

Entangled Photon Virtual-State Spectroscopy

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In Partial Fulfillment of the Requirements for the degree of
Bachelor of Science in Physics



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Dedicated to

my family, a special feeling of gratitude to my loving parents, Luis and Gelen Gutiérrez whose words of encouragement have pushed me forward throughout my life. My Brother Andres how has has been a constant source of inspiration. I also dedicate this dissertation to my dog Ozai and friends.

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Abstract

In recent years, the process of absorbing two photons using a non-classical light sources has been studied thoroughly. In particular, it has been experimentally demonstrated that the absorption cross section of two entangled photons is much greater than the absorption cross section of two uncorrelated photons. In this dissertation a source of entangled photons produced through spontaneous parametric down conversion or SPDC was constructed and characterized. It was found that for a pump laser of 411.32 nm and a BBO crystal of 1 mm the produced photons followed the same optical path as the laser, it was also found that the joint spectrum is symmetric at the degenerate wavelength of $822 \pm 0.5\text{ nm}$. This work conclude by showing a first approximation to execute a measurement of the Hong-Ou-Mandel effect. The objective of the measurements of joint spectrum and the Hong-Ou-Mandel effect are to understand the correlation properties of the photon source, with this information a virtual state spectroscopy technique can be implemented on the experiment constructed.

Declaration

The work in this thesis is based on research carried out at the Quantum Optics, the Department of Physics, Bogota. No part of this thesis has been submitted elsewhere for any other degree or qualification and it is all my own work unless referenced to the contrary in the text.

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Chapter 1

Overview

Two-photon absorption (2PA) was first described in 1931 [1, 2], since then it has become a very important tool as a spectroscopic technique, giving us a model to describe the process by which, a molecule or material, absorbs a pair of photons. Where the energy of the transition must be equal to the sum of the energies of the photon pair. Stimulating a 2PA transition using non-classical light has shown remarkable properties such as their ionization characteristics [3], two- and three-photon microscopy to enhanced resolution in biophysics [4] and nonlinear pulsed spectroscopy which has been proven as a powerful tool for investigating the properties of materials [5].

Two photon absorption induced by non-classical light [6] (entangled light) is a recently developed tool that gives us a frame work to extract information about a intermediate states that contributed to a two-photon excitation. This latter has become a spectroscopy technique of its own known as Virtual state spectroscopy (VSS) [7]. The 2PA allows us to force an *angular momentum non-conserving atomic transition* by taking an intermediate step in the total transition, thus making all transitions permitted by quantum mechanics. This intermediate state is known as a virtual state [6]. In VSS the intermediate state is unique to the material that is being induced to 2PA transitions, thus revealing an intrinsic property of this material. The technique of VSS has shown to be able to induced unique characteristics such as two-photon induced transparency [8], the ability to select different states in

complex biological aggregates [9] and the control of entanglement in matter [10,11].

The main goal of this project is to implement a part of a theoretical scheme proposed by Svozilík to exploit VSS [12]. The technique proposes to introduce a time delay and a frequency chirp to change the frequency correlations between photon pairs before the interaction with sample. The entangled 2PA probability depends on the frequency correlation between photons and it is modulated by the temporal delay between photon pairs. Specifically we propose to measure the modulation of the entangled two photon absorption signal in a sample as function of the temporal delay.

A broadly studied and used source of entangled light is generated by Spontaneous Parametric Down Conversion (SPDC) [13]. This technique consist in laser pumping a nonlinear crystal to produce pairs of photons correlated in different degrees of freedom, in this implementation a Beta Barium Borate (BBO) Crystal as the nonlinear crystal. The light produced satisfies the conservation laws of energy and momentum, known as phase matching conditions. SPDC light can be produced with different types of correlations, such as spacial, frequency and polarization correlations between photons pairs. In our case we are interested in frequency correlation since the atomic transitions are completely dependent on the frequency of the stimulating photons. The light produced by BBO is know as type-I SPDC, where the photon pairs have the same polarization and are correlated spatially or in frequency depending in the transverse profile.

Virtual state spectroscopy being a theoretical scheme that gives a new route to ultra sensitive detections [14] and novel applications of quantum phenomenon [15–17], has not been broadly implemented due to its experiemntal cumbursome. The ease with which it can be performed is limited due to the low rate of 2PA and its experimental cumbersome of modulating the frequency correlations. Svozilík et al. [12] experimental scheme is intended to successfully over come these problems by modulating the time delay between photon pairs, thus altering frequency correlations and secondly using SPDC light to produce a large number of entangled light known

as intense twin beams. The general objective of this thesis is to develop an experimental setup that produces a reliable and reproducible delay between entangled photons to study its effect on the Two photo abortionist signal of a sample. To accomplish this objective it is necessary to build a collinear type-I SPDC entangled photon source, and to characterize the frequency correlations of this light source by measuring its second order coherence degree and the joint spectrum. After this, a temporal delay line has to be constructed that can reliably and consistently produce time delays. Lastly, it is necessary to measure the absorption signal of the entangled two photons of a sample as a function of the delay.

The Quantum Optics lab had to my disposal a laser with $\lambda_p = 411.33nm$, $\sigma = 0.148nm$, with this specifications we had to look for a material that had a quantum transition in resonance with this wavelength. Finding a material that had an atomic transition that fulfilled our criteria was done with relative ease. This material is atomic Cesium which has a transition from 6s-8s (refer to Fig.1.1) given by $\lambda_{6s-8s} = 411.32nm$, notice that for this transition $\Delta l = 0$ thus making this a dipolar forbidden transition. Cesium for this transition has states that can be used as an intermediate step between 6s-6p, making it a viable option for a proof of principle for the proposal made by Svozilik.

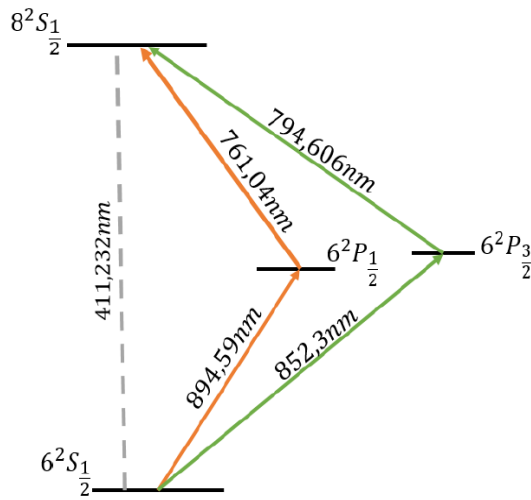


Figure 1.1: Proposed Cesium transition

With this in mind we had to determine the type of SPDC needed to produce correlated light in the spectrum that coincides with the intermediate states of the sample. The SPDC light is determined by the group velocity of pump light and the length of BBO crystal. With these parameters calculated we obtained that collinear type-I SPDC with a 1 mm thick BBO crystal would be the best option for our implementation. With the parameters of the wavelength and the crystal length set,

it was calculated that the produced light would have a wavelength $\lambda_{i,s} = 822nm \pm 130nm$ which covers the virtual states, thus being ideal for 2PA.

Once the entangled photon source is finished the next step would be to construct the delay-line that produces a difference in the arrival time between identical photons. Initially we have proposed a delay using quartz plates as shown in Fig. 3.5. A 50/50 beam splitter produces two branches. In one branch, the quartz plates would be placed such that there is a difference in the optical path producing the delay between arrivals. With these two parts complete, we can introduce the sample and measure the absorption signal. This signal is detected by single photon counters and analyzed with a FPGA detection array. The experimental results will be cross reference to the theoretical calculation from Svozilik.

In the next chapter, the theory and experimental implementation of a SPDC photon source is discussed in detail. Chapter three treats the design, calculation and the implementation of a temporal delay line. Further more, an attempt to characterize the delay line is shown by measuring the Hong-Ou-Mandel effect, this being done to acquire a tool to quantify the temporal difference between photons. Chapter four is a description of how light and matter interact by means of two photon absorption, and a further extension of the experimental setup constructed is discussed. The last chapter concludes on the overall results of this thesis giving a general layout of the future if this project.

Chapter 2

Entangled Photon Source

In this chapter the theoretical and the experimental implementation of an entangled photon source is carried out. The light source is constructed with the intention of being used in a experiment of virtual state spectroscopy. Thus, understanding the frequency and time correlation properties of the photon source is of great interest.

2.1 Quantum Entanglement

A two-particle system is described by a pure state, that is composed of two spatially separated subsystems

$$\hat{\rho} = |\psi\rangle \langle\psi|, \quad |\psi\rangle = \sum_{a,b} c(a,b) |a\rangle |b\rangle \quad (2.1.1)$$

where $\{|a\rangle\}, \{|b\rangle\}$ are two set of orthogonal vectors for each subsystem 1 and 2, respectively, and $\hat{\rho}$ is the density matrix. The state of the two-particle system depends on the factorization properties of $c(a,b)$, that can be written as:

$$c(a,b) \begin{cases} a) = f(a) \otimes g(b) \rightarrow |\psi\rangle = \sum_a f(a) |a\rangle \otimes \sum_b g(b) |b\rangle \\ b) \neq f(a) \otimes g(b) \rightarrow |\psi\rangle = \sum_{a,b} c(a,b) |a\rangle |b\rangle \end{cases} \quad (2.1.2)$$

If $c(a,b)$ does not factor as product of independent states as shown in Eq. (2.1.2) b), the density matrix of the system cannot be written as a product of states for

subsystem 1 and 2:

$$\hat{\rho} \neq \hat{\rho}_1 \otimes \hat{\rho}_2 \quad (2.1.3)$$

and therefore the two-particle system is entangled [18].

