for the moment the quantum fields; the idea here is to treat the classical coupling field to all orders, as is commonly done in EIT]:

$$
\begin{aligned}
|\dot{\psi}_2\rangle &= -\frac{i}{2}\,\Omega_c\,|\psi_3\rangle, \\
|\dot{\psi}_3\rangle &= -\frac{i}{2}\,\Omega_c\,|\psi_2\rangle - \gamma\,|\psi_3\rangle, \qquad (4.11)
\end{aligned}
$$

which can be expressed in a more compact form as

$$\frac{d}{dt}\begin{pmatrix}|\psi_3\rangle\\|\psi_2\rangle\end{pmatrix}=\underbrace{\begin{pmatrix}-\gamma & -i\,\Omega_c/2\\-i\,\Omega_c/2 & 0\end{pmatrix}}_{M}\begin{pmatrix}|\psi_3\rangle\\|\psi_2\rangle\end{pmatrix}. \tag{4.12}$$

Our motivation here is to express $|\psi_2\rangle$ and $|\psi_3\rangle$ as a linear combination of the eigenvectors of M. Since M is a 2×2 matrix, we obviously have two eigenvalues given by

$$\lambda' = -\frac{1}{2}\left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right),$$

and

$$\lambda'' = -\frac{1}{2}\left(\gamma - \sqrt{\gamma^2 - \Omega_c^2}\right).$$

We shall denote the eigenvectors corresponding to λ' and λ'' as $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$, respectively, which can be written as

$$|\tilde{\psi}_2\rangle = \begin{pmatrix}\left[\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right]/\Omega_c\\i\end{pmatrix},$$

and

$$|\tilde{\psi}_3\rangle = \begin{pmatrix}\left[\gamma - \sqrt{\gamma^2 - \Omega_c^2}\right]/2\gamma\\i\,\Omega_c/2\gamma\end{pmatrix}.$$

If we define $|\psi_2\rangle \equiv \begin{pmatrix} 0 \\ 1 \end{pmatrix}$ and $|\psi_3\rangle \equiv \begin{pmatrix} 1 \\ 0 \end{pmatrix}$, we could then rewrite $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$ as

$$|\tilde{\psi}_2\rangle = \left(\frac{\gamma + \sqrt{\gamma^2 - \Omega_c^2}}{\Omega_c} \right) |\psi_3\rangle + i \, |\psi_2\rangle, \tag{4.13}$$

and

$$|\tilde{\psi}_3\rangle = \left(\frac{\gamma - \sqrt{\gamma^2 - \Omega_c^2}}{2\gamma} \right) |\psi_3\rangle + i \left(\frac{\Omega_c}{2\gamma} \right) |\psi_2\rangle. \tag{4.14}$$

Our next step is to invert Eqs. (4.13) and (4.14) so that we can express $|\psi_2\rangle$ and $|\psi_3\rangle$ in terms of $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$. This procedure yields

$$|\psi_2\rangle = -i\frac{\gamma}{\Omega_c} \left(1 + \frac{\gamma}{\sqrt{\gamma^2 - \Omega_c^2}} \right) |\tilde{\psi}_3\rangle - \frac{i}{2} \left(1 - \frac{\gamma}{\sqrt{\gamma^2 - \Omega_c^2}} \right) |\tilde{\psi}_2\rangle, \tag{4.15}$$

and

$$|\psi_3\rangle = \frac{\Omega_c}{2\sqrt{\gamma^2 - \Omega_c^2}} |\tilde{\psi}_2\rangle - \frac{\gamma}{\sqrt{\gamma^2 - \Omega_c^2}} |\tilde{\psi}_3\rangle. \tag{4.16}$$

Following this, we now have to express the equations of motion in Eqs. (4.3) and (4.6) in terms of $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$.

Eq. (4.3) can be rewritten as

$$|\dot{\psi}_1\rangle = -i\frac{g_{13}^*}{\sqrt{\gamma^2 - \Omega_c^2}} \, \hat{A}^\dagger(t) \left(\frac{\Omega_c}{2} |\tilde{\psi}_2\rangle - \gamma |\tilde{\psi}_3\rangle \right). \tag{4.17}$$

Later, we shall come back to the previous equation, once we have the solution for $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$. Now, we shall consider the time derivative of Eq. (4.13) which is given by

$$|\dot{\tilde{\psi}}_2\rangle = \frac{\gamma + \sqrt{\gamma^2 - \Omega_c^2}}{\Omega_c} \, |\dot{\psi}_3\rangle + i \, |\dot{\psi}_2\rangle. \tag{4.18}$$

Next, on substituting for $|\dot{\psi}_2\rangle$ and $|\dot{\psi}_3\rangle$ from Eqs. (4.4) and (4.5) in Eq. (4.18), we obtain

$$|\dot{\tilde{\psi}}_2\rangle = -i\frac{g_{13}}{\Omega_c} \left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right) \hat{A}(t) \, |\psi_1\rangle + g_{24}^* \, \hat{B}^\dagger(t) \, c_4(t) \, |0\rangle$$
$$- \frac{i}{2} \left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right) |\psi_2\rangle + \left(\frac{\Omega_c^2 - 2\gamma\left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right)}{2\Omega_c}\right) |\psi_3\rangle. \tag{4.19}$$

Finally, on substituting for $|\psi_2\rangle$ and $|\psi_3\rangle$ in the previous equation, in terms of $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$ from Eqs. (4.15) and (4.16), respectively, and carrying out some simplification, we get

$$|\dot{\tilde{\psi}}_2\rangle + \frac{1}{2} \left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right) |\tilde{\psi}_2\rangle = -i \, \frac{g_{13}}{\Omega_c} \left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right) \hat{A}(t) \, |\psi_1\rangle$$
$$+ g_{24}^* \, C_4(t) \, \hat{B}^\dagger(t) \, |0\rangle, \tag{4.20}$$

which can be formally integrated to give

$$|\tilde{\psi}_2\rangle = -i \, \frac{g_{13}}{\Omega_c} \left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right) \int_0^t dt' \, e^{-\left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right)(t-t')/2} \, \hat{A}(t') \, |\psi_1(t')\rangle$$
$$+ g_{24}^* \int_0^t dt' \, e^{-\left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right)(t-t')/2} \, C_4(t') \, \hat{B}^\dagger(t') \, |0\rangle. \tag{4.21}$$

Our next task is to consider the time derivative of Eq. (4.14) which is given by

$$|\ddot{\psi}_3\rangle = \frac{\gamma - \sqrt{\gamma^2 - \Omega_c^2}}{2\gamma} \, |\dot{\psi}_3\rangle + i \, \frac{\Omega_c}{2\gamma} \, |\dot{\psi}_2\rangle. \tag{4.22}$$

Next, on substituting for $|\dot{\psi}_2\rangle$ and $|\dot{\psi}_3\rangle$ from Eqs. (4.4) and (4.5), we obtain

$$|\ddot{\psi}_3\rangle = -i \, \frac{g_{13}}{2\gamma} \left(\gamma - \sqrt{\gamma^2 - \Omega_c^2} \right) \hat{A}(t) \, |\psi_1\rangle + \frac{\Omega_c g_{24}^*}{2\gamma} \, C_4(t) \, \hat{B}^\dagger(t) \, |0\rangle$$
$$- i \, \frac{\Omega_c}{4\gamma} \left(\gamma - \sqrt{\gamma^2 - \Omega_c^2} \right) |\psi_2\rangle + \left[\frac{\Omega_c^2 - \left(2\gamma^2 - 2\gamma\sqrt{\gamma^2 - \Omega_c^2} \right)}{4\gamma} \right] |\psi_3\rangle. \tag{4.23}$$

Finally, substituting for $|\psi_2\rangle$ and $|\psi_3\rangle$ in the previous equation, in terms of $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$ from Eqs. (4.15) and (4.16), respectively, and carrying out some simplification yields

$$|\ddot{\tilde{\psi}}_3\rangle + \frac{1}{2} \left(\gamma - \sqrt{\gamma^2 - \Omega_c^2} \right) |\dot{\tilde{\psi}}_3\rangle = -i \, \frac{g_{13}}{2\gamma} \left(\gamma - \sqrt{\gamma^2 - \Omega_c^2} \right) \hat{A}(t) \, |\psi_1\rangle$$
$$+ \frac{\Omega_c \, g_{24}^*}{2\gamma} \, C_4(t) \, \hat{B}^\dagger(t) \, |0\rangle, \tag{4.24}$$

which can be formally integrated to give

$$|\tilde{\psi}_3\rangle = -i \, \frac{g_{13}}{2\gamma} \left(\gamma - \sqrt{\gamma^2 - \Omega_c^2} \right) \int_0^t dt' \, e^{-\left(\gamma - \sqrt{\gamma^2 - \Omega_c^2} \right)(t-t')/2} \, \hat{A}(t') \, |\psi_1(t')\rangle$$
$$+ \frac{\Omega_c \, g_{24}^*}{2\gamma} \int_0^t dt' \, e^{-\left(\gamma - \sqrt{\gamma^2 - \Omega_c^2} \right)(t-t')/2} \, C_4(t') \, \hat{B}^\dagger(t') \, |0\rangle. \tag{4.25}$$

Our next task is to deal with Eq. (4.6), i.e. the equation of motion for $C_4(t)$. On substituting for $|\psi_2\rangle$ in Eq. (4.6), in terms of $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$ using Eq. (4.15), we obtain

$$\dot{C}_4 + i\,\Delta_b\,C_4 = -\frac{\gamma\,g_{24}}{\Omega_c}\left(1 + \frac{\gamma}{\sqrt{\gamma^2 - \Omega_c^2}}\right)\hat{B}(t)\,|\tilde{\psi}_3\rangle - \frac{g_{24}}{2}\left(1 - \frac{\gamma}{\sqrt{\gamma^2 - \Omega_c^2}}\right)\hat{B}(t)\,|\tilde{\psi}_2\rangle.$$

(4.26)

