Single Photon Sources with Surface Plasmons

Angelika Ghazale

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Dr. Nancy Rahbany

Lebanese American University

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Abstract. Single photon sources play a crucial role in the leading quantum technologies however, there is still no ideal single photon emitter on demand. In this study, we will visit the concept of single photon sources, surface plasmon polaritons (SPPs) and the coupling of surface plasmons with the single photon emitters to discuss the effects on the emission of single photons and its efficiency. First, we will prove the existence of single photon sources from the Mandel Dip experiment and the second order correlation. Then we will study the effects of coupling SPPs with single photon emitters through photon counting. The time elapsed between two consecutive emitted single photonsor the lifetime is our predictor of efficiency. As the results show that when the emitters were coupled, the time decreased for the emission of a photon after the system returned to its de-excitated state.

Keywords: Single Photon Sources, Surface Plasmons, Coupling.

1. Introduction

New research has been studying non- conventional light sources to produce streams of photons with adjustable quantum correlation, in particular single photon emitters (SPEs). These SPEs play a central role in several quantum computing aspects such as quantum walks, quantum simulation, and precise measurement [1]. Moreover, SPEs can also be useful in secure communication and optics. The use of these quantum objects allows for a more efficient and secure outcomes. Hence the ideal choice for this application is encoding the information in the quantum states of the photon through momentum, energy, or polarization. This is because photons can be manipulated with linear optics and travel at light speed causing little noise and loss. The definition of a photon as the quantum of energy $h\nu$ is crucial to the quantum revolution, it branches from the quantization of the electromagnetic field, which implies the excitation of a single discrete energy mode [2].

There has been considerable effort to develop single photon sources and the first model was using atomic transition of sodium atoms demonstrated by Kimble et al. in 1977 [1]. In recent years, advancement in single photon sources has been made yet, efficiency, reliability and the probability of absorption are still prone to improvements. The absorption probability when aiming the photon towards the single emitter is very low and achieving high probabilities is challenging. In order to enhance these single photon sources, the emitter is placed inside a cavity where the photon will bounce continuously to increase the absorption probability. However, this technique is limited to narrowband photons and emitters. So, the optimal way of achieving higher absorption probabilities is to place the emitter within a plasmonic mode or what we are concerned with, surface plasmon polaritons.

1.1 Ideal Single Photon Sources

An ideal single photon source is when a single photon is emitted at any defined time. The probability of a single photon emission is 100% and the probability of emitting multiple photons is 0%. The photons emitted are indistinguishable and the rate of emission or pulse is very fast meaning that the duration between two consecutive photons emitted is small. It is highly photostable with defined spatial and spectral properties. Such source must have a lifetime in the sub-nanosecond and should be polarized in both emission and absorption. In

reality, single photon emitters deviate from these ideal characteristics, but suitable emitters have been developed to meet some of the above criteria [2].

1.2 Characteristics of Single Photon Emitters

The quality of a single photon source is measured by the purity of the emitted photon, emission of one photon per time frame and the efficiency of the rate at which the single photon is emitted. A single photon source is characterized by the second order correlation function $g^{(2)}(\tau)$ which verifies the emission of a single photon [2].

A single photon source must have $g^{(2)}(0) = 0$ as this is the quantity that is correlated with the probability of multi-photon emission at a specific time. Moreover, to measure the efficiency of the single emitter the system is modeled to determine the luminescence lifetime [2]. In the following sections, we will discuss in detail these two aspects of the single photon sources.

1.3 Single Photon Source Systems

The operation of a single photon emitter is simple and requires an external force to excite the system into its excited energy state. While the de-excitation to a lower energy state occurs, the system will emit a single photon.

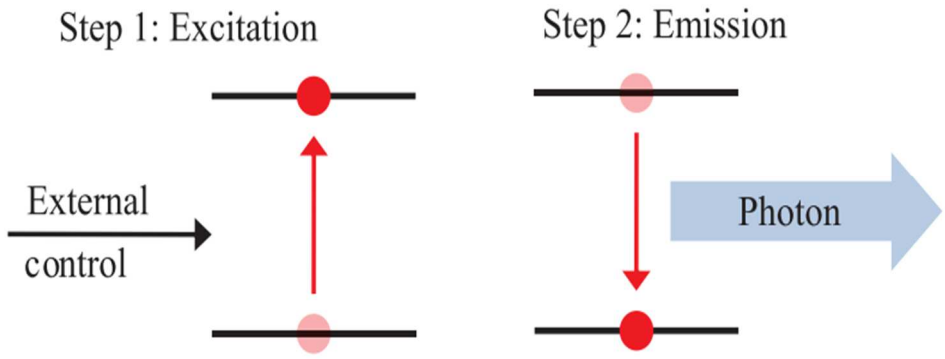

Figure 1: Excitation and de-excitation process [2].

There are numerous systems that have been developed to produce single photon sources such as single neutral atoms, single ions, single molecules, quantum dots (QD) and

color centers. However, they all have the same concept of excitation, de-excitation system [2].

2. Types of Single Photon Sources

2.1 Single Neutral atoms

A single neutral atom can emit only one photon at a time. A single photon is generated when a laser pulse is applied to a trapped atom. When a single atom is put between two reflective mirrors (known as a cavity) all the photons will be sent in the same direction [10].