Some well known example of two-particle entangled state are the Einstein, Podolsky and Rosen (EPR) [20] or EPR state Eq.(2.1.4), the singlet state [21] of two spin $\frac{1}{2}$ particles Eq. (2.1.5) and the odd and even Shrödinger cat state [22]:

$$|\psi\rangle = \sum_{a,b} \delta(a+b-c_0) |a\rangle |b\rangle \quad (2.1.4)$$

$$|\chi_+\rangle = \frac{1}{\sqrt{2}} (|\uparrow\rangle_1 |\downarrow\rangle_2 - |\downarrow\rangle_1 |\uparrow\rangle_2) \quad (2.1.5)$$

$$|\psi_e\rangle = \mathcal{N}_e(|\alpha\rangle + |-\alpha\rangle), \quad |\psi_o\rangle = \mathcal{N}_o(|\alpha\rangle - |-\alpha\rangle) \quad (2.1.6)$$

In EPR state (2.1.4) a and b can be momentum or position of particle 1 and 2 respectively, in the singlet state (2.1.5) the ket vectors represent the direction of the spin for each particle and the Shrödinger cat state (2.1.6) describes the entanglement between to phases shifts of the same coherent state $|\alpha\rangle$ where each individual state is a classical-like state but the superposition of them has non-classical properties. These states as examples lets us understand that quantum entanglement can happen between different physical properties.

2.2 Spontaneous Parametric Down Conversion

A two photon state of a signal-idler photon pair is generated when a pump laser beam is incident onto an nonlinear optical material, this is know as *Spontaneous Parametric Down Conversion* or SPDC [19]. The state of the photon pair can be derived mathematically by applying perturbation theory to the nonlinear interaction Hamiltonian that can be written as

$$|\psi\rangle = \sum_{s,i} \delta(w_s + w_i - w_p) \delta(\vec{k}_s + \vec{k}_i - \vec{k}_p) a_s^\dagger(\vec{k}_s) a_i^\dagger(\vec{k}_i) |0\rangle \quad (2.2.1)$$

where s, i and p stand for signal, idler and pump respectably, $w_j, \vec{k}_j$ are the frequency and the wave-vector. The wave function of the photon pair of SPDC has the

same form as an EPR state (2.1.4) showing entanglement in frequency and momentum. Its also important to notice that $w_p, \vec{k}_p$ can be consider constant for a stable monochromatic CW pump laser, $a_s^\dagger$ and $a_i^\dagger$ are the creation operators for signal and idler photons. Thus, we can see from Eq. (2.2.1) that in the SPDC process a photon par (signal, idler) are created when the pump interacts with an nonlinear crystal. As a photon travels through the crystal occasionally the nonlinear interaction annihilates a high frequency pump photon but simultaneously two lower frequency photons are created. The created photon pair satisfies the phase matching conditions [23] as shown below:

$$w_p = w_s + w_i \quad , \quad \vec{k}_p = \vec{k}_s + \vec{k}_i \quad (2.2.2)$$

One can notice that this condition arises from the delta function in Eq. (2.2.1). The SPDC process can produce pair photons with the same polarization or orthogonal polarization. When the photons have the same polarization they are referred to as type-I SPDC, and when the produced photons have orthogonal polarization there know as type-II SPDC. The process is said to be degenerated when the SPDC photon pair has the same wavelength ($\lambda_s = \lambda_i = 2\lambda_p$) and non-degenerate other wise. Traditionally, when the direction of the photon pair is different than the direction of pump beam, it is said to be non-collinear process as shown in Fig. 2.1

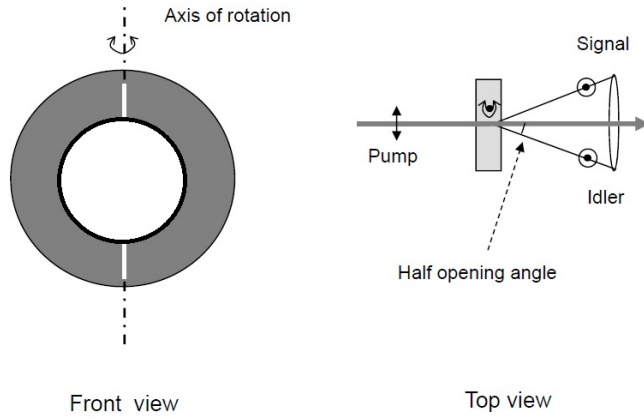


Figure 2.1: Type-I non-collinear SPDC visual representation. Photons from the pump beam are annihilated and a photon pair signal and idler is created. The signal and idler photons have identical polarization, which is orthogonal to the pump photon polarization.

The SPDC process arises due to the non-linear crystal interacting with a pump beam. A crystal is said to be nonlinear if the polarization or the dipole moment per unit volume depends of terms of $\chi^{(m)}$ (the m th order electrical susceptibility tensor), given by:

$$P_i = \chi_{i,j}^{(1)} E_j + \chi_{i,j,k}^{(2)} E_j E_k + \chi_{i,j,k,l}^{(3)} E_j E_k E_l + \dots \quad (2.2.3)$$

In SPDC, the main driver of this process is the second order nonlinear susceptibility $\chi^{(2)}$ [19]. Thus, one can write the second order nonlinear interaction Hamiltonian as:

$$H = \epsilon_0 \int_V d^3r \chi_{i,j,k}^{(2)} E_i E_j E_k \quad (2.2.4)$$

where the integral is taken over the interaction volume V . The electrical fields in Eq.(2.2.4) are written as their Fourier representation.

$$E(\vec{r}, t) = \int d^3\vec{k} \left[\vec{E}^{(-)}(\vec{k}) e^{-i(w(\vec{k})t - \vec{k}\vec{r})} + \vec{E}^{(+)}(\vec{k}) e^{i(w(\vec{k})t - \vec{k}\vec{r})} \right] \quad (2.2.5)$$

It is reasonable to consider the pump field as classical, since it is usually a laser beam, and quantize the produced signal and idler photons. Thus we write the

electrical fields $\vec{E}^{(+)}, \vec{E}^{(-)}$ as :

$$\vec{E}^{(-)}(\vec{k}) = i\sqrt{\frac{2\pi\hbar\omega}{V}}a^\dagger(\vec{k}), \quad \vec{E}^{(+)}(\vec{k}) = i\sqrt{\frac{2\pi\hbar\omega}{V}}a(\vec{k}) \quad (2.2.6)$$

Substituting (2.2.5) into (2.2.4) and selecting the terms in the interaction Hamiltonian where the pump photon is annihilated and two photons are created, given by the interpretation of Eq. (2.2.6), one gets:

$$H_{int}(t) = \epsilon_0 \int d^3\vec{k}_s d^3\vec{k}_i \times \\ \chi^{(2)} E_p^{(+)} e^{i(w_p t - \vec{k}_p \vec{r})} E_s^{(-)} e^{-i(w_s(\vec{k}_s)t - \vec{k}_s \vec{r})} E_i^{(-)} e^{-i(w_i(\vec{k}_i)t - \vec{k}_i \vec{r})} + h.c \quad (2.2.7)$$

where h.c. stands for the Hermitian conjugate. The state of the produced photon pair can be estimated by using first order perturbation theory. Assuming an infinite interaction time the wave function for the photon pair can be written as:

$$|\Psi\rangle = A \int d^3\vec{k}_s d^3\vec{k}_i \delta(w_p - w_s(\vec{k}_s) - w_i(\vec{k}_i)) \delta(\vec{k}_p - \vec{k}_s - \vec{k}_i) a_s^\dagger(\vec{k}_s) a_i^\dagger(\vec{k}_i) |0\rangle \quad (2.2.8)$$

2.3 Joint Spectrum

A more realistic representation of the SPDC process is to consider a finite interaction volume. The SPDC quantum state that describes this process can not be written as Eq. (2.2.8) but it has a close resemblance, differing in terms that take into account the length of the crystal and the interaction area [25]. Now let me discuss for a moment the infinite interaction time, that implies an infinite interaction volume. This assumption lets us write the interaction volume as:

$$\frac{1}{(2\pi)^3} \int d^3\vec{r} e^{i(\vec{k}_p - \vec{k}_s - \vec{k}_i)\vec{r}} = \delta(\vec{k}_p - \vec{k}_s - \vec{k}_i) \quad (2.3.1)$$

which is a good approximation. Lets consider a crystal of length L, and write the interaction volume in terms of the transverse and longitudinal components of momentum ($\vec{k} = \vec{q} + k_z \hat{z}$) and position ($\vec{r} = \vec{x} + r \hat{z}$) vectors:

$$\int d^3\vec{r} e^{i(\vec{k}_p - \vec{k}_i - \vec{k}_s)\vec{r}} = \int_0^L dr e^{i(k_{zp} - k_{zi} - k_{zs})r} \int d^2\vec{x} e^{-i(q_s + q_i)\vec{x}} \quad (2.3.2)$$