On substituting for $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$ from Eqs. (4.21) and (4.25), respectively, in Eq. (4.26), we get

$$\dot{C}_4 + i\,\Delta_b\,C_4 = i\,\frac{g_{24}\,g_{13}\,\Omega_c}{2\sqrt{\gamma^2 - \Omega_c^2}}\int_0^t dt'\,e^{-\left(\gamma - \sqrt{\gamma^2 - \Omega_c^2}\right)(t-t')/2}\,\hat{B}(t)\,\hat{A}(t')\,|\psi_1(t')\rangle$$

$$-\frac{|g_{24}|^2}{2}\left(1 + \frac{\gamma}{\sqrt{\gamma^2 - \Omega_c^2}}\right)\int_0^t dt'\,e^{-\left(\gamma - \sqrt{\gamma^2 - \Omega_c^2}\right)(t-t')/2}\,C_4(t')\,\hat{B}(t)\,\hat{B}^\dagger(t')\,|0\rangle$$

$$- i\,\frac{g_{24}\,g_{13}\,\Omega_c}{2\sqrt{\gamma^2 - \Omega_c^2}}\int_0^t dt'\,e^{-\left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right)(t-t')/2}\,\hat{B}(t)\,\hat{A}(t')\,|\psi_1(t')\rangle$$

$$-\frac{|g_{24}|^2}{2}\left(1 + \frac{\gamma}{\sqrt{\gamma^2 - \Omega_c^2}}\right)\int_0^t dt'\,e^{-\left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right)(t-t')/2}\,C_4(t')\,\hat{B}(t)\,\hat{B}^\dagger(t')\,|0\rangle. \quad (4.27)$$

In order to simplify the previous equation further, we shall make use of the following results from the commutation relation between $\hat{B}$ and $\hat{B}^\dagger$:

$$\hat{B}(t)\,\hat{B}^\dagger(t') = \hat{B}^\dagger(t')\,\hat{B}(t) + \delta(t - t'),$$

and

$$\hat{B}^{\dagger}(t')\ \hat{B}(t)|0\rangle = 0.$$

On exploiting these results, we shall be left with a δ function in the second and fourth terms in Eq. (4.27), and to enforce the δ function, we shall resort to the following identity:

$$\int_0^t dt'\ f(t')\ \delta(t - t') = \frac{1}{2}\ f(t).$$

On using this result and furthermore, defining

$$\frac{1}{2}\left(\gamma - \sqrt{\gamma^2 - \Omega_c^2}\right) \equiv \Gamma_-,$$

and

$$\frac{1}{2}\left(\gamma + \sqrt{\gamma^2 - \Omega_c^2}\right) \equiv \Gamma_+,$$

and carrying out some simplification, we obtain

$$\dot{C}_4 + \left(\frac{|g_{24}|^2}{2} + i\,\Delta_b\right) C_4 = i\,\frac{g_{24}\ g_{13}\ \Omega_c}{2\ \sqrt{\gamma^2 - \Omega_c^2}} \int_0^t dt'\left(e^{-\Gamma_-(t-t')} - e^{-\Gamma_+(t-t')}\right)$$

$$\times \hat{B}(t)\ \hat{A}(t')\ |\psi_1(t')\rangle, \tag{4.28}$$

which can be formally integrated to give

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$$C_4(t) = i \, \frac{g_{24} \, g_{13} \, \Omega_c}{2 \, \sqrt{\gamma^2 - \Omega_c^2}} \int_0^t dt' \; e^{-\left(|g_{24}|^2/2 + i\Delta_b\right)(t-t')} \; \hat{B}(t')$$

$$\times \int_0^{t'} dt'' \; \left(e^{-\Gamma_-(t'-t'')} - e^{-\Gamma_+(t'-t'')}\right) \; \hat{A}(t'') \, |\psi_1(t'')\rangle. \qquad (4.29)$$

Having obtained the formal solution for C_4, the next step is to substitute this result in the solutions for $|\tilde{\psi}_2\rangle$ and $|\tilde{\psi}_3\rangle$, in Eqs. (4.21) and (4.25), respectively.

We shall first consider $|\tilde{\psi}_2\rangle$. Now, on substituting Eq. (4.29) in Eq. (4.21), we obtain

$$|\tilde{\psi}_2(t)\rangle = -2 \, i \, \Gamma_+ \frac{g_{13}}{\Omega_c} \int_0^t dt' \; e^{-\Gamma_+(t-t')} \; \hat{A}(t') \, |\psi_1(t')\rangle + i \, \frac{|g_{24}|^2 \, g_{13} \, \Omega_c}{2 \, \sqrt{\gamma^2 - \Omega_c^2}} \int_0^t dt' \; e^{-\Gamma_+(t-t')} \; \hat{B}^\dagger(t')$$

$$\times \int_0^{t'} dt'' \; e^{-\left(|g_{24}|^2/2 + i\Delta_b\right)(t'-t'')} \; \hat{B}(t'') \int_0^{t''} dt''' \; \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')}\right) \; \hat{A}(t''') \, |\psi_1(t''')\rangle.$$

$$(4.30)$$

Next, we shall consider $|\tilde{\psi}_3\rangle$. On substituting Eq. (4.29) in Eq. (4.25), we get

$$|\tilde{\psi}_3(t)\rangle = -i \, \Gamma_- \frac{g_{13}}{\gamma} \int_0^t dt' \; e^{-\Gamma_-(t-t')} \; \hat{A}(t') \, |\psi_1(t')\rangle + i \, \frac{\Omega_c}{2\gamma} \frac{|g_{24}|^2 \, g_{13} \, \Omega_c}{2\sqrt{\gamma^2 - \Omega_c^2}} \int_0^t dt' \; e^{-\Gamma_-(t-t')} \; \hat{B}^\dagger(t')$$

$$\times \int_0^{t'} dt'' \; e^{-\left(|g_{24}|^2/2 + i\Delta_b\right)(t'-t'')} \; \hat{B}(t'') \int_0^{t''} dt''' \; \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')}\right) \; \hat{A}(t''') \, |\psi_1(t''')\rangle.$$

$$(4.31)$$

Following this, we shall substitute Eqs. (4.30) and (4.31) in Eq. (4.17) and carry out some simplification which finally yields the following closed-form equation for $|\dot{\psi}_1\rangle$:

$$
|\dot{\psi}_1\rangle = \frac{|g_{13}|^2}{\sqrt{\gamma^2 - \Omega_c^2}} \int_0^t dt' \left(\Gamma_- \, e^{-\Gamma_-(t-t')} - \Gamma_+ \, e^{-\Gamma_+(t-t')} \right) \hat{A}^\dagger(t) \, \hat{A}(t') \, |\psi_1(t')\rangle
$$

$$
+ \frac{|g_{13}|^2 \, |g_{24}|^2 \, \Omega_c^2}{4(\gamma^2 - \Omega_c^2)} \int_0^t dt' \left(e^{-\Gamma_+(t-t')} - e^{-\Gamma_-(t-t')} \right) \int_0^{t'} dt'' \, e^{-\left(|g_{24}|^2/2 + i\Delta_b\right)(t'-t'')}
$$

$$
\times \int_0^{t''} dt''' \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')} \right) \hat{A}^\dagger(t) \, \hat{A}(t''') \, \hat{B}^\dagger(t') \, \hat{B}(t'') \, |\psi_1(t''')\rangle. \quad (4.32)
$$

Until now, we have been working with field states without resorting to any specific representation. However, at this stage, we would switch to the frequency-space using a wavepacket to the represent the a and the b photons in a multimode framework. We shall thus, define

$$
|\psi_1(t)\rangle \equiv \int d\omega' \int d\omega'' \, f(\omega', \omega'', t) \, \hat{a}_{\omega'}^\dagger \, \hat{b}_{\omega''}^\dagger \, |0\rangle.
$$

The operators for the field interacting with an atom at a location z' are

$$
\hat{A}\left(t - \frac{z'}{c}\right) \equiv \frac{1}{\sqrt{2\pi}} \int d\omega_a \, e^{-i\omega_a(t - z'/c)} \, \hat{a}_{\omega_a}
$$

and

$$
\hat{B}\left(t - \frac{z'}{c}\right) \equiv \frac{1}{\sqrt{2\pi}} \int d\omega_b \, e^{-i\omega_b(t - z'/c)} \, \hat{b}_{\omega_b}.
$$

We assume that the medium extends from $-L/2$ to $L/2$ and we approximate the sum over all the atoms with coordinates z' by an integral over a continuous distribution $\rho(z')$.

On inserting these definitions in Eq. (4.32) and furthermore, introducing the atomic density $\rho(z')$ in the previous equation, we end up getting the following equation of motion for the wavepacket:

$$
\begin{aligned}
\frac{\partial}{\partial t} f(\omega_a, \omega_b, t) =& \frac{|g_{13}|^2}{2\pi\sqrt{\gamma^2 - \Omega_c^2}} \int_{-L/2}^{L/2} dz' \, \rho(z') \int_0^t dt' \left(\Gamma_- \, e^{-\Gamma_-(t-t')} - \Gamma_+ \, e^{-\Gamma_+(t-t')}\right) \\
& \times \int d\omega_a' \, f(\omega_a', \omega_b, t) \, e^{i\omega_a(t-z'/c)} \, e^{-i\omega_a'(t'-z'/c)} + \frac{|g_{13}|^2 \, |g_{24}|^2 \, \Omega_c^2}{16 \, \pi^2 \, (\gamma^2 - \Omega_c^2)} \int_{-L/2}^{L/2} dz' \, \rho(z') \\
& \times \int_0^t dt' \left(e^{-\Gamma_+(t-t')} - e^{-\Gamma_-(t-t')}\right) \int_0^{t'} dt'' \, e^{-(|g_{24}|^2/2 + i\Delta_b)(t'-t'')} \\
& \times \int_0^{t''} dt''' \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')}\right) \int d\omega_a' \int d\omega_b' \, f(\omega_a', \omega_b', t''') \\
& \times e^{i\omega_a(t-z'/c)} \, e^{i\omega_b(t'-z'/c)} \, e^{-i\omega_a'(t'''-z'/c)} \, e^{-i\omega_b'(t''-z'/c)}.
\end{aligned}
\tag{4.33}
$$

For computational convenience, we shall rewrite Eq. (4.33) in terms of k instead of ω. We shall assume that $\gamma \gg \Omega_c$ as a result of which we can approximate $\gamma^2 - \Omega_c^2 \approx \gamma^2$. Thus, the previous equation can be rewritten in terms of k under the assumption that we have just made, in the following form:

$$\frac{\partial}{\partial t} f(k_a, k_b, t) = \frac{|g_{13}|^2\ c}{2\pi\gamma}\ \int_{-L/2}^{L/2} dz'\ \rho(z')\ \int_0^t dt' \left(\Gamma_-\ e^{-\Gamma_-(t-t')} - \Gamma_+\ e^{-\Gamma_+(t-t')}\right)$$

$$\times \int dk'_a\ f(k'_a, k_b, t)\ e^{ik_a(ct-z')}\ e^{-ik'_a(ct'-z')} + \frac{|g_{13}|^2\ |g_{24}|^2\ \Omega_c^2\ c^2}{16\ \pi^2\ \gamma^2}\ \int_{-L/2}^{L/2} dz'\ \rho(z')$$

$$\times \int_0^t dt' \left(e^{-\Gamma_+(t-t')} - e^{-\Gamma_-(t-t')}\right) \int_0^{t'} dt''\ e^{-(|g_{24}|^2/2+i\Delta_b)(t'-t'')}$$

$$\times \int_0^{t''} dt''' \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')}\right) \int dk'_a \int dk'_b\ f(k'_a, k'_b, t''')$$

$$\times e^{ik_a(ct-z')}\ e^{ik_b(ct'-z')}\ e^{-ik'_a(ct'''-z')}\ e^{-ik'_b(ct''-z')}. \tag{4.34}$$