Using the new definition of the interaction volume given in Eq. (2.3.2), the SPDC quantum state can be expressed in terms of the spectral amplitude $\Psi(\Delta_k L)$,

$$L\Psi(\Delta_k L) \equiv \int_0^L dr e^{i(k_{zp} - k_{zi} - k_{zs})r} \quad (2.3.3)$$

where the phase mismatch in the longitudinal direction is given by $\Delta_k = k_{zp} - k_{zi} - k_{zs}$. The second term in Eq. (2.3.2) is the integral over the interaction area and it can be considered as

$$h_{ir}(\Delta_{\vec{q}}) = \int d^2\vec{x} e^{-i(\vec{q}_s + \vec{q}_i)\vec{x}} \quad (2.3.4)$$

where $\Delta_{\vec{q}} = \vec{q}_s + \vec{q}_i$ is the phase mismatch in the transverse direction. Using our prior definitions, we can write the SPDC quantum state for the case of finite interaction volume as:

$$|\Psi\rangle = A_2 \int d^3\vec{k}_s \int d^3\vec{k}_i h_{ir}(\Delta_{\vec{q}}) \Psi(\Delta_k L) \delta(w_p - w_s - w_i) a_s^\dagger(\vec{k}_s) a_i^\dagger(\vec{k}_i) |0\rangle \quad (2.3.5)$$

It's more useful to work with the spectrum of SPDC radiation in one fixed direction of propagation. Experimentally the measurement of the power spectrum of the signal idler pair can be achieved with relative ease. Making it a valuable resource and it also reduces our calculation by not working with the total output coming from the SPDC Crystal. The power spectrum of the signal idler pair is given by the modulus squared of the spectral amplitude,

$$F(\nu) = |\Psi(\Delta_k L)|^2 \quad (2.3.6)$$

where ν is the detuning variable [25]. Now we can rewrite the spectral amplitude given in Eq.(2.3.3) as follows,

$$L\Psi(\Delta_k L) = \int_0^L dr e^{i(k_{zp} - k_{zi} - k_{zs})r} \quad (2.3.7)$$

$$= e^{\frac{i\Delta_k L}{2}} \text{sinc}\left(\frac{\Delta_k L}{2}\right). \quad (2.3.8)$$

Notice that the joint spectrum intensity function Eq.(2.3.6),

$$F(\nu) \approx \text{sinc}^2 \left(\frac{\Delta_k L}{2} \right) \quad (2.3.9)$$

contains the spectral properties of SPDC photons as discussed in [26]. They also showed that for a type-I SPDC, the joint spectrum can be written in terms of a longitudinal phase mismatch(Δ_k) that takes into account the dispersion and the finite thickness of the crystal which is of interest for our experimental implementation.

2.4 Coherence degree

The Quantum mechanical description of the statistical and coherence properties of an electromagnetic field are given by the degree of coherence [27]. In its simplest form, the first order coherence degree, $g^{(1)}$, can be used for quantifying the coherence between two electric fields. The second order coherence degree, $g^{(2)}$, is typically used to describe the statistical character of the intensity fluctuations. It is important to note that the first order correlation is actually the amplitude-amplitude correlation and the second order correlation is the intensity-intensity correlation.

2.4.1 First Order Coherence

The normalized first order correlation function is written as [23]

$$g^{(1)}(\vec{r}_1, t_1; \vec{r}_2, t_2) = \frac{\langle E^*(\vec{r}_1, t_1) E(\vec{r}_2, t_2) \rangle}{[\langle |E(\vec{r}_1, t_1)|^2 \rangle \langle |E(\vec{r}_2, t_2)|^2 \rangle]^{1/2}} \quad (2.4.1)$$

where r_i , t_i are the position and the time of measurement of one detector, and $\langle \rangle$ accounts for the statistical average. Consider the case where one deals with a state that its statistical properties do not change with time, a stationary state, the ensemble average can be taken as a time average. Furthermore, let's consider only one detector $\vec{r}_1 = \vec{r}_2$. For this case the time dependence is in terms only of a delay between measurements given by $\tau = t_1 - t_2$. Thus, allowing us to write the first order coherence degree as

$$g^{(1)}(\tau) = \frac{\langle E^*(t) E(t + \tau) \rangle}{\langle |E(t)|^2 \rangle}. \quad (2.4.2)$$

In optical interferometers such as the Michelson interferometer, Mach-Zehnder interferometer, or Sagnac interferometer, one splits an electric field into two components, and introducing a time delay to one of the components with respect to the other, and then recombines them. The intensity of the resulting field is measured as a function of the time delay as show in Eq.(2.4.2), where generally, $g^{(1)}(\tau) = g^{(1)}(-\tau)$, $g^{(1)}(0) = 1$. For example, a single frequency laser has degree of first order coherence given by $g^{(1)}(\tau) = \exp(-i\omega_0\tau)$ where ω_0 is the central frequency of the light [28].

2.4.2 Second Order Coherence

Now lets consider the second order correlation function written as:

$$g^{(2)}(\vec{r}_1, t_2; \vec{r}_2, t_2) = \frac{\langle E^*(\vec{r}_1, t_1)E^*(\vec{r}_2, t_2)E(\vec{r}_1, t_1)E(\vec{r}_2, t_2) \rangle}{\langle |E(\vec{r}_1, t_1)|^2 \rangle \langle |E(\vec{r}_2, t_2)|^2 \rangle} \quad (2.4.3)$$

One can show that in general the second order coherence degree is even $g^{(2)}(\tau) = g^{(2)}(-\tau)$. Now, if we apply the Cauchy-Schwarz inequality to Eq.(2.4.3) one gets that $1 \leq g^{(2)}(\tau) \leq \infty$ and that $g^{(2)}(\tau) \leq g^{(2)}(0)$ [24]. If we consider the output of a beamsplitter, the Transmitted and Reflected beams, and treat the output fields as classical fields one can write the second order coherence as:

$$g_{T,R}^{(2)}(\tau) = \frac{\langle I_T(t+\tau)I_R(t) \rangle}{\langle I_T(t+\tau) \rangle \langle I_R(t) \rangle} \quad (2.4.4)$$

Which has a resemblance to Eq.(2.4.2), but its worth noting that the second order coherence is not a generalization of the first order coherence. Now if we consider the second coherence degree of a laser one gets $g^{(2)}(\tau) = 1$ which is drastically different to the Gaussian result for the first order coherence. Thus revealing, that in some sense the second order coherence gives a measurement of how non-classical is the light source, being constant for the a completely classical light source.

2.5 Experimental Implementation

Experimentally construing an entangled photon source of SPDC light consists of two parts, first, building the source of light second, characterizing it by measuring its

correlations as discussed previously. To build an SPDC photon source, one needs a pump laser and a nonlinear Beta Barium Borate (BBO) crystal. In this experimental implementation the pump laser used was a L405P20 fabricated by Thorlabs. This laser has a temperature and current controller as seen in FIG.2.2 that allows us to change the central laser wavelength from 400 nm up to 415 nm. To ensure we had a well defined single wavelength pump laser a diffraction grating was used to select the second order diffraction at a wave length of $\lambda_p = 411.33$ nm which can be tune by changing the temperature or the current being applied.



Figure 2.2: Diode laser used in our experimental implementation, with its respective current and temperature controllers

2.5.1 Transverse spatial profile

With this well defined monochromatic pump laser impinging on a 1mm BBO crystal, which is a nonlinear optical crystal, we built the SPDC source. To focus the pump laser on the BBO crystal a lens with a focal length of 200mm was used producing a waist of $132.5\ \mu\text{m}$ at the crystal as show in Fig. 2.3. After the BBO a lens with focal length of 75mm was placed to collimate the produced SPDC light and a filter FEL700 was used which is a high pass filter. With this setup the first measurement we were interested in executing was the transverse spacial profile. To capture the the spacial profile a CCD camera was used to collect images of the produced SPDC photons.

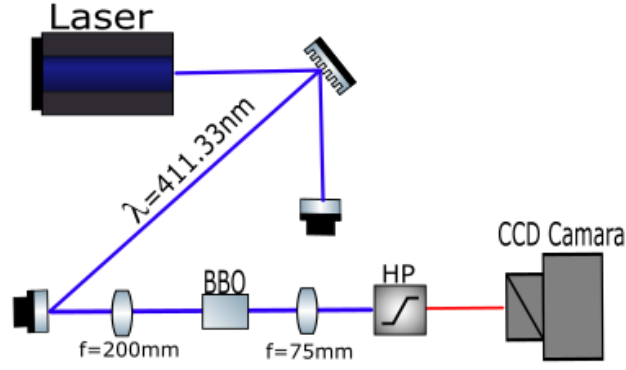


Figure 2.3: Experimental setup used to capture the transverse spatial profile using a CCD camera. HPF is a high pass filter of 700nm to remove the pump laser light left after the BBO

Theoretically for a type-I SPDC light one expects to get a transvers spatial profile as shown in FIG.2.1 which shows the spatial distribution for one fixed position between the propagation direction of the beam and the optical axis of the BBO crystal. In our implementation several images were taken for different positions of the optical axes, where moving the optical axis consist in rotating the BBO crystal. By doing this we observed the complete transition form a collinear configuration to noncollinear one as shown in FIG 2.4. Notice that in FIG.2.3 we are using a High pass filter PEL700 fabricated by Thorlabs, which filters out must of the pump laser and leaves us with the SPDC light, the residual pump laser can still be observed as a point in the center of FIG.2.4.