We shall define a "two-photon wavefunction" $\xi(z_a, z_b, t)$ as

$$\xi(z_a, z_b, t) \equiv \frac{1}{2\pi} \int dk_a \int dk_b\ e^{ik_a z_a}\ e^{ik_b z_b}\ f(k_a, k_b, t).$$

So,

$$\frac{\partial}{\partial t} \xi(z_a, z_b, t) = \frac{1}{2\pi} \int dk_a \int dk_b\ e^{ik_a z_a}\ e^{ik_b z_b}\ \frac{\partial}{\partial t} f(k_a, k_b, t). \tag{4.35}$$

On substituting Eq. (4.34) in Eq. (4.35), we get

$$
\frac{\partial}{\partial t}\xi(z_a, z_b, t) = \frac{|g_{13}|^2 c}{(2\pi)^2 \gamma} \int_{-L/2}^{L/2} dz' \, \rho(z') \int_0^t dt' \left(\Gamma_- \, e^{-\Gamma_-(t-t')} - \Gamma_+ \, e^{-\Gamma_+(t-t')} \right) \int dk_a \, e^{ik_a z_a}
$$

$$
\times \int dk_b \, e^{ik_b z_b} \int dk'_a \, f(k'_a, k_b, t) \, e^{ik_a(ct-z')} \, e^{-ik'_a(ct'-z')} + \frac{|g_{13}|^2 \, |g_{24}|^2 \, \Omega_c^2 \, c^2}{(2\pi)16 \, \pi^2 \, \gamma^2}
$$

$$
\times \int_{-L/2}^{L/2} dz' \, \rho(z') \int_0^t dt' \left(e^{-\Gamma_+(t-t')} - e^{-\Gamma_-(t-t')} \right) \int_0^{t'} dt'' \, e^{-(|g_{24}|^2/2 + i\Delta_b)(t'-t'')}
$$

$$
\times \int_0^{t''} dt''' \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')} \right) \int dk_a \, e^{ik_a z_a} \int dk_b \, e^{ik_b z_b}
$$

$$
\times \int dk'_a \int dk'_b \, f(k'_a, k'_b, t''') \, e^{ik_a(ct-z')} \, e^{ik_b(ct'-z')} \, e^{-ik'_a(ct'''-z')} \, e^{-ik'_b(ct''-z')}
$$

$$
= T_1 + T_2,
$$

$$(4.36)$$

where the "linear" term

$$
T_1 \equiv \frac{|g_{13}|^2 c}{(2\pi)^2 \gamma} \int_{-L/2}^{L/2} dz' \, \rho(z') \int_0^t dt' \left(\Gamma_- \, e^{-\Gamma_-(t-t')} - \Gamma_+ \, e^{-\Gamma_+(t-t')} \right)
$$

$$
\times \int dk_a \, e^{ik_a z_a} \int dk_b \, e^{ik_b z_b} \int dk'_a \, f(k'_a, k_b, t) \, e^{ik_a(ct-z')} \, e^{-ik'_a(ct'-z')},
$$

involves only the interaction of the field $\hat{A}$ with the EIT medium and the "nonlinear" term

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$$T_2 \equiv \frac{|g_{13}|^2 \ |g_{24}|^2 \ \Omega_c^2 \ c^2}{(2\pi)16 \ \pi^2 \ \gamma^2} \int_{-L/2}^{L/2} dz' \ \rho(z') \int_0^t dt' \left(e^{-\Gamma_+(t-t')} - e^{-\Gamma_-(t-t')} \right)$$

$$\times \int_0^{t'} dt'' \ e^{-(|g_{24}|^2/2+i\Delta_b)(t'-t'')} \int_0^{t''} dt''' \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')} \right)$$

$$\times \int dk_a \ e^{ik_a z_a} \int dk_b \ e^{ik_b z_b} \int dk_a' \int dk_b' \ f(k_a', k_b', t''') \ e^{ik_a(ct-z')} \ e^{ik_b(ct'-z')}$$

$$\times \ e^{-ik_a'(ct'''-z')} \ e^{-ik_b'(ct''-z')},$$

captures the interaction of the two fields $\hat{A}$ and $\hat{B}$. We shall first simplify T_1. Here, we shall assume that the pulse never leaves the medium. So, we shall let $L \to \infty$ and furthermore, we shall assume a constant atomic density ρ_0. Under these assumptions, we have

$$T_1 = \frac{|g_{13}|^2 c \ \rho_0}{(2\pi)^2 \gamma} \int_0^t dt' \left(\Gamma_- \ e^{-\Gamma_-(t-t')} - \Gamma_+ \ e^{-\Gamma_+(t-t')} \right) \int dk_a \ e^{ik_a z_a} \int dk_b \ e^{ik_b z_b}$$

$$\times \ e^{ik_a ct} \int dk_a' \ f(k_a', k_b, t') \ e^{-ik_a' ct'} \underbrace{\int_{-\infty}^{\infty} dz' \ e^{i(k_a'-k_a)z'}}_{2\pi\delta(k_a'-k_a)}. \tag{4.37}$$

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On enforcing the δ function in the previous equation, we obtain

$$
T_1 = \frac{|g_{13}|^2 \; c \; \rho_0}{\gamma} \int_0^t dt' \left(\Gamma_- \; e^{-\Gamma_-(t-t')} - \Gamma_+ \; e^{-\Gamma_+(t-t')} \right)
$$

$$
\times \; \underbrace{\frac{1}{2\pi} \int dk_a \; e^{ik_a[z_a+c(t-t')]} \int dk_b \; e^{ik_b z_b} \; f(k_a, k_b, t')}_{\xi(z_a+c(t-t'), z_b, t')}
$$

$$
= \frac{|g_{13}|^2 \; c \; \rho_0}{\gamma} \int_0^t dt' \left(\Gamma_- \; e^{-\Gamma_-(t-t')} - \Gamma_+ \; e^{-\Gamma_+(t-t')} \right) \; \xi(z_a + c(t-t'), z_b, t'). \quad (4.38)
$$

Next, we shall expand the terms in the parenthesis, in the previous equation, which then gives us

$$
T_1 = \frac{|g_{13}|^2 \; c \; \rho_0}{\gamma} \; \Gamma_- \; e^{-\Gamma_- t} \int_{-\infty}^t dt' \; e^{\Gamma_- t'} \; \xi(z_a + c(t-t'), z_b, t')
$$

$$
- \frac{|g_{13}|^2 \; c \; \rho_0}{\gamma} \; \Gamma_+ \; e^{-\Gamma_+ t} \int_{-\infty}^t dt' \; e^{\Gamma_+ t'} \; \xi(z_a + c(t-t'), z_b, t').
$$

$$
(4.39)
$$

We shall evaluate both the integrals in the previous equation by parts and furthermore, impose the condition that ξ vanishes as $t \to -\infty$. This yields

$$
T_1 = \frac{|g_{13}|^2 \; c \; \rho_0}{\gamma} \int_0^t dt' \left(e^{-\Gamma_+(t-t')} - e^{-\Gamma_-(t-t')} \right) \left(\frac{\partial \xi}{\partial t'} - c \, \frac{\partial \xi}{\partial z} \right), \quad (4.40)
$$

where we have defined $z_a + c\,(t - t') \equiv z$. In order to simplify the mathematical analysis, we shall make an approximation here. Since $\Gamma_+ \gg \Gamma_-$, $e^{-\Gamma_+(t-t')} \ll e^{-\Gamma_-(t-t')}$.

This approximation will be good except for very short times around $t = t'$, of the order of $|t - t'| \sim 1/\Gamma_+$. So, we shall retain only the exponential term involving Γ_- in the previous equation which then reduces Eq. (4.40) to

$$T_1 = -\frac{|g_{13}|^2 \, c \, \rho_0}{\gamma} \int_{-\infty}^{t} dt' \, e^{-\Gamma_-(t-t')} \left(\frac{\partial \xi}{\partial t'} - c \, \frac{\partial \xi}{\partial z} \right). \tag{4.41}$$

Now, we shall make a plain adiabatic approximation to the evaluation of the integral in Eq. (4.41). We will assume that the terms in the parenthesis in Eq. (4.41) vary much more slowly than $\exp(-\Gamma_- t')$ in t' as a result of which we can evaluate $\partial \xi / \partial t' - c \, \partial \xi / \partial z$ at $t' = t$, and pull them outside the integral.

Thus,

$$\left(\frac{\partial \xi}{\partial t'} - c \, \frac{\partial \xi}{\partial z} \right)_{t'=t} \rightarrow \frac{\partial \xi}{\partial t} - c \, \frac{\partial \xi}{\partial z_a}.$$

This makes

$$T_1 = -\frac{|g_{13}|^2 \, c \, \rho_0}{\gamma} \left(\frac{\partial \xi}{\partial t} - c \, \frac{\partial \xi}{\partial z_a} \right) \underbrace{\int_{-\infty}^{t} dt' \, e^{-\Gamma_-(t-t')}}_{1/\Gamma_-}$$

$$= -\frac{|g_{13}|^2 \, c \, \rho_0}{\gamma \, \Gamma_-} \left(\frac{\partial}{\partial t} - c \, \frac{\partial}{\partial z_a} \right) \xi(z_a, z_b, t). \tag{4.42}$$

Next, we shall simplify T_2 under the same assumptions which we imposed on T_1.

$$T_2 = \frac{|g_{13}|^2 \ |g_{24}|^2 \ \Omega_c^2 \ c^2 \ \rho_0}{16 \ \pi^2 \ \gamma^2} \int_0^t dt' \left(e^{-\Gamma_+(t-t')} - e^{-\Gamma_-(t-t')}\right)$$

$$\times \int_0^{t'} dt'' \ e^{-(|g_{24}|^2/2+i\Delta_b)(t'-t'')} \int_0^{t''} dt''' \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')}\right)$$

$$\times \int_{-\infty}^\infty dz' \underbrace{\int dk_a \ e^{ik_a(z_a-z'+ct)}}_{2\pi \ \delta(z_a-z'+ct)} \underbrace{\int dk_b \ e^{ik_b(z_b-z'+ct')}}_{2\pi \ \delta(z_b-z'+ct')}$$

$$\times \left(\frac{1}{2\pi}\right) \underbrace{\int dk_a' \int dk_b' \ e^{ik_a'(z'-ct''')} \ e^{ik_b'(z'-ct'')} \ f(k_a', k_b', t''')}_{\xi(z'-ct''',z'-ct'',t''')}, \tag{4.43}$$

which gives us

$$T_2 = \frac{|g_{13}|^2 \ |g_{24}|^2 \ \Omega_c^2 \ c^2 \ \rho_0}{4 \ \gamma^2} \int_0^t dt' \left(e^{-\Gamma_+(t-t')} - e^{-\Gamma_-(t-t')}\right)$$

$$\times \int_0^{t'} dt'' \ e^{-(|g_{24}|^2/2+i\Delta_b)(t'-t'')} \int_0^{t''} dt''' \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')}\right)$$

$$\times \int_{-\infty}^\infty dz' \ \delta(z_a - z' + ct) \ \delta(z_b - z' + ct') \ \xi(z' - ct''', z' - ct'', t'''). \tag{4.44}$$

In the previous equation, we shall first enforce the δ function on the left, i.e.
$\delta(z_a - z' + ct)$ to get rid of the integral over z' which makes $z' = z_a + ct$.