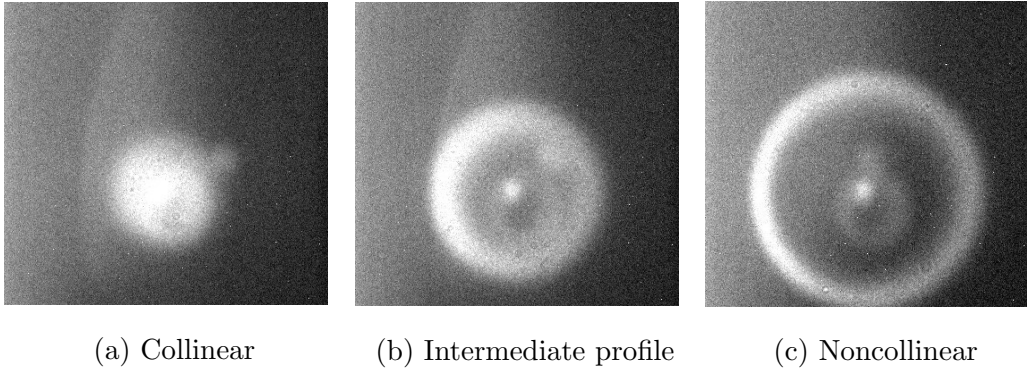


Figure 2.4: Images of the transverse distribution of the SPDC light for different angles between the optical axes of the BBO and the direction of propagation of the pump light

With this measurement of the transverse spatial profile, we conclude that the crystal is effectively a type-I crystal, due to the fact that the spatial profile FIG.2.4 is the same as predicted by the SPDC theory and shown in FIG.2.1. For the rest of this experiment, the collinear configuration was kept constant which is shown in FIG.2.4a.

2.5.2 Second Order Coherence degree

At this point the crystal and the laser have been set up in such a way that the light produced is a collinear SPDC type-I source. Now we need to determine if the light produced has a time correlation as discussed in section 2.4.2. To implement the measurement of the second order coherence degree, the produced SPDC light was split and coupled into two different detectors as shown in FIG. 2.5. The technique followed to implement this measurement and the analysis of the data was the one used by Thorn in Ref [27].

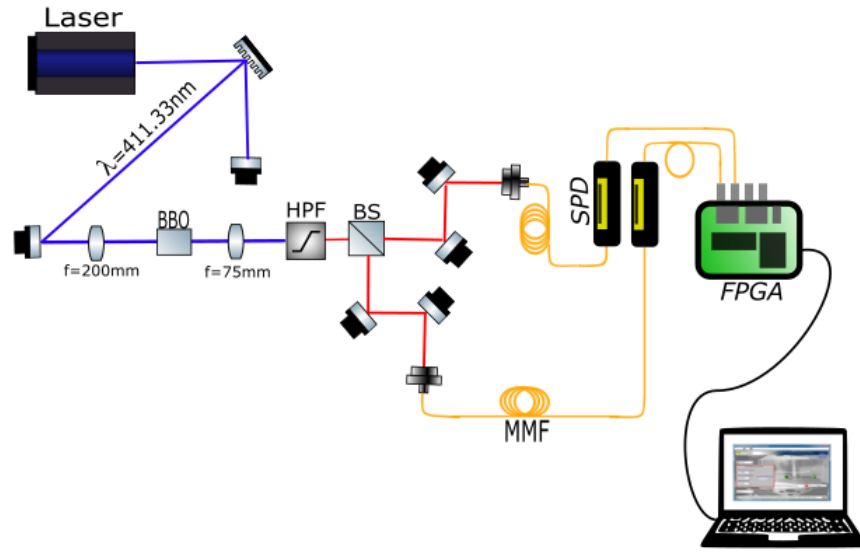


Figure 2.5: Experimental setup implemented to measure the second order coherence degree. SPD stands for Single Photon Detector, where each out put of the Beam Splitter (BS) has an SPD system, the counts or detection's are processed for coincidences by the FPGA and analyzed by the computer. MMF stands for multimode fiber.

No selection of wavelength was involved in the this measurement, due to the fact that all the light coupled into the multimode fiber is being sent to a single photon detection system, then the FPGA is checking for coincidences in all the light detected. Since a 50/50 BS was used we can expect that the output a and b of the BS has an intensity $E_{a,out}, E_{b,out} < E_{in}$. Each arm was built to have approximately the same optical length, and they each have there own SPD system. The recollection of the data was executed as follows, one arm was left with a fixed detection time t_1 and the second arm swept different detection time t_2 produced by introducing an electronic temporal delay with the FPGA. The amount of coincidences between outputs of the BS is stored for the different time delays. In Fig 2.6 the data collected is shown, where in the vertical axis we have the normalized second order degree intensity and in the horizontal axis the temporal difference between the time of measurement $\tau = t_2 - t_1$ as defined in Section 2.4.2.

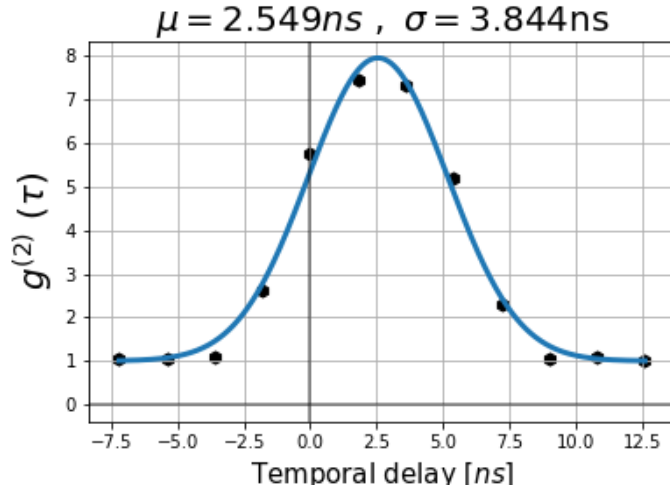


Figure 2.6: Data recollected of the Second order coherence and fitted with a Gaussian profile. There is a electrical delay between the two detectors that corresponds to μ , as seen by the Gaussian having a mean different than zero

Note that Fig. 2.6 does follow the theoretically imposed conditions on the second order coherence degree, that was discussed in section 2.4.2. A valuable piece of information can be extracted from this measurement, let us recall that the maximum coincidences are expected to be when $g^2(0)$, but from Fig. 2.6 we can see that the maximum is at 2.6ns telling us that there is a slight difference in the detection time between the detection system of each arm. With this insight the difference in detection times is tuned so we get the maximum of coincidences threw out the rest of the experiment.

2.5.3 Joint Spectrum

We have determined that the light source in our experiment is a type-I SPDC and it has a collinear transverse spatial profile, we have also determined that it has a second order degree that presents high coherence. Thus, revealing that this light has non classical statistical properties or its quantum mechanically correlated. Now we are interested on determining exactly what wavelengths are present in these correlations, this can be achieved by measuring the joint spectrum as mentioned previously in section 2.3.

In the previous measurement all the coupled light was sent straight through to the SPD system, now we are interested in separating the different wavelengths and then checking for coincidences. Two monochromators were used Scientech, one for each arm, to separate the different wavelengths of the SPDC light as seen in Fig. 2.7. The monochromators allow us to select a single wavelength at a cost of reducing the total count of coincidences.