We thus have,

$$T_2 = \frac{|g_{13}|^2 \; |g_{24}|^2 \; \Omega_c^2 \; c^2 \; \rho_0}{4 \; \gamma^2} \int_0^t dt' \left(e^{-\Gamma_+(t-t')} - e^{-\Gamma_-(t-t')} \right)$$

$$\times \int_0^{t'} dt'' \; e^{-(|g_{24}|^2/2+i\Delta_b)(t'-t'')} \int_0^{t''} dt''' \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')} \right)$$

$$\times \xi[z_a + c(t - t'''), z_a + c(t - t''), t'''] \; \delta[z_b - z_a - c(t - t')]. \qquad (4.45)$$

We shall now enforce the δ function in the previous equation and get rid of the integral over t'. This makes $t' = t - [(z_b - z_a)/c]$. It is important to note that the integral over t' goes from 0 to t. So $t' < t$ means that $z_b - z_a$ must be positive. In order to incorporate this requirement, we shall introduce a step function $\Theta(z_b - z_a)$ in the expression for T_2.

We thus obtain,

$$T_2 = \frac{|g_{13}|^2 \; |g_{24}|^2 \; \Omega_c^2 \; c \; \rho_0}{4 \; \gamma^2} \Theta(z_b - z_a) \left(e^{-\Gamma_+(z_b-z_a)/c} - e^{-\Gamma_-(z_b-z_a)/c} \right)$$

$$\times \int_0^{t-[(z_b-z_a)/c]} dt'' \; e^{-(|g_{24}|^2/2+i\Delta_b)(t-t''-[(z_b-z_a)/c])}$$

$$\times \int_0^{t''} dt''' \left(e^{-\Gamma_-(t''-t''')} - e^{-\Gamma_+(t''-t''')} \right) \xi[z_a + c(t - t'''), z_a + c(t - t''), t''']. \quad (4.46)$$

From the expression for T_2 in the previous equation, it is quite clear that the analytical calculation for this problem in its current form might be intractable. In order to make the problem little simpler, we make an approximation for ξ. We shall assume that ξ varies slowly such that we will evaluate it at $t'' = t''' = t - [(z_b - z_a)/c]$. This is again essentially

an adiabatic approximation to the integrals over t'' and t'''.

We thus have,

$$\xi[z_a + c(t - t'''), z_a + c(t - t''), t''']_{t''=t'''=t-[(z_b-z_a)/c]} = \xi(z_b, z_b, t - [(z_b - z_a)/c]).$$

This assumption simplifies T_2 to the following form:

$$T_2 = \frac{|g_{13}|^2 \, |g_{24}|^2 \, \Omega_c^2 \, c \, \rho_0}{4 \, \gamma^2} \, \Theta(z_b - z_a) \, \left(e^{-\Gamma_+(z_b-z_a)/c} - e^{-\Gamma_-(z_b-z_a)/c}\right)$$

$$\times \, \xi(z_b, z_b, t - [(z_b - z_a)/c]) \, e^{-(|g_{24}|^2/2+i\Delta_b)(t-[(z_b-z_a)/c])}$$

$$\times \, \underbrace{\int_{-\infty}^{t-[(z_b-z_a)/c]} dt'' \, e^{(|g_{24}|^2/2+i\Delta_b)t''}}_{I'}$$

$$\times \, \left(\underbrace{\int_{-\infty}^{t''} dt''' \, e^{-\Gamma_-(t''-t''')}}_{1/\Gamma_-} - \underbrace{\int_{-\infty}^{t''} dt''' \, e^{-\Gamma_+(t''-t''')}}_{1/\Gamma_+}\right), \tag{4.47}$$

where

$$I' = \frac{e^{(|g_{24}|^2/2+i\Delta_b)(t-[(z_b-z_a)/c])}}{|g_{24}|^2/2 + i\Delta_b}.$$

Note that in the previous equation, we have extended the lower limits of all the integrals to $-\infty$ since this would not change anything. This is because the pulse starts at $t = 0$. This means that we can safely assume that at $t = 0$, it is negligible. So, this implies that the pulse is negligible for $t < 0$ which would then allow us to harmlessly extend the lower limit

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of all the integrals over time to $-\infty$. Eq. (4.47) now reduces to a more compact form:

$$T_2 = \frac{|g_{13}|^2 \; |g_{24}|^2 \; \Omega_c^2 \; c \; \rho_0}{4 \; \gamma^2} \; \Theta(z_b - z_a) \; \left(e^{-\Gamma_+(z_b - z_a)/c} - e^{-\Gamma_-(z_b - z_a)/c}\right)$$

$$\times \left(\frac{1}{\Gamma_-} - \frac{1}{\Gamma_+}\right) \; \frac{1}{|g_{24}|^2/2 + i\Delta_b} \; \xi(z_b, z_b, t - [(z_b - z_a)/c]). \tag{4.48}$$

We shall further assume that $1/\Gamma_- \gg 1/\Gamma_+$. Therefore, $1/\Gamma_- - 1/\Gamma_+ \approx 1/\Gamma_-$.

Moreover, we shall assume that $\Delta_b \gg |g_{24}|^2$ which makes $|g_{24}|^2/2 + i\Delta_b \approx i\Delta_b$. These

additional assumptions simplify T_2 further which finally results in

$$T_2 = -i\frac{|g_{13}|^2 \; |g_{24}|^2 \; \Omega_c^2 \; c \; \rho_0}{4 \; \gamma^2 \; \Gamma_- \; \Delta_b} \; \Theta(z_b - z_a) \; \left(e^{-\Gamma_+(z_b - z_a)/c} - e^{-\Gamma_-(z_b - z_a)/c}\right)$$

$$\times \; \xi(z_b, z_b, t - [(z_b - z_a)/c]). \tag{4.49}$$

On substituting Eqs. (4.42) and (4.49) in Eq. (4.36), we obtain

$$\frac{\partial}{\partial t}\xi(z_a, z_b, t) = -\frac{|g_{13}|^2 \; c \; \rho_0}{\gamma \; \Gamma_-} \frac{\partial}{\partial t}\xi(z_a, z_b, t) + \frac{|g_{13}|^2 \; c^2 \; \rho_0}{\gamma \; \Gamma_-} \frac{\partial}{\partial z_a}\xi(z_a, z_b, t)$$

$$-i\frac{|g_{13}|^2 \; |g_{24}|^2 \; \Omega_c^2 \; c \; \rho_0}{4 \; \gamma^2 \; \Gamma_- \; \Delta_b} \; \Theta(z_b - z_a) \; \left(e^{-\Gamma_+(z_b - z_a)/c} - e^{-\Gamma_-(z_b - z_a)/c}\right)$$

$$\times \; \xi(z_b, z_b, t - [(z_b - z_a)/c]). \tag{4.50}$$

On moving the first term on the right hand side of Eq. (4.50) to the left and defining

the following parameters:

$$\frac{|g_{13}|^2 \, c \, \rho_0}{\gamma \, \Gamma_-} \equiv A',$$

$$\frac{|g_{13}|^2 \, |g_{24}|^2 \, \Omega_c^2 \, c \, \rho_0}{4 \, \gamma^2 \, \Gamma_- \, \Delta_b} \equiv B',$$

and

$$c \left(\frac{A'}{1 + A'} \right) \equiv v,$$

we can rewrite Eq. (4.50) in a more compact form that reads

$$\left(\frac{\partial}{\partial t} - v \frac{\partial}{\partial z_a} \right) \xi(z_a, z_b, t) = -i \left(\frac{B'}{1 + A'} \right) \Theta(z_b - z_a) \left(e^{-\Gamma_+ (z_b - z_a)/c} - e^{-\Gamma_- (z_b - z_a)/c} \right)$$

$$\times \xi(z_b, z_b, t - [(z_b - z_a)/c]). \tag{4.51}$$

Eq. (4.51) is the final simplified dynamical equation that we need to solve to obtain the

final state of the outgoing photons.

We note that the right-hand side of the previous equation vanishes for $z_b < z_a$. This

makes $\Theta(z_b - z_a) = 0$. So, we start by solving Eq. (4.51) in that region where it reduces to

$$\left(\frac{\partial}{\partial} - v \frac{\partial}{\partial z_a} \right) \xi(z_a, z_b, t) = 0. \tag{4.52}$$

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We will then evaluate the result on the line $z_a = z_b$ and use that on the right-hand side of Eq. (4.51) for $z_a > z_b$. In this way, an exact solution to Eq. (4.51) is possible.

We shall make the following co-ordinate transformation: $t' = t$ and $z' = z_a + vt$. We thus have,

$$\frac{\partial}{\partial t} - v \frac{\partial}{\partial z_a} = \frac{\partial}{\partial t'},$$

which then transforms Eq. (4.52) to

$$\frac{\partial}{\partial t'} \xi(z', z_b, t') = 0, \tag{4.53}$$

whose solution can be written as

$$\xi(z', z_b, t') = \eta(z', z_b), \tag{4.54}$$

which is a constant in t'. In terms of the original variables, we have when $z_b < z_a$,

$$\xi(z_a, z_b, t) = \eta(z_a + vt, z_b). \tag{4.55}$$

It is straightforward to verify that $\eta(z_a + vt, z_b)$ indeed satisfies Eq. (4.52). When $z_b < z_a$ and at $t = 0$, $\xi(z_a, z_b, 0) = \eta(z_a, z_b)$.

The fact that there is no derivative with respect to z_b on Eq. (4.51) means that we are working in a reference frame where the b photon is at rest. We shall assume that the center of the wavepacket corresponding to the b photon is at $z_b = 0$. At $t = 0$, the center of the wavepacket corresponding to the a photon is at $z_a = z_0$ and it starts moving to the left (toward the b photon) as time evolves. Although, in principle, it is possible to write down the formal solution of Eq. (4.51) for an arbitrary initial wavepacket, for definiteness in what follows we will assume that the initial state is a Gaussian pulse.

Note that

$$\xi(z_a, z_b, 0) = \frac{1}{2\pi} \int dk_a \ e^{ik_a z_a} \int dk_b \ e^{ik_b z_b} \ f(k_a, k_b, 0).$$

If we assume our initial state to be a Gaussian, then

$$\xi(z_a, z_b, 0) = \frac{1}{\sigma\sqrt{\pi}} \ e^{-(z_a-z_0)^2/2\sigma^2} \ e^{-z_b^2/2\sigma^2}.$$

When $z_b < z_a$, we know from Eq. (4.55) that

$$\xi(z_a, z_b, t) = \frac{1}{\sigma\sqrt{\pi}} \ e^{-(z_a+vt-z_0)^2/2\sigma^2} \ e^{-z_b^2/2\sigma^2}.$$

When $z_b \geq z_a$, the time evolved state $\xi[z_b, z_b, t - ((z_b - z_a)/c)]$ for a Gaussian pulse can be written as

$$\xi(z_b, z_b, t - ((z_b - z_a)/c)) = \frac{1}{\sigma\sqrt{\pi}} \ e^{-[z_b-z_0+vt-v/c(z_b-z_a)]^2/2\sigma^2} \ e^{-z_b^2/2\sigma^2}. \tag{4.56}$$

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On substituting Eq. (4.56) in Eq. (4.51), we get

$$\left(\frac{\partial}{\partial} - v\frac{\partial}{\partial z_a}\right)\xi(z_a, z_b, t) = -\frac{i}{\sigma\sqrt{\pi}}\left(\frac{B'}{1 + A'}\right)\Theta(z_b - z_a)\left(e^{-\Gamma_+(z_b - z_a)/c} - e^{-\Gamma_-(z_b - z_a)/c}\right)$$

$$\times \, e^{-[z_b - z_0 + vt - v/c(z_b - z_a)]^2/2\sigma^2} \, e^{-z_b^2/2\sigma^2}.$$

(4.57)

We shall once again make the same co-ordinate transformation that we made earlier, i.e. $t' = t$ and $z' = z_a + vt$, which would then transform Eq. (4.57) to the following form:

$$\frac{\partial}{\partial t'}\xi(z', z_b, t') = \frac{-i}{\sigma\sqrt{\pi}}\left(\frac{B'}{1 + A'}\right)\Theta(z_b - z' + vt')\left(e^{-\Gamma_+(z_b - z')/c}\,e^{-\beta\,\Gamma_+ t'} - e^{-\Gamma_-(z_b - z')/c}\,e^{-\beta\,\Gamma_- t'}\right)$$

$$\times \, e^{-z_b^2/2\sigma^2} \, e^{-[\alpha\,z_b - z_0 + \beta\,z' + \alpha\,v\,t']^2/2\sigma^2},$$

(4.58)

where we have defined $1 - v/c \equiv \alpha$ and $v/c \equiv \beta$.

The step function in Eq. (4.58) reads

$$\Theta(z_b - z' + vt') = \begin{cases} 1, & \text{if } z_b - z' + vt' \geq 0 \\ \\ 0, & \text{otherwise.} \end{cases}$$

(4.59)

This sets the lower limit for t', i.e. $t' \geq (z' - z_b)/v$. On incorporating this lower bound for t' in Eq. (4.58) and formally integrating, we obtain

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$$\xi(z', z_b, t') - \xi(z', z_b, 0) = -\frac{i}{\sigma\sqrt{\pi}} \left(\frac{B'}{1 + A'}\right) e^{-z_b^2/2\sigma^2} \left(e^{-\Gamma_+(z_b - z')/c}\right.$$

$$\times \underbrace{\int_{(z'-z_b)/v}^{t'} dt'' \; e^{-\beta \; \Gamma_+ t''} \; e^{-[\alpha \; z_b - z_0 + \beta \; z' + \alpha \; v \; t'']^2/2\sigma^2}}_{I_I}$$

$$- e^{-\Gamma_-(z_b - z')/c} \underbrace{\int_{(z'-z_b)/v}^{t'} dt'' \; e^{-\beta \; \Gamma_- t''} \; e^{-[\alpha \; z_b - z_0 + \beta \; z' + \alpha \; v \; t'']^2/2\sigma^2}}_{I_{II}} \right),$$

$$(4.60)$$

where

$$I_I = \sqrt{\frac{\pi}{2}} \frac{\sigma}{\alpha v} e^{-\beta\Gamma_+(-\alpha \; z_b + z_0 - \beta \; z')/\alpha v} e^{\beta^2\Gamma_+^2\sigma^2/2\alpha^2 v^2}$$

$$\times \left(\mathrm{erf}\left[\frac{\alpha \; z_b - z_0 + \beta \; z' + \alpha \; v \; t'}{\sqrt{2} \; \sigma} + \frac{\beta \; \Gamma_+ \; \sigma}{\sqrt{2} \; \alpha \; v}\right]\right.$$

$$\left. - \mathrm{erf}\left[\frac{z' - z_0}{\sqrt{2}\sigma} + \frac{\beta \; \Gamma_+ \; \sigma}{\sqrt{2} \; \alpha \; v}\right]\right),$$

$$(4.61)$$

and

$$I_{II} = \sqrt{\frac{\pi}{2}} \frac{\sigma}{\alpha v} e^{-\beta\Gamma_-(-\alpha \; z_b + z_0 - \beta \; z')/\alpha v} e^{\beta^2\Gamma_-^2\sigma^2/2\alpha^2 v^2}$$

$$\times \left(\mathrm{erf}\left[\frac{\alpha \; z_b - z_0 + \beta \; z' + \alpha \; v \; t'}{\sqrt{2} \; \sigma} + \frac{\beta \; \Gamma_- \; \sigma}{\sqrt{2} \; \alpha \; v}\right]\right.$$

$$\left. - \mathrm{erf}\left[\frac{z' - z_0}{\sqrt{2}\sigma} + \frac{\beta \; \Gamma_- \; \sigma}{\sqrt{2} \; \alpha \; v}\right]\right).$$

$$(4.62)$$

Next, we shall substitute Eqs. (4.61) and (4.62) in Eq. (4.60). Furthermore, we will

express the final state in terms of the original coordinates (t, z_a), and we shall explicitly

substitute for the functional form of a Gaussian pulse for the initial state

$\xi(z_a + vt, z_b, 0) = (1/(\sigma\sqrt{\pi})) \exp[-(z_a + vt - z_0)^2/2\sigma^2] \exp[-z_b^2/2\sigma^2]$ in Eq. (4.60). This

yields

$$\xi(z_a, z_b, t) = \frac{1}{\sigma\sqrt{\pi}} e^{-(z_a+vt-z_0)^2/2\sigma^2} e^{-z_b^2/2\sigma^2} - \frac{i}{\sqrt{2}\,\alpha\,v} \left(\frac{B'}{1+A'}\right) e^{-z_b^2/2\sigma^2}$$
$$\times \left[e^{-\Gamma_+(z_b-z_a-vt)/c} e^{-\beta\Gamma_+[-\alpha z_b+z_0-\beta(z_a+vt)]/\alpha v} e^{\beta^2\Gamma_+^2\sigma^2/2\alpha^2 v^2} \right.$$
$$\times (S_1 - S_2) - e^{-\Gamma_-(z_b-z_a-vt)/c} e^{-\beta\Gamma_-[-\alpha z_b+z_0-\beta(z_a+vt)]/\alpha v}$$
$$\left. \times e^{\beta^2\Gamma_-^2\sigma^2/2\alpha^2 v^2} (S_3 - S_4) \right], \tag{4.63}$$

where

$$S_1 \equiv \mathrm{erf}\left[\frac{\alpha z_b - z_0 + \beta z_a + vt}{\sqrt{2}\sigma} + \frac{\beta\Gamma_+\sigma}{\sqrt{2}\alpha v}\right], \tag{4.64}$$

$$S_2 \equiv \mathrm{erf}\left[\frac{z_a + vt - z_0}{\sqrt{2}\sigma} + \frac{\beta\Gamma_+\sigma}{\sqrt{2}\alpha v}\right], \tag{4.65}$$

$$S_3 \equiv \mathrm{erf}\left[\frac{\alpha z_b - z_0 + \beta z_a + vt}{\sqrt{2}\sigma} + \frac{\beta\Gamma_-\sigma}{\sqrt{2}\alpha v}\right], \tag{4.66}$$

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and

$$S_4 \equiv \mathrm{erf}\left[\frac{z_a + vt - z_0}{\sqrt{2}\sigma} + \frac{\beta\Gamma_-\sigma}{\sqrt{2}\alpha v}\right]. \tag{4.67}$$

It is evident that the expression for the final state given by Eqs. (4.63) through (4.67) is quite complicated. Hence, it would be helpful if we make some approximations here to reasonably simplify the final state. It is important to keep in mind that we are only interested in the state long after the interaction is over, i.e. we are concerned only with the limit $t \to \infty$. Furthermore, in our analysis $\beta > \alpha$. This is because in the EIT, the a photon travels with a velocity v_g ($v_g < c$) and the b photon travels at c. However, we are implicitly in a reference frame as seen in Eq. (4.51) where the b photon is at rest. In this frame, the a photon travels toward the b photon with a velocity $c - v_g \equiv v$. This means $1 - v_g/c = v/c$. Thus, $\alpha \equiv 1 - v/c = v_g/c$ and $\beta \equiv v/c = 1 - v_g/c$. We assume in our calculation that v_g/c is in the range of 0.05 to 0.1. This makes $\beta > \alpha$. Thus, in the long time limit and given the fact that $\beta > \alpha$, we shall approximate

$$S_1 \equiv \mathrm{erf}\left[\frac{\alpha z_b - z_0 + \beta z_a + vt}{\sqrt{2}\sigma} + \frac{\beta\Gamma_+\sigma}{\sqrt{2}\alpha v}\right] \approx 1,$$

and

$$S_3 \equiv \mathrm{erf}\left[\frac{\alpha z_b - z_0 + \beta z_a + vt}{\sqrt{2}\sigma} + \frac{\beta\Gamma_-\sigma}{\sqrt{2}\alpha v}\right] \approx 1,$$