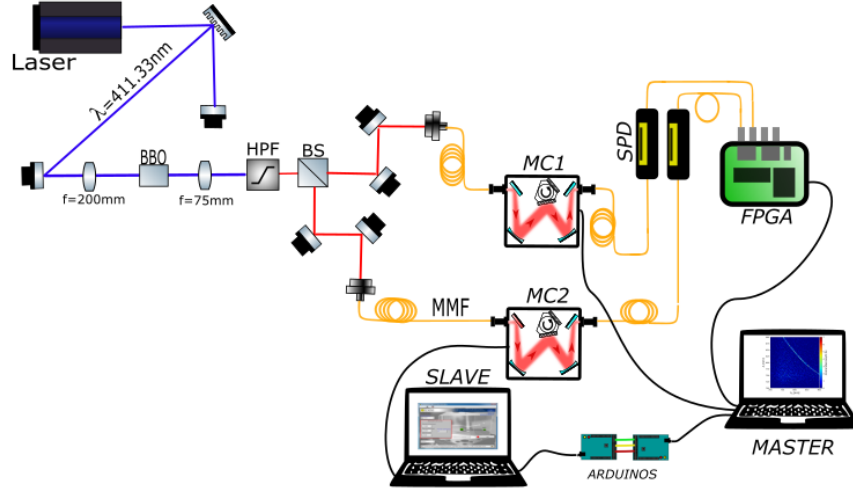
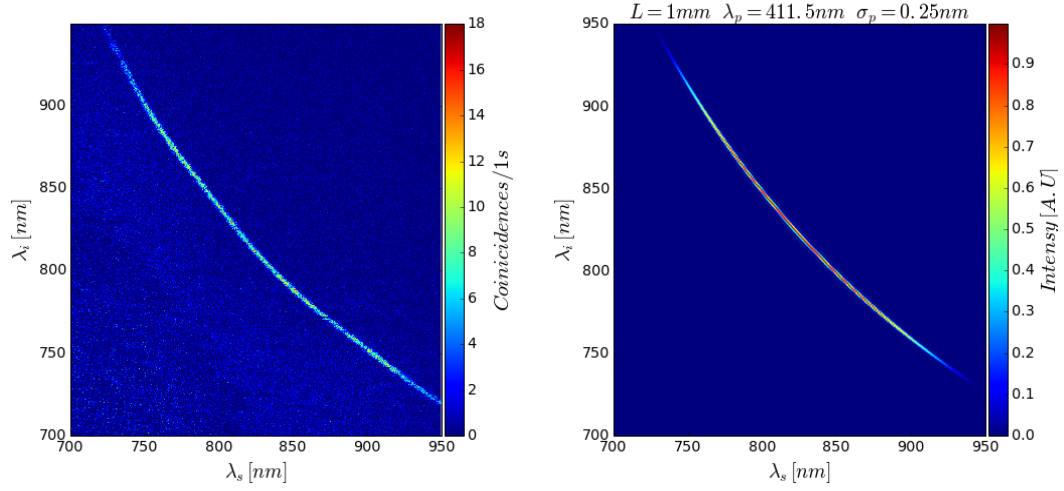


Figure 2.7: Experimental setup implemented to measure the joint spectrum. MC stands for Monochromators that are used to select a single frequency before sending it to the SPD system.

The data was recollected by sweeping a range of wavelengths in steps of 0.5 nm and a collection time of one second for each set of wavelength. Both MC1 and MC2 sweep the range 700 nm - 950 nm, but point by point MC1 was left at a fixed wavelength and MC2 scanned the full range, then when MC2 is done MC1 changes its wavelength by 0.5 nm and MC2 scans all the range again. The theoretical model was calculated by implementing what was shown in Sec. 2.3, its further analysis and comparison with the measurement executed was done by an other member of the Quantum optics group.

As its theoretically expected the joint spectrum is anticorrelated and symmetric about the degenerate wavelength 822 nm. The SPDC light produced can be seen to have correlation for nondegenerate wavelengths, where one can find a pair of



(a) Experimental measurement of the two-photon joint spectrum (b) Theoretically expected two-photon joint spectrum

Figure 2.8: Experimental Results and the theoretical prediction of the joint spectrum for a type-I collinear SPDC. One can see that what was measured has a great resemblance to the theoretical prediction of the Joint Spectrum

correlated photons, signal and idler, with the wavelength required in the overview seen in Fig 1.1. With the two photon joint spectrum measured we can conclude that the SPDC photon source was constructed in such a way that the required wavelengths for the VSS experiment are produced. With this photon source we now need to produce a temporal delay in the arrival time to sample. To characterize this delay line we implemented a technique of two-photon interference, known as the Hong-Ou-Mandel effect, it will be discussed in detail in the next chapter.

Chapter 3

Interferometer Calibration

Interferometry is a powerful technique for extracting information about various types of waves, this being a process where two waves are superimposed upon each other to determine various quantities of interest. This technique has proven to be remarkably robust and dependable as a experimental tool, due to its experimental precision and the wide variety of fields in which it has applications [23]. Interferometers are widely used in science and industry for the measurement of small displacements, refractive index changes and surface irregularities. In an interferometer, light from a single source is split into two beams that travel different optical paths, then combined again to produce interference [24]. The resulting interference fringes give information about the difference in optical path length. This technique are employed in this work to characterize the temporal delay between photon pairs by measuring the Hong-Ou-mandel deep.

3.1 Hong-Ou-Mandel Effect

Exploiting the quantum mechanical nature of two identical photons passing through a beam splitter has shown to be an important scientific tool, yielded in the form of the Hong-Ou-Mandel (HOM) interferometer [29]. Where its clearest purpose is testing the degree of indistinguishability between photon generated from any light source. However, it can also be used to determine path differences between two identical photons. There are two possibilities for an incident photon in a beam splitter, being

reflected or transmitted as shown in Fig. 3.1. For a 50:50 BS one would expect that the intensity of the out put fields would be half of the total intensity, independent in what position of the optical path one places the BS, or the statistical properties of the incident light, but for a two photon input a non-intuitive effect emerges.

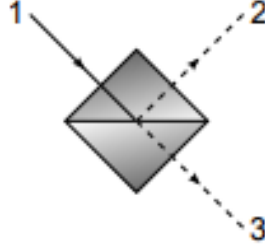


Figure 3.1: Beam splitter single photon out comes, incident photon 1, reflected photon 2, transmitted photon 3. There only is two possible out comes for an incident beam

3.1.1 Two Photon interference

For a 50:50 beam splitter there are exactly four different options for the reflection or transmission of the photons. Both photons can either be reflected, or transmitted, photon 1 can transmit and photon 2 can reflect, or vice versa. This is visually represented in Fig. 3.2a using two different colors for photon 1 and 2. One can tell that options 2 and 3 are in a sense conjugate possibilities, where the only difference is the color of the reflected and transmitted photons. Now consider the situation where we can not distinguish between photons (they both have the same color), this is illustrated in Fig. 3.2b where we can see that the conjugate possibility, 2 and 3 cancel each other out, thus interfering destructively between them, which only leaves options 1 and 2, naturally implying that no coincidence can be detected, thus revealing the HOM effect.

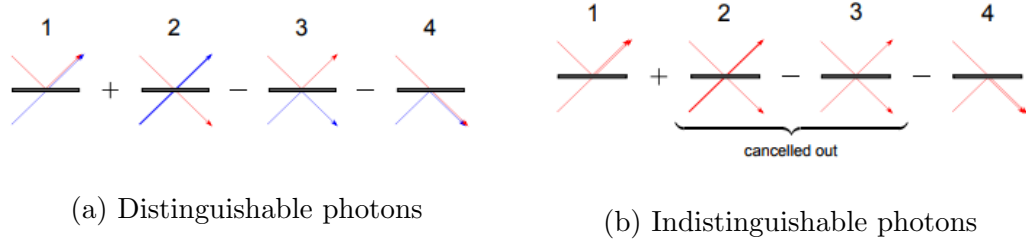


Figure 3.2: Visual representation of the two photon propagation possibilities through a BS considering distinguishable photons, different colors, and indistinguishable photons, same color. The sign is a representation of the signs read from the BS wave equation.

In more mathematical terms the two inputs can be described as a Fock state of $|1_{01}, 1_{02}\rangle$ where the field modes on the input side of the beam splitter are labeled 01, 02 and the output side 1, 2. The wave function for the two photons can then be resolved in general as follows: [29]

$$|\Psi_{out}\rangle = (R - T) |1_1, 1_2\rangle + i(2RT)^{1/2} |2_1, 0_2\rangle + i(2RT)^{1/2} |0_1, 2_2\rangle \quad (3.1.1)$$

Here R is the reflectivity and T is the transmissivity of the beam splitter, for a 50:50 beam splitter $R = T = \frac{1}{2}$. The first term of this wave function describes the state where each photon takes a different output $|1_1, 1_2\rangle$, since we are considering an ideal BS this term cancels out. This cancellation is the reason no coincidental measurement should be made, or as said in Ref. [30] "the quantum mechanical nature of the system requires us to add each of the possible scenarios together in order to determine the wave function and in doing so the scenarios in which both photons exit in different direction add to zero". A detailed calculation of the wave function is shown in the appendix.

3.1.2 Theoretical Implementation

However, in practice the two photons are never monochromatic Hong et al. derived the following expression for the number of coincident photons, by representing the wave function of the two photon state produced in the nonlinear crystal by a linear

superposition given by Eq. (3.1.2), where this formulation was already used implicitly in Sec. 2.3 in a slightly different context. In this case ϕ is the weight function where this weight function was the spectral amplitude in the case of Sec. 2.3.

$$|\Psi\rangle = \int dw \phi(w_1, w_0 - w_1) |w_1, w_0 - w_1\rangle \quad (3.1.2)$$

From here a series of manipulations are executed in Ref. [29], which are out of the scope of this work. To derive the following relation for the number of coincidence measured.

$$N_c = C \left[T^2 + R^2 - 2RT \frac{\int_{-\infty}^{\infty} g(\tau)g(\tau - 2\delta\tau)d\tau}{\int_{-\infty}^{\infty} g^2(\tau)d\tau} \right] \quad (3.1.3)$$

Where $g(\tau)$ and $g^2(\tau)$ are the first and second coherence degrees as previously seen in Sec. 2.4. The expected number N_C of observed photon coincidences is modulated by the temporal difference $\delta\tau$ between photons. It follows that if there is no temporal difference between photons $\delta\tau = 0$, then $N_C = C(R - T)^2$. Note that N_C vanishes when considering an ideal beam splitter as expected from Sec. 3.1.1. This effect of vanishing of photon coincidence is a purely quantum-mechanical feature of the fourth-order interference as shown in [31]. Lets consider now the special case where the first order coherence degree is a Gaussian in w with bandwidth Δw . Then $g(\tau) = \exp((-\Delta w\tau)^2/2)$, replacing in Eq. 3.1.3 gives us a expected number of observed photon coincidence [29]

$$N_c = C(T^2 + R^2) \left[1 - \frac{2RT}{R^2 + T^2} e^{-(\Delta w\delta t)^2} \right] \quad (3.1.4)$$

This expression describes the HOM deep, considering the spectral properties of correlated light source.