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which then simplifies Eq. (4.63) to the following form:

$$
\begin{aligned}
\xi(z_a, z_b, t) = {} & \frac{1}{\sigma\sqrt{\pi}} \, e^{-(z_a + vt - z_0)^2/2\sigma^2} \, e^{-z_b^2/2\sigma^2} - \frac{i}{\sqrt{2}\,\alpha\,v} \left(\frac{B'}{1 + A'} \right) e^{-z_b^2/2\sigma^2} \\
& \times \left[e^{-\Gamma_+(z_b - z_a - vt)/c} \, e^{-\beta\Gamma_+[-\alpha z_b + z_0 - \beta(z_a + vt)]/\alpha v} \, e^{\beta^2\Gamma_+^2\sigma^2/2\alpha^2 v^2} \right. \\
& \times \operatorname{erfc}\left(\frac{z_a + vt - z_0}{\sqrt{2}\sigma} + \frac{\beta\Gamma_+\sigma}{\sqrt{2}\alpha v} \right) - e^{-\Gamma_-(z_b - z_a - vt)/c} \, e^{-\beta\Gamma_-[-\alpha z_b + z_0 - \beta(z_a + vt)]/\alpha v} \\
& \left. \times e^{\beta^2\Gamma_-^2\sigma^2/(2\alpha^2 v^2)} \, \operatorname{erfc}\left(\frac{z_a + vt - z_0}{\sqrt{2}\sigma} + \frac{\beta\Gamma_-\sigma}{\sqrt{2}\alpha v} \right) \right].
\end{aligned}
$$

$$(4.68)$$

Our next task is to simplify Eq. (4.68). In order to save space and make the simplification easier to follow, we shall split the second term on the right hand side of the previous equation into two parts, viz.

$$
\Lambda_1 \equiv e^{-\Gamma_+(z_b - z_a - vt)/c} \, e^{-\beta\Gamma_+[-\alpha z_b + z_0 - \beta(z_a + vt)]/\alpha v} \, e^{\beta^2\Gamma_+^2\sigma^2/2\alpha^2 v^2} \, \operatorname{erfc}\left(\frac{z_a + vt - z_0}{\sqrt{2}\sigma} + \frac{\beta\Gamma_+\sigma}{\sqrt{2}\alpha v} \right),
$$

and

$$
\Lambda_2 \equiv e^{-\Gamma_-(z_b - z_a - vt)/c} \, e^{-\beta\Gamma_-[-\alpha z_b + z_0 - \beta(z_a + vt)]/\alpha v} \, e^{\beta^2\Gamma_-^2\sigma^2/(2\alpha^2 v^2)} \, \operatorname{erfc}\left(\frac{z_a + vt - z_0}{\sqrt{2}\sigma} + \frac{\beta\Gamma_-\sigma}{\sqrt{2}\alpha v} \right).
$$

We shall simplify Λ_1 and Λ_2 separately and to accomplish this, we shall make use of the following asymptotic expansion [39]:

$$e^{z^2} \operatorname{erfc}(z) \sim \frac{1}{\sqrt{\pi}z} + \frac{1}{\sqrt{\pi}z} \sum_{m=1}^{\infty} (-1)^m \frac{1.3...(2m-1)}{(2z^2)^m}, \quad z \to \infty.$$

For our case, we are only going to retain the first term in the asymptotic expansion. So, we simply have

$$e^{z^2} \operatorname{erfc}(z) \sim \frac{1}{\sqrt{\pi}z}. \tag{4.69}$$

Note that we are working in the long-time limit, i.e. as $t \to \infty$. Thus, in this limit, the argument of the complimentary error function in both Λ_1 and Λ_2 becomes very large, i.e. $(z_a + vt - z_0)/\sqrt{2}\sigma + \beta\Gamma_{\pm}\sigma/\sqrt{2}\alpha v \to \infty$. Hence, we can legitimately exploit the approximation in Eq. (4.69) to simplify Λ_1 and Λ_2.

We shall start with Λ_1.

$$\Lambda_1 \equiv e^{-\Gamma_+(z_b-z_a-vt)/c} \; e^{-\beta\Gamma_+[-\alpha z_b+z_0-\beta(z_a+vt)]/\alpha v} \; e^{\beta^2\Gamma_+^2\sigma^2/2\alpha^2 v^2} \; \text{erfc}\left(\frac{z_a+vt-z_0}{\sqrt{2}\sigma} + \frac{\beta\Gamma_+\sigma}{\sqrt{2}\alpha v}\right)$$

$$= e^{-\Gamma_+(z_b-z_a-vt)/c} \; e^{-(z_a+vt-z_0)^2/2\sigma^2} \; \underbrace{e^{-\beta\Gamma_+[-\alpha z_b+z_0-\beta(z_a+vt)]/\alpha v} \; e^{-\beta\Gamma_+(z_a+vt-z_0)/\alpha v}}_{\exp[\Gamma_+(z_b-z_a-vt)/c]}$$

$$\times \underbrace{e^{(z_a+vt-z_0)^2/2\sigma^2} \; e^{\beta\Gamma_+(z_a+vt-z_0)/\alpha v} \; e^{\beta^2\Gamma_+^2\sigma^2/2\alpha^2 v^2}}_{T'}$$

$$\times \text{erfc}\left(\frac{z_a+vt-z_0}{\sqrt{2}\sigma} + \frac{\beta\Gamma_+\sigma}{\sqrt{2}\alpha v}\right),$$

$$(4.70)$$

where

$$T' = \exp\left[\left(\frac{z_a+vt-z_0}{\sqrt{2}\sigma} + \frac{\beta\Gamma_+\sigma}{\sqrt{2}\alpha v}\right)^2\right].$$

In the previous equation, we can see that $\exp[-\Gamma_+(z_b - z_a - vt)/c]$ cancels with its inverse and on explicitly substituting for T' in Eq. (4.70) and making the asymptotic expansion in Eq. (4.69), we obtain

$$\Lambda_1 = \frac{1}{\sqrt{\pi}} \frac{e^{-(z_a+vt-z_0)^2/2\sigma^2}}{[(z_a+vt-z_0)/\sqrt{2}\sigma] + [(\beta\Gamma_+\sigma)/\sqrt{2}\alpha v]}. \qquad (4.71)$$

Similarly, we can simplify Λ_2 along the same line which would then yield

$$\Lambda_2 = \frac{1}{\sqrt{\pi}} \frac{e^{-(z_a+vt-z_0)^2/2\sigma^2}}{[(z_a+vt-z_0)/\sqrt{2}\sigma] + [(\beta\Gamma_-\sigma)/\sqrt{2}\alpha v]}. \qquad (4.72)$$

On substituting Eqs. (4.71) and (4.72) in Eq. (4.68), we obtain

$$
\xi(z_a, z_b, t) = \frac{1}{\sigma\sqrt{\pi}} \, e^{-(z_a + vt - z_0)^2/2\sigma^2} \, e^{-z_b^2/2\sigma^2} \left[1 - i \left(\frac{\sigma}{\alpha v} \right) \left(\frac{B'}{1 + A'} \right) \right.
$$
$$
\left. \times \left(\frac{1}{[(z_a + vt - z_0)/\sigma] + [(\beta\Gamma_+ \sigma)/\alpha v]} - \frac{1}{[(z_a + vt - z_0)/\sigma] + [(\beta\Gamma_- \sigma)/\alpha v]} \right) \right]. \quad (4.73)
$$

In the reference frame in which we are working here, the b photon is at rest. The a photon is traveling toward the b photon with a velocity v. Initially, the center of the wavepacket of the a photon is at $z_a = z_0$, far away from the b photon. We shall assume that the wavepacket of the b photon is centered at $z_b = 0$ and it stays there. In this frame of reference, the wavepacket of the a photon sweeps across the wavepacket of the b photon and after the interaction, the a pulse moves away from the b pulse.

In the long-time limit, i.e. for very large values of t, the center of the wavepacket of the a photon would be far away from the b photon. So, we can approximate $z_a + vt \approx z_0$ in the denominator of Eq. (4.73), which would then simplify this equation to

$$
\xi(z_a, z_b, t) = \frac{1}{\sigma\sqrt{\pi}} \, e^{-(z_a + vt - z_0)^2/2\sigma^2} \, e^{-z_b^2/2\sigma^2} \left[1 - \frac{i}{\beta} \left(\frac{B'}{1 + A'} \right) \left(\frac{1}{\Gamma_+} - \frac{1}{\Gamma_-} \right) \right]. \quad (4.74)
$$

Let us recall that we had earlier assumed that $\Gamma_+ \gg \Gamma_-$ as a result of which we can approximate $1/\Gamma_+ - 1/\Gamma_- \approx -1/\Gamma_-$, which would then further simplify Eq. (4.74) to

$$\xi(z_a, z_b, t_{final}) = \underbrace{\frac{1}{\sigma\sqrt{\pi}} \, e^{-(z_a+vt-z_0)^2/2\sigma^2} \, e^{-z_b^2/2\sigma^2}}_{\xi(z_a,z_b,t_{initial})} \left[1 + \frac{i}{\beta}\left(\frac{B'}{1+A'}\right)\left(\frac{1}{\Gamma_-}\right)\right]. \tag{4.75}$$

Note that

$$\Gamma_- = \frac{\gamma}{2}\left[1 - \left(1 - \frac{\Omega_c^2}{\gamma^2}\right)^{1/2}\right].$$

When $\gamma \gg \Omega_c$, we can expand $(1 - \Omega_c^2/\gamma^2)^{1/2}$ and retain only the term first-order in $(\Omega_c/\gamma)^2$ which would then yield

$$\Gamma_- \simeq \frac{\gamma}{2}\left[1 - \left(1 - \frac{1}{2}\frac{\Omega_c^2}{\gamma^2}\right)\right] = \frac{\Omega_c^2}{4\gamma}.$$

Earlier we had set

$$c\left(\frac{A'}{1+A'}\right) = v,$$

from which we obtain

$$A' = \frac{v/c}{1 - v/c} = \frac{\beta}{\alpha}. \tag{4.76}$$

We shall once again recall the definitions of A' and B' for convenience.

$$A' \equiv \frac{|g_{13}|^2 \, c \, \rho_0}{\gamma \, \Gamma_-},$$

and

$$B' \equiv \frac{|g_{13}|^2 \ |g_{24}|^2 \ \Omega_c^2 \ c \ \rho_0}{4 \ \gamma^2 \ \Delta_b \ \Gamma_-}.$$