3.2 Experimental Implementation

A theoretical calculation of the HOM deep is shown in Fig.3.3, it was executed by a member of the quantum optics group, where we can see that the resolution of the measurement is in the subpicosecond regimen. It is also worth noting that it is a very slim deep, needing precise measurements in the range of $\tau = [-20fs, 20fs]$.

Now we need to find way to produce the delays with some fine resolution so we can measure the HOM deep.

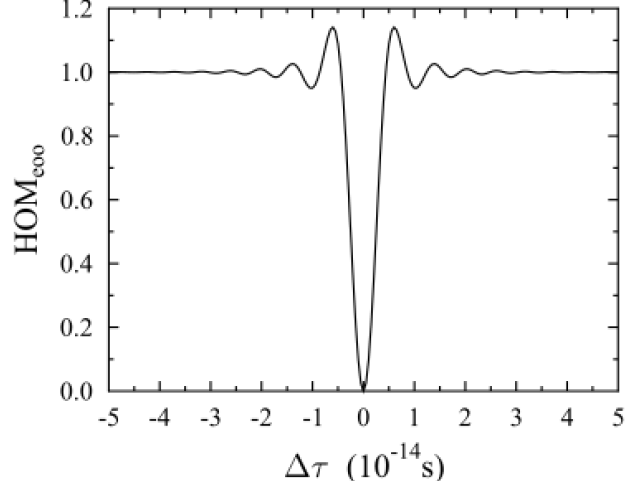


Figure 3.3: Theoretical calculated HOM deep for the SPDC photon source constructed. Note that HOM_{ooo} stands for the polarization considered extraordinary input an ordinary output.

The experimental implementation proposed is to produce this temporal difference by introducing a quartz plate in the optical path of one photon of the SPDC photon pair. The experimental setup is shown in Fig.3.5. A closer look to the quartz plates and some critical values delays are shown in Fig.3.4 and Table 3.1 respectively. The quartz plates are mounted on transitional stages that has a movement of $500 \mu m$ per revolution. Since the movement is diagonal to the optical path, as seen in Fig.3.5 the actual thickness of quartz added to the optical path is $\Delta x_0 = 0.29 mm$, in terms of the time difference it corresponds to a $\Delta \tau_0 = 8.51 fs$ which allows us to make atleast a couple of measurements in the range of τ need to observe the deep.

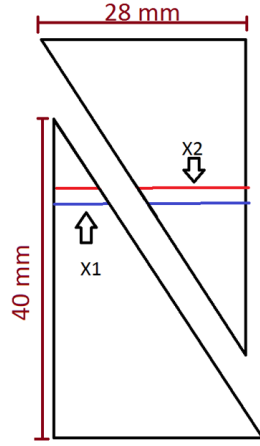


Figure 3.4: Quartz plates implemented to generate the time delay between photons where x_i represent the width traveled by the photons

$x = x_1 + x_2$	τ
11.46 μm	0.34 fs
0.29 mm	8.51 fs
13.7 mm	0.41 ps
17.1 mm	0.51 ps
18.4 mm	0.55 ps

Table 3.1: Relevant conversion of quartz width to temporal delay produced. The values shown were calculate considering 810 nm light [24]. The finest resolution used in the experiment is the first element of this table, the second term is the Δx for each measurement done, from there the initial, middle and last width are shown.

In the experimental setup implemented a set of two mirrors and a beam splitter were added to the setup in Fig. 2.8a to build an interferometer as shown in Fig. 3.5. The alignment of the outputs of the second beam splitter were coupled in such a way that they had the same optical path, overlapping in the detection system. In the Fig. 3.5 we have two sets of quartz plates, one set has a tunable thickness (top set) and the second one was placed at a constant width (bottom set). This second set of quartz was placed due to the theoretical necessity to measure negative time delays. This implies that we need to add a fixed time delay t_{02} to one arm thus, moving the HOM deep location. The HOM deep will now be seen when $t_1 = t_{02}$ such that $\Delta\tau = t_{02} - t_1 = 0$. With the top set of quartz, a time interval $t_1 < t_{02}$ is possible. This can be achieve by setting the top set with a smaller width than the bottom set of quartz, thus, producing a $\Delta\tau < 0$.

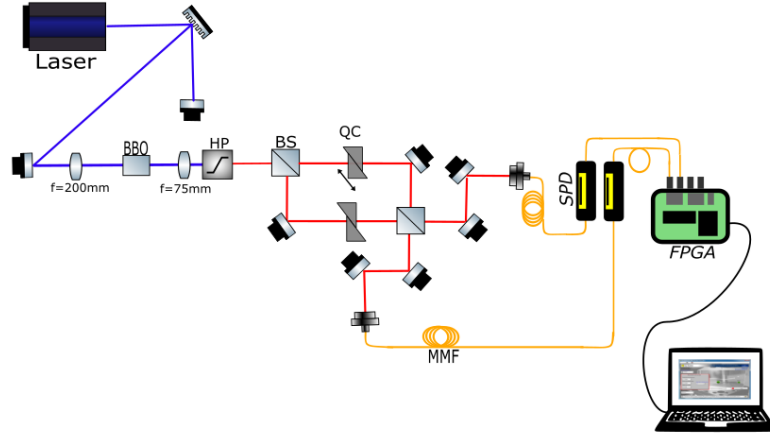
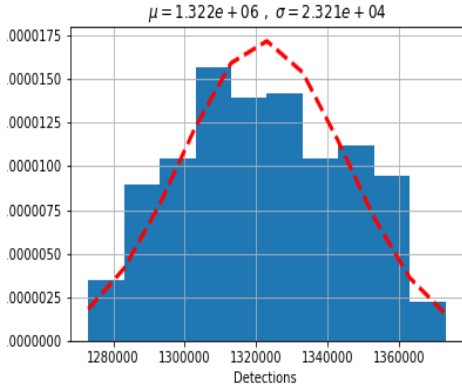
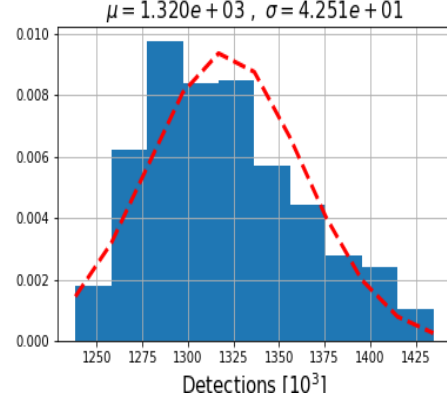


Figure 3.5: Experimental setup implemented, where QC stands for quartz plate. Note that one set of the quartz plate stays at a fixed position and the other set of quartz plate vary in width.

The Data recollected from the measurement is shown in Fig. 3.6-3.7, the total amount of detection, per arm, for each step executed, is shown in Fig. 3.6. From which it can be concluded that there is an oscillation in the total amount of counts in both arms, due to the large sigma seen in Fig. 3.6a-3.6b. This tendency of non constant total single photon detection generates noise, making it harder to distinguish the detection of the HOM deep. Fig. 3.7b is the normalized coincidences (as discussed in Sec.2.4.2) from the plot one can see that there seem to be a drop in the amount of coincidences, for positive values of $\tau = [0, 40] \text{ fs}$, this could be a the first approximation to observing the HOM deep. The measurement is not conclusive, its needed to examine the interval of $\tau = [-20, 80] \text{ fs}$ with this new range it can be determined if what is being observed is effectively the HOM deep.

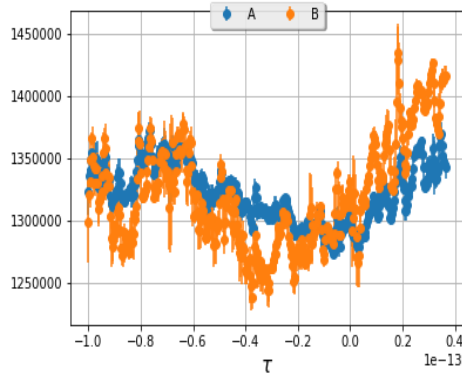


(a) Beam splitter side A output

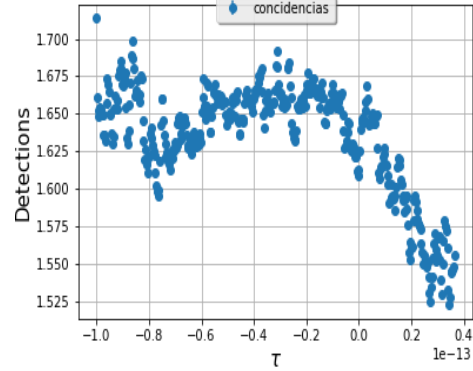


(b) Beam splitter side B output

Figure 3.6: Total Photon count per set, over the length of the measurement, detection's varied within $\sigma_1 = 23k$ detection's for arm A and $\sigma_2 = 42k$ detection's for arm B. This variation imply that there can be large fluctuation in the amount of coincidence between these two arms, since the total amount of light detected dose not stay at a constant rate.