We shall now try to express B' in terms of the other parameters defined earlier.

$$B' = \frac{|g_{24}|^2}{\Delta_b} \ \underbrace{\frac{\Omega_c^2}{4 \ \gamma}}_{\Gamma_-} \ \underbrace{\left(\frac{|g_{13}|^2 \ c \ \rho_0}{\gamma \ \Gamma_-}\right)}_{A'= \ \beta/\alpha}$$

$$= \frac{|g_{24}|^2}{\Delta_b} \ \Gamma_- \ \frac{\beta}{\alpha}. \tag{4.77}$$

On substituting Eqs. (4.76) and (4.77) in Eq. (4.75) and carrying out some trivial simplifications, we get

$$\xi(z_a, z_b, t_{final}) = \xi(z_a, z_b, t_{initial}) \ \left(1 + i \ \frac{|g_{24}|^2}{\Delta_b}\right). \tag{4.78}$$

We have already assumed that $\Delta_b \gg |g_{24}|^2$, i.e. we have assumed the detuning to be very large so as to prevent the b photon from getting absorbed in the atomic medium. This makes $|g_{24}|^2/\Delta_b \ll 1$. To the same order of validity as of Eq. (4.78), then, we can write

$$\xi(z_a, z_b, t_{final}) = \xi(z_a, z_b, t_{initial}) \ e^{i|g_{24}|^2/\Delta_b}, \tag{4.79}$$

as long as we remember that the argument of the exponential (the phase shift) must be

small. We can thus infer from Eq. (4.79) that in the long-time limit and on the condition that the detuning from level 4 (Δ_b) is much larger compared to the coupling strength between levels 2 and 4 (g_{24}), the final state ends up being the same as the initial state with only a small phase shift. One of the reasons for the smallness of this phase shift is that whatever phase gets built comes out of the interaction between the two photons and only one atom. In other words, when the a and the b photons travel through a gas of atoms, it is necessary for both of them to be present at the same atom in order to interact and the likelihood that they would be together in more than one atom is negligible since they are moving at different velocities. On the other hand, the phase-shift per atom is small, because we have to keep the detuning Δ_b large enough to prevent the b photon from getting absorbed in the atomic medium.

Another crucial point to observe is the fact that the norm is not preserved in Eq. (4.78). Formally speaking, it exceeds unity. It is however, equal to 1 to first-order in the small quantity $|g_{24}|^2/\Delta_b$ as shown in Eq. (4.79).

The reason for the norm not being preserved, in general, in the expression for the final state in Eq. (4.78) is in the various approximations we made to derive Eq. (4.50), such as the adiabatic approximations to Eqs. (4.40) and (4.46). We had to do this to simplify the mathematical analysis but the price that we have paid is the non-preservation of the norm of the final state.

Now, we shall calculate the fidelity using the expression for the final state in Eq. (4.73) without any further approximation.

Let us recall that the combined fidelity-phase is given by

$$\sqrt{F}\ e^{i\phi} = \int dz_a \int dz_b\ \xi^*(z_a, z_b, t_{initial})\ \xi(z_a, z_b, t_{final})$$

and the initial state is given by

$$\xi(z_a, z_b, t_{initial}) = \frac{1}{\sigma\sqrt{\pi}}\ e^{-(z_a+vt-z_0)^2/2\sigma^2}\ e^{-z_b^2/2\sigma^2}.$$

On substituting the initial and the final state [from Eq. (4.73)] in the expression for the fidelity, we get

$$\sqrt{F}\ e^{i\phi} = 1 - \frac{i}{\pi}\left(\frac{1}{\alpha\ v\ \sigma}\right)\left(\frac{B'}{1+A'}\right)\int dz_a \int dz_b\ \frac{e^{-(z_a+vt-z_0)^2/\sigma^2}\ e^{-z_b^2/\sigma^2}}{[(z_a+vt-z_0)/\sigma] + [(\beta\Gamma_+\sigma)/\alpha v]}$$
$$+ \frac{i}{\pi}\left(\frac{1}{\alpha\ v\ \sigma}\right)\left(\frac{B'}{1+A'}\right)\int dz_a \int dz_b\ \frac{e^{-(z_a+vt-z_0)^2/\sigma^2}\ e^{-z_b^2/\sigma^2}}{[(z_a+vt-z_0)/\sigma] + [(\beta\Gamma_-\sigma)/\alpha v]}.$$

$$(4.80)$$

It is useful to work in terms of dimensionless variables and parameters. So, we shall define $z_a/\sigma \equiv z'$ and $z_b/\sigma \equiv z''$ which would then make $dz_a\ dz_b = \sigma^2\ dz'\ dz''$. Furthermore, we shall introduce two more dimensionless parameters viz. $\tau \equiv vt/\sigma$ and $\tilde{z} \equiv z_0/\sigma$. In terms of all these dimensionless variables and parameters, Eq. (4.80) can be rewritten as

$$\sqrt{F}\ e^{i\phi} = 1 - \frac{i}{\pi}\left(\frac{\sigma\ B'}{v}\right)\left(\frac{1}{1+A'}\right)\frac{1}{\alpha}\int dz' \int dz'' \frac{e^{-(z'+\tau-\tilde{z})^2}\ e^{-z''^2}}{(z'+\tau-\tilde{z}) + [(\beta\Gamma_+\sigma)/\alpha v]}$$

$$+ \frac{i}{\pi}\left(\frac{\sigma\ B'}{v}\right)\left(\frac{1}{1+A'}\right)\frac{1}{\alpha}\int dz' \int dz'' \frac{e^{-(z'+\tau-\tilde{z})^2}\ e^{-z''^2}}{(z'+\tau-\tilde{z}) + [(\beta\Gamma_-\sigma)/\alpha v]}. \qquad (4.81)$$

Note that A', α and β are dimensionless parameters; B has the dimensions of inverse time, v is the velocity and σ has the dimensions of length. Thus, $\sigma B'/v \equiv \chi$ is dimensionless.

Next, we shall consider $(\beta\Gamma_+\sigma)/(\alpha v)$.

$$\frac{\beta\ \Gamma_+\ \sigma}{\alpha\ v} = \frac{\beta}{\alpha}\underbrace{\left(\frac{\Gamma_+\ \sigma}{c}\right)}_{\Gamma'}\underbrace{\frac{c}{v}}_{1/\beta} = \frac{\Gamma'}{\alpha},$$

where $\Gamma' \equiv \Gamma_+\sigma/c$ is another dimensionless parameter.

Similarly,

$$\frac{\beta\ \Gamma_-\ \sigma}{\alpha\ v} = \frac{\Gamma''}{\alpha},$$

where $\Gamma'' \equiv \Gamma_-\sigma/c$ is dimensionless.

Thus, in terms of Γ' and Γ'', Eq. (4.81) can be rewritten as

$$
\begin{aligned}
\sqrt{F} \, e^{i\phi} = 1 &- \frac{i}{\pi} \left(\frac{\chi}{1 + A'} \right) \int dz' \int dz'' \, \frac{e^{-(z'+\tau-\tilde{z})^2} \, e^{-z''^2}}{\alpha(z' + \tau - \tilde{z}) + \Gamma'} \\
&+ \frac{i}{\pi} \left(\frac{\chi}{1 + A'} \right) \int dz' \int dz'' \, \frac{e^{-(z'+\tau-\tilde{z})^2} \, e^{-z''^2}}{\alpha(z' + \tau - \tilde{z}) + \Gamma''}.
\end{aligned}
\tag{4.82}
$$

The expression for the fidelity in Eq. (4.82) is written completely in terms of

dimensionless quantities. This makes it easier to assign numerical values for various

parameters while numerically evaluating the fidelity.

Next, we shall numerically compute the fidelity for certain values of the parameters

which show up in the previous equation.

In the lab frame, we have the a photon traveling with velocity v_g (its group velocity)

and the b photon traveling at c. However, in the reference frame in which we have solved

the final dynamical equation [see Eq. (4.51)], the b photon is at rest and the a photon is

moving toward the b photon with a velocity $v \equiv c - v_g$. This implies that $1 - v_g/c = v/c$ or

$\beta \equiv v/c = 1 - v_g/c$ and $\alpha \equiv 1 - v/c = v_g/c$. If we set $v_g/c = 0.1$, then $\alpha = 0.1$ and $\beta = 0.9$.

This makes $A' = \beta/\alpha = 9$.

We have already imposed the condition that $\Delta_b \gg |g_{24}|^2$. So, we could set

$|g_{24}|^2/\Delta_b = 0.1$.

Let us recall that $\chi \equiv \sigma B'/v$. If we substitute explicitly for B' from Eq. (4.51) in the expression for χ, we get

$$\begin{aligned}
\chi &= \frac{|g_{24}|^2}{\Delta_b} \left(\frac{\sigma \Gamma_-}{v} \right) A' \\
&= \frac{|g_{24}|^2}{\Delta_b} \underbrace{\left(\frac{\sigma \Gamma_-}{c} \right)}_{\Gamma''} \underbrace{\frac{c}{v}}_{1/\beta} \underbrace{A'}_{\beta/\alpha} \\
&= \frac{|g_{24}|^2}{\Delta_b} \frac{\Gamma''}{\alpha}.
\end{aligned}$$

If we set $\Gamma'' = 10$ and $\Gamma' = 100\ \Gamma'' = 1000$, then $\chi = 10$. Note that Γ'' must be much greater than Γ' since Γ'' and Γ' are directly proportional to Γ_+ and Γ_-, respectively, and in our model, $\Gamma_+ \gg \Gamma_-$. We shall set $\tilde{z} = 5\ \sigma = 5$ and for τ, we shall consider the range $0 \leq \tau \leq 2\tilde{z}$ or $0 \leq \tau \leq 10$. Physically speaking, τ gives the ratio of the distance traveled by the a pulse after interaction to its characteristic length. The larger the value of τ, the farther away is the a photon from the b photon after interaction.

When we put all the numerical values of all these parameters in Eq. (4.82) and compute the fidelity as a function of τ in the range specified above, we obtain $\sqrt{F}\ e^{i\phi} = 1 + 0.099005i$. As a matter of fact, for all values of τ from 0 to 10, we get the same result for the fidelity. It is very evident that the absolute value exceeds unity for the same reason discussed earlier. This is the consequence of the approximations that we have made to derive Eq. (4.50) to keep the problem analytically tractable. More importantly, we can observe that the phase shift is extremely low as predicted by our analytical calculations.