(a) Total single photon count for A and B



(b) Normalized Coincidences between A and B

Figure 3.7: Experimental Results showing a first approximation of the HOM deep. The measurement of τ is in seconds for both plots. In b) the normalized coincidences in function of the time difference is plotted where e can see a reduction in counts in the last segment of the data.

Chapter 4

Light - Matter interaction

The aim of this chapter is to introduce the theoretical frame work and a possible experimental implementation for the interaction of a delayed photon pair discussed previously with a cesium sample. The interaction between a photon and a two level system has been studied thoroughly [32] and are of interest to many fields outside of physics. A two level system is the standard way to represent matter in quantum field theory, where one can consider the photon as an excitation in the electromagnetic field. Its with in the interest of this work to study the two photon absorption signal of a sample as a function of the delay between the interacting photons.

4.1 Two Photon Absorption *TPA*

Two-photon absorption (TPA) is the process by which a molecule or material absorbs a pair of photons, the sum of whose energy equals the transition energy. The probability that a molecule undergoes TPA when exposed to light of a given photon energy, or combination of photon energies, depends on certain intrinsic characteristics of the material and of the environment in which the molecule is immersed [33]. This information can be different from, and complementary to, what is obtainable from one-photon absorption spectroscopy, because different selection rules are applied in one photon absorption and two photon absorption.

The simultaneous absorption of two photons of energies E_1 and E_2 by a two level

system occurs when the energy $E_1 + E_2$ is in resonance with one of the electronic states of the system. As a result of the absorption process, the two absorbed photon are lost and the molecule is brought to an excited state (state f in Fig. 4.1), at $E_1 + E_2$ above the ground state (state g). This process can be thought of as an initial interaction of a photon of energy E_1 with the molecule, which is thus left in a temporary virtual state of energy E_1 above the ground state. This state exists only for a short time interval, τ_ν which has to be short than the coherence time between photon pairs. If during τ_ν a photon of energy E_2 interacts with the molecule, it can be excited to state f. The order of magnitude for τ_ν is $10^{-15} - 10^{-16}$ s for photons energies in the visible and near-IR ranges [35]. This is not sufficient per se to lead to the absorption of the two photons, symmetry selection rules also need to be satisfied for the TPA process to be allowed. These selection rules have been derived and described within the dipole approximation in many contributions in the past as shown in Ref. [36].

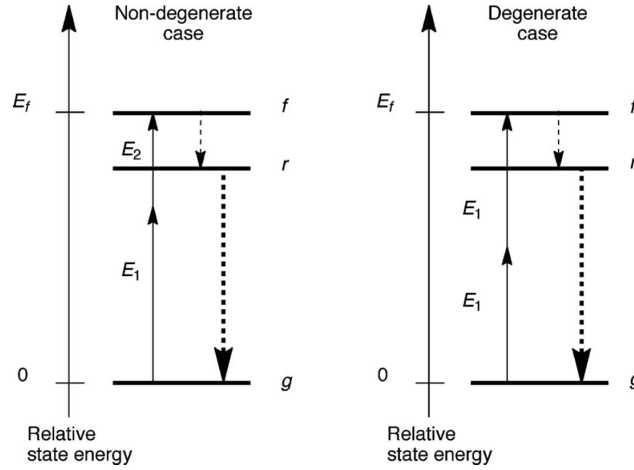


Figure 4.1: Energy level diagram showing the excitation of a two level system from the ground state to an excited state $g \rightarrow f$, located at energy E_f above the g state by the absorption of two photons (vertical solid arrows). The photons can have the same energy known as the degenerate case $E_f = 2E_1$, or different energies E_1 and E_2 nondegenerate case where $E_f = E_1 + E_2$. After the excitation, the system quickly relaxes to state r , the lowest vibronic level of the lowest-energy excited state, by internal conversion or vibrational relaxation (dashed arrow). The system finally returns to the ground state by radiative or nonradiative pathways (bold dashed arrow).

The effect of TPA can be quantified by measuring the rate of change of in the flux of photons registered, which is defined by

$$\frac{d\phi}{dz} = -\sigma N_g \phi - \delta N_g \phi^2 \quad (4.1.1)$$

where ϕ is the photon flux (number of photons per unit area and time), N_g is the number of molecules in the ground state and z is the propagation direction [37]. In Eq. 4.1.1 there are two cross section present, σ and δ where they each describe the absorption rate for a TPA process. The cross section σ describes the entangled photon flux and δ is the absorption rate for non correlated photons. From this expression one can see why there is interest in entangled two photon absorption, this is given by the linear dependence of σ with the flux, this implies that a lower flux of entangled light is need to have an absorption rate equal to the absorption rate of chaotic light (non correlated) which depends on the square of the flux . The low

flux needed for entangled photon absorption is an advantage since the lower intensity allows us to study delicate samples, nonclassical light demands a high intensity to produce a TPA signal potentially destroying delicate samples.

4.2 Virtual State Spectroscopy VSS

In the theoretical technique described by Svozilík et al. [12], it uses the fact that the probability that the medium is excited to this final state is given by

$$P_{g \rightarrow f} = \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{t_2} dt_1 \int_{-\infty}^{\infty} dt'_2 \int_{-\infty}^{dt'_2} dt'_1 M^*(t_1, t_2) G^{(2)}(t_2, t_1 : t'_2, t'_1) M(t_1, t_2) \quad (4.2.2)$$

where G^2 is the non-normalized second order coherence degree described as

$$G^{(2)}(t_2, t_1 : t'_2, t'_1) = \langle \hat{E}^{(-)}(t_2) \hat{E}^{(-)}(t_1) \times \hat{E}^{(+)}(t'_2) \hat{E}^{(+)}(t'_1) \rangle \quad (4.2.3)$$

and $M(t_1, t_2)$ describes the response of the medium to the applied fields:

$$M(t_1, t_2) = \sum_j \frac{\mu_{fj} \mu_{jg}}{\hbar} \exp[i(\epsilon_f - \epsilon_j)t_2 + i(\epsilon_j - \epsilon_g)t_1] \quad (4.2.4)$$

where μ_{fj} , μ_{jg} are the transition matrix elements of the dipole-moment operator between the states specified in the subscript. Where Svozilík et al. show that one can extract this information by measuring the changes in the absorption signal of the sample in question.

4.3 Experimental proposal implement VSS

An experimental setup is proposed for the measurement of VSS, by extending the setup shown in Fig. 3.5. This setup consists in changing the second BS from Fig. 3.5 to a Polarization beam splitter (PBS) and introducing a half wave plate to rotate the polarization of the bottom arm of the interferometer as shown in Fig. 4.2. With this experimental setup one can measure the changes in the intensity of the two photon absorption signal by the sample, as required by Svozilík. Initially and as shown in Fig. 4.2 we propose a single channel detection but this may change to a 2 channel

detection , where this is dependent in which setup gives a better signal to noise ratio.

Replacing the BS with a PBS and introducing a half wave plate are done to have all correlated photons exit threw single face of the beam splitter. With the single source the sample can be introduce, by measuring the of amount light that pass threw the sample and knowing the relative delay between the photon pairs on can obtain the TPA signal. A large set delay intervals would have to be measured, it is also necessary to have a very stable source of entangled light to have high resolution in changes in absorption rates. Due to this facts it is necessary to keep working and further calibrate the previous section of this experiment to be able to reliably measure the TPA signal.

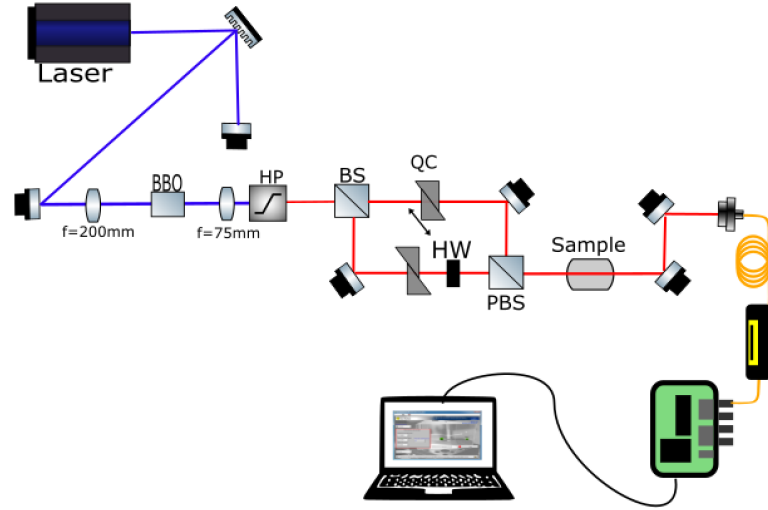


Figure 4.2: Experimental setup proposed to execute a measurement of a VSS for a cesium sample by extending the setup previously shown in Fig. 3.5. HW stand for Half wave plate and PBS is polarization beam splitter.

Chapter 5

Analysis and Conclusions

The main goal of this work was to construct a experimental setup that could be extended to a virtual state spectroscopy experiment. To achieve this we first constructed an entangled photon source and characterize its frequency and time correlation properties. A SPDC photon source construed was studied, where we specifically showed that the second order coherence degree for the photon source showed a non-classical behavior. The transverse spacial profile is in the a collinear formation, implying that a photon pair travels in the same direction as the pump photons. The frequency correlations were also studied by measuring the joint spectrum, where we saw that SPDC source has a symmetric and anti-correlated spectrum. The symmetric center wavelengths are in resonance with the atomic transitions of cesium sample, which is the one we are interested in studying in the VSS experiment.

The next step towards a measurement of the VSS in cesium sample is to produce a reproducible time delay between two correlated photons, characterizing the time differences between photons with out changing the total amount of photons detected. In order to archive this goal we built a time delay using quartz plates, but it presents difficulties at keeping a constant amount of coupled light. The measurement of the time delay produces by this mechanism was quantified by the first measurement of the HOM deep, from this measurement we can see that the oscillation of the total detected light can produce oscillations on the same scale as are signal.

To continue with this project its need to finely calibrate the produces time difference between photon pairs, also we propose an experimental setup to measure the TPA signal in a sample as is shown in Fig. 4.2 by introducing a few optical elements. This experimental technique with entangled photons can used in different application such as dealing with life samples, where conventional techniques don't work do to the need of high photon flux that can be damaged the sample. In this cases the low photon flux regimen that we are using can be very effective to achieve high sensitivity and low intensity imaging.

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Appendix A

Auxiliary Results

A.1 Beam splitter without dissipation

Lets consider a light separator or beam splitter (BS) with out dissipation. The input fields are $E_{a,in}$, $E_{b,in}$ are transformed to $E_{a,out}$, $E_{b,out}$ as follows: [24]

$$\begin{pmatrix} E_{a,out} \\ E_{b,out} \end{pmatrix} = \begin{pmatrix} \cos \theta & i \sin \theta \\ i \sin \theta & \cos \theta \end{pmatrix} \begin{pmatrix} E_{a,in} \\ E_{b,in} \end{pmatrix} \quad (\text{A.1.1})$$

Where θ is associated with the reflectivity and transmissivity of the BS as $R = \sin^2 \theta$, $T = \cos^2 \theta$. Classically if there is only on input, lets say a, which implies $E_{b,in} = 0$ producing a output filed $E_{a,out} = \cos \theta E_{a,in} < E_{a,in}$ thus producing an attenuation. Lets consider the quantum field theory approach, such that $E_a \rightarrow \hat{a}$, $E_b \rightarrow \hat{b}$, lets try to proof that the action of the BS can be described as Unitary operator $\hat{U} = e^{i\theta(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b})}$. Since its a unitary operator on has that $\hat{U}^\dagger(\theta) = \hat{U}^{-1}(\theta) = e^{-i\theta(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b})} = \hat{U}(-\theta)$. The action of this unitary operator on a is given by:

$$\begin{aligned} \hat{a}_{out} &= \hat{U}^\dagger \hat{a} \hat{U} = e^{-i\theta(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b})} \hat{a} e^{i\theta(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b})} \\ \hat{b}_{out} &= \hat{U}^\dagger \hat{b} \hat{U} = e^{-i\theta(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b})} \hat{b} e^{i\theta(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b})} \end{aligned} \quad (\text{A.1.2})$$

Applying the BCH identity:

$$e^{\lambda G} A e^{-\lambda G} = \sum_{n=0}^{\infty} \frac{\lambda^n}{n!} C_n, \quad C_0 = A, \quad C_1 = [G, C_0], \quad C_2 = [G, C_1], \dots \quad (\text{A.1.3})$$

when the main commutation computation done are

$$[\hat{a}\hat{b}^\dagger + \hat{a}\hat{b}, \hat{a}] = -\hat{b}, \quad [\hat{a}\hat{b}^\dagger + \hat{a}\hat{b}, \hat{b}] = -\hat{a}. \quad (\text{A.1.4})$$

With this one can find that there is a tendency dependent in the parity of n , if n is even $C_n = \hat{a}$ and if n is odd $C_n = i^2\hat{b}$, from here one can find that the output fields can be written as:

$$\hat{a}_{out} = \hat{U}^\dagger \hat{a} \hat{U} = \hat{a} \cos \theta + i\hat{b} \sin \theta \quad (\text{A.1.5})$$

$$\hat{b}_{out} = \hat{U}^\dagger \hat{b} \hat{U} = \hat{b} \cos \theta + i\hat{a} \sin \theta \quad (\text{A.1.6})$$

thus one can write the action of $\hat{U}$ as a matrix

$$\begin{pmatrix} \hat{a}_{out} \\ \hat{b}_{out} \end{pmatrix} = \begin{pmatrix} \cos \theta & i \sin \theta \\ i \sin \theta & \cos \theta \end{pmatrix} \begin{pmatrix} \hat{a} \\ \hat{b} \end{pmatrix} \quad (\text{A.1.7})$$

Now we can see that this is the same matrix transformation seen in Eq. A.1.1, thus the action of the BS can be described in quantum field theory as unitary operator. Further more, let's see what is the effect of the operator over a single input $|\Psi\rangle_{in} = |1\rangle_a \otimes |0\rangle_b$

$$|\Psi_{out}\rangle = \hat{U} |\Psi_{in}\rangle, \quad \hat{U} = e^{i\theta(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b})} = \sum_{m=0}^{\infty} \frac{(i\theta)^m}{m!} (\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b})^m \quad (\text{A.1.8})$$

thus is of interest to compute

$$(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b}) (|1\rangle_a \otimes |0\rangle_b) = |0\rangle_a \otimes |1\rangle_b \quad (\text{A.1.9})$$

$$(\hat{a}\hat{b}^\dagger + \hat{a}^\dagger\hat{b}) (|0\rangle_a \otimes |1\rangle_b) = |1\rangle_a \otimes |0\rangle_b \quad (\text{A.1.10})$$

One can show that if m is odd then the produce output state is $|0\rangle_a \otimes |1\rangle_b$ and if m is even one gets the output state $|1\rangle_a \otimes |0\rangle_b$. With this one can write the total output state of the BS as

$$|\psi_{out}\rangle = \cos \theta |1\rangle_a \otimes |0\rangle_b + i \sin \theta |0\rangle_a \otimes |1\rangle_b \quad (\text{A.1.11})$$

Now from Chapter 2 we can tell that this is an entangled state, while the input state can be considered as a classical field. Now let's find the action of the BS over a two input state $|1\rangle_a \otimes |1\rangle_b$, but now let's use a slightly different approach

for the execution of this calculation. Since $\hat{U}$ is a unitary operator it follows that $\hat{U}\hat{U}^\dagger = \hat{U}^\dagger\hat{U} = I$, let us note that:

$$|\Psi_{out}\rangle = \hat{U} |\Psi_{in}\rangle = \hat{U} |1\rangle_a \otimes |1\rangle_b = \hat{U} |1, 1\rangle = \hat{U}\hat{a}^\dagger |0, 1\rangle. \quad (\text{A.1.12})$$

Now lets use the fact or the operator unitary properties and apply the same operation to the input b

$$\hat{U}\hat{a}^\dagger |0, 1\rangle = \hat{U}\hat{a}^\dagger\hat{U}^\dagger\hat{U} |0, 1\rangle = \hat{U}\hat{a}^\dagger\hat{U}^\dagger\hat{U}\hat{b}^\dagger\hat{U}^\dagger\hat{U} |0, 0\rangle \quad (\text{A.1.13})$$

Note that $\hat{U} |0, 0\rangle = |0, 0\rangle$ and lets use Eq. A.1.2

$$\hat{U}\hat{a}^\dagger\hat{U}^\dagger\hat{U}\hat{b}^\dagger\hat{U}^\dagger |0, 0\rangle = (\hat{a} \cos \theta + i\hat{b} \sin \theta)(\hat{b} \cos \theta + i\hat{a} \sin \theta) |0, 0\rangle \quad (\text{A.1.14})$$

after some algebra and careful computation one gets that the total output state of the beam splitter is

$$|\Psi_{out}\rangle = (\cos^2 \theta - \sin^2 \theta) |1, 1\rangle + i\sqrt{2} \sin \theta \cos \theta (|2, 0\rangle + |0, 2\rangle) \quad (\text{A.1.15})$$

$$|\Psi_{out}\rangle = (T - R) |1, 1\rangle + i\sqrt{2} \sin \theta \cos \theta (|2, 0\rangle + |0, 2\rangle) \quad (\text{A.1.16})$$

The physical analysis of this wave function is executed in Sec.3.1.1. In a 50:50 BS $\theta = \frac{\pi}{4}$ giving us a theoretical model for the wave function of the HOM effect:

$$|\Psi_{out}\rangle = \frac{i}{\sqrt{2}} (|2, 0\rangle + |0, 2\rangle) \quad (\text{A.1.17})$$