For the sake of completeness, we shall choose a different numerical value for α keeping the same values for Γ', Γ'', $\tilde{z}$. If we set $\alpha = 0.05$, we get $\beta = 0.95$, $A' = 19$ and $\chi = 20$. On substituting these new values in Eq. (4.82), we obtain $\sqrt{F}\, e^{i\phi} = 1 + 0.0990013i$ for the same range of values for τ. We can thus see that the result is no different.
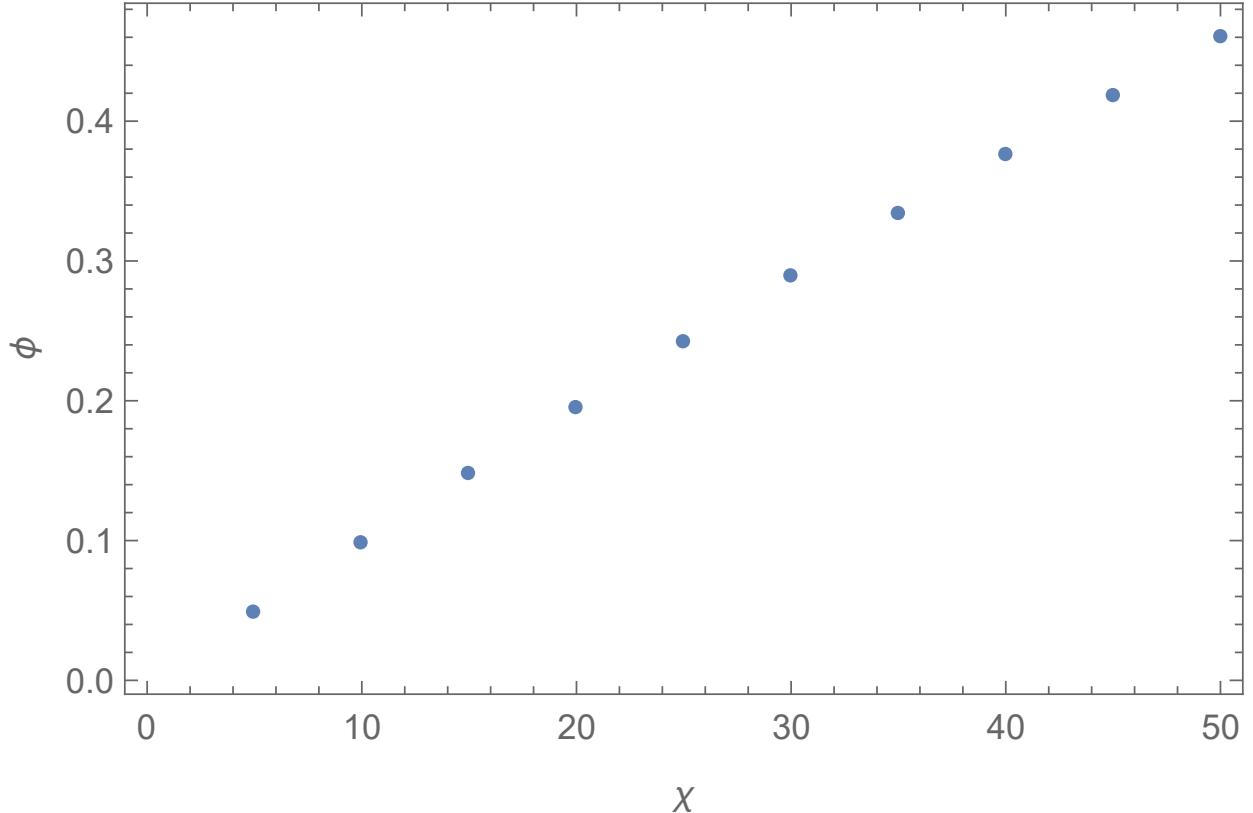

Figure 4.2: Plot of the phase (ϕ) as a function of χ when $\alpha = 0.1$, $\beta = 0.9$, $A' = 9$, $\tilde{z} = 5$, $\tau = 10$.

Finally, we shall try to see how the phase changes if we vary $|g_{24}|^2/\Delta_b$ provided we keep all the other parameters fixed. For this numerical calculation, we shall set $\Gamma'' = 10$, $\Gamma' = 1000$, $\alpha = 0.1$, $\beta = 0.9$, $A' = 9$, $\tilde{z} = 5$ and $\tau = 10$. Let us recall that $\chi = (|g_{24}|^2/\Delta_b)(\Gamma''/\alpha)$. For the chosen values of Γ'' and α, we have $\chi = 100\,(|g_{24}|^2/\Delta_b)$.

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Now we shall vary $|g_{24}|^2/\Delta_b$ from 0.05 to 0.5 in steps of 0.05, i.e. $0.05 \le |g_{24}|^2/\Delta_b \le 0.5$. This would in turn fix the following range for χ: $5 \le \chi \le 50$. Note that we cannot arbitrarily increase $|g_{24}|^2/\Delta_b$ since one of the important assumptions in our model is a very large detuning.

Figure (4.2) shows a plot of ϕ versus χ. We can clearly see that for this range of values of χ, it is very nearly equal to $|g_{24}|^2/\Delta_b$, as predicted by Eq.(4.79).

Thus, we can discern that the Kerr effect is very weak which is why the accumulated phase in this model is to small to be of any practical significance.

4.3 Conclusion

Our aim in this chapter has been to find a conceivable way to physically realize a conditional phase gate in a laboratory. So, we explored the most obvious candidate, "giant Kerr effect" in EIT, to see if we could get obtain unit fidelity with a π phase shift. However, we have seen through our detailed analysis with certain approximations that this model doesn't give us the desired result. One of the reasons for this negative result is that in a completely quantum mechanical model that we have considered here, the two photons when they propagate through an ensemble of atoms eventually interact with only one atom in the whole ensemble. This is because in order to facilitate the atom-photon interaction, the two pulses should be present at the same atom, at the same time and the probability that this can happen in more than one atom is negligible. Thus, all the phase shift essentially comes out of the interaction between the two photons and only one atom which is why the interaction is weak as a consequence of which the total phase that is built is low.

Another reason that has contributed to this outcome is the large detuning. We shall recall that in our model, we had assumed a large detuning so as to prevent the b photon from getting absorbed in the atomic medium. This resulted in a weak Kerr effect which has yielded a negligible phase-shift.

Yet another consequence of this weak interaction between the two pulses is the absence of distortion of the outgoing pulses which is why we obtained almost unit fidelity.

One way to overcome this challenge and come up with a scheme that could possibly be used to construct a phase gate is to have an array of identical atoms coupled losslessly to a one-dimensional waveguide [40].

Another setup that might be helpful for the physical realization of a conditional phase gate is to have two counterpropagating photons travel through a discrete chain of cross-Kerr sites [29, 30].

Chapter 5

Summary

Photons are one of the most effective carriers of quantum information and numerous nonlinear optical schemes have been proposed to implement quantum logical gates at the single-photon level. Despite the many inherent advantages that the photons have as qubits for quantum logical operations, there are some serious challenges to the physical realization of a CPHASE gate. In this dissertation, we have focused on theoretically studying some of these nonlinear optical schemes to construct a conditional phase gate.

Difficulties to the realization of such gates with high fidelity have been pointed out in Kerr media (third-order optical nonlinearities) due to the time-nonlocality of conventional nonlinear media and spectral entanglement of the final state. A few years ago, several authors proposed a scheme based on second-order nonlinearity, and on the coherent evolution of a two-photon state through successive up- and down-conversion processes (with both the photons co-propagating with equal velocities). Motivated by this proposal, we have carried out a rigorous multimode quantized field analysis of this scheme, in chapter 2, to examine the feasibility of using a second-order optical nonlinearity to realize a conditional phase gate between two single-photon pulses. We concluded that even here the spectral entanglement is an important fidelity degrading mechanism. In other words, once the two incident photons are destroyed in the nonlinear medium, the "re-created" two-photon state is constrained only by the conservation of momentum and energy, and it need not spectrally resemble the initial state very much. This indeed degrades the gate performance. We thus inferred that this approach (involving second-order nonlinearity)

suffers from the same difficulties as third-order ones, for schemes involving co-propagating photons with equal velocities.

However, over the last few years, there have been assertions that appear to contest the view that there is an unavoidable trade-off between high fidelity and large phase shift, in a finite bandwidth medium. In particular, we were strongly motivated by couple of theoretical papers viz. one by Xia *et al* and the other one by Brod *et al* which showed that it is indeed possible to achieve unit fidelity with a π phase shift. We were strongly inspired by the scheme suggested by Xia *et al* (which they studied only numerically) where the two photons co-propagate with different velocities in a spatially nonlocal medium. We have developed an analytical model in chapter 3, for the scheme suggested by Xia *et al*. We have generalized their results here to deal with an arbitrary response, initial state and pulse velocity. Our results support the numerical observation in Xia *et al* that a π phase shift with unit fidelity is possible, in principle, in an appropriate limit. We have explicitly shown through our analysis that for the scheme considered here where the two photons co-propagate with different velocities in a spatially nonlocal medium, conservation of energy and momentum lead to non-equivalent algebraic conditions on the wavevectors and frequencies of the interacting photons, which when enforced simultaneously remove the spectral entanglement of the final state. Furthermore, we realized that the role of spatial nonlocal response for the medium is to restrict the bandwidth in order to make the theory well behaved. This is equivalent to truncating the medium's bandwidth by hand (by introducing "cut-offs" in the Hamiltonian). Both these approaches yield similar results which we have explicitly verified toward the end in chapter 3.

It is, however, to be noted that the model developed in chapter 3 whose results were encouraging cannot be directly verified in a laboratory. It just gives us conceptual framework and the necessary conditions to achieve a conditional phase shift. Thus, in chapter 4, we turned our attention to search for a realistic atomic system to build a phase gate at the single-photon level. With this goal, we studied the interaction of two single-photon wavepackets with an ensemble of five-level atoms. We have tried to develop an analytical model by looking at the "giant Kerr" effect in electromagnetically induced transparency to see whether such a system can indeed be used to build a phase gate. Based on our analysis, we had to conclude that this model doesn't yield the desired result, i.e. unit fidelity with a π phase shift. The major reason for this negative outcome is the weak atom-photon interaction in this scheme. This is because in order to facilitate the interaction between the photons and the atomic medium, the two single-photon pulses should be present at the same atom, at the same time. The probability that this can happen in more than one atom is negligible. Thus, all the phase shift essentially comes out of the interaction between the two photons and only one atom which is why the interaction is weak, and as a result, the total phase built is low. Another consequence of this weak interaction between the two photons is the absence of distortion of the outgoing pulses which is why we obtained almost unit fidelity.

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