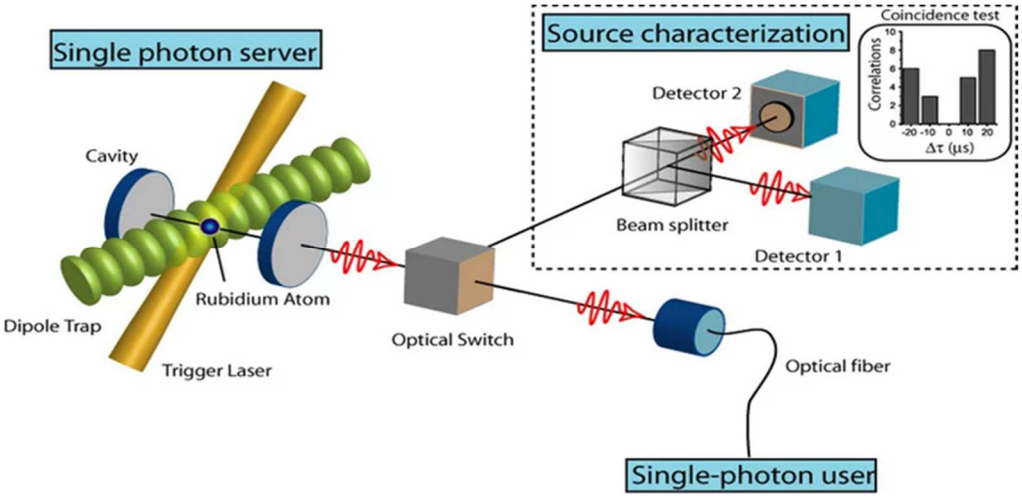

Figure 2: SPE generated by a single atom [10].

2.2 Single Ions

Single ions for generating single photon source are similar to single atoms where the single ion is trapped in a cavity and an outside source excites the ion in order to emit a single photon [4].

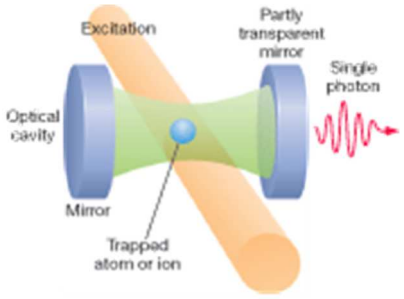

Figure 3: Excitation of an ion trapped in a cavity [4].

2.3 Single Molecules

A single molecule system consists of three states, a ground state, first excited state and an intermediate state. The electron is excited to the first excited state and then it is de-excited to its ground state emitting a single photon [7]. This single photon emission occurs in every cycle of an electron in its ground state to excited state then back to the ground state until the electron is transitioned to the intermediate state.

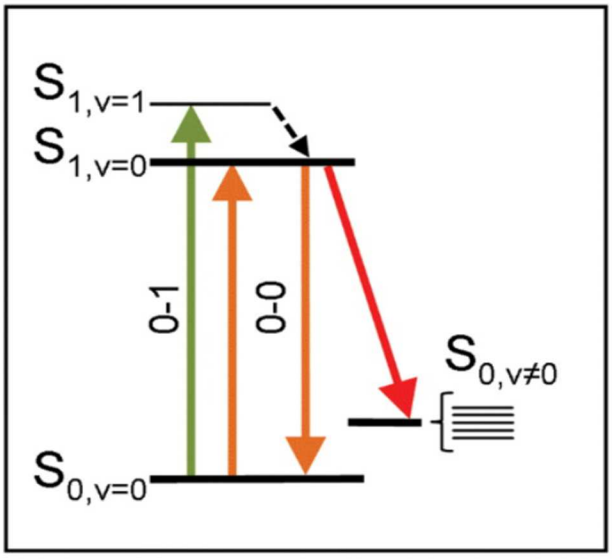

Figure 4: Three state system of a single molecule [7].

2.4 Quantum Dots (QD)

A single quantum dot confined inside a cavity, acts as a SPE when a laser pulse excites an exciton (electron and electron hole bound by Coulomb force) in the quantum dot [14]. The exciton decays which leads to the emission of a single photon.

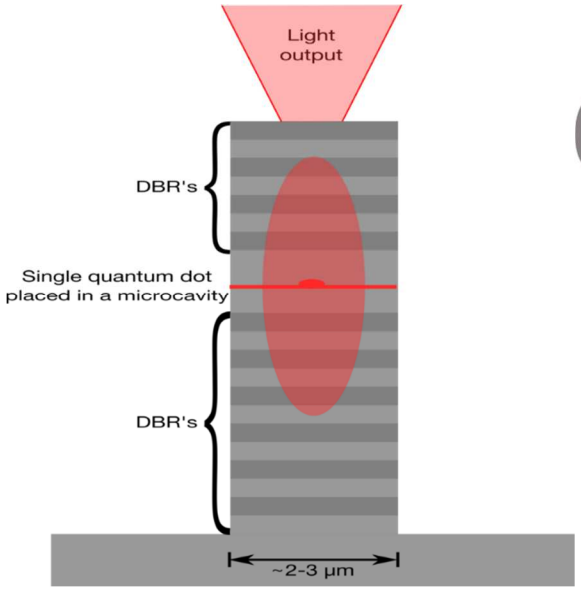

Figure 5: Single quantum dot in microcavity [14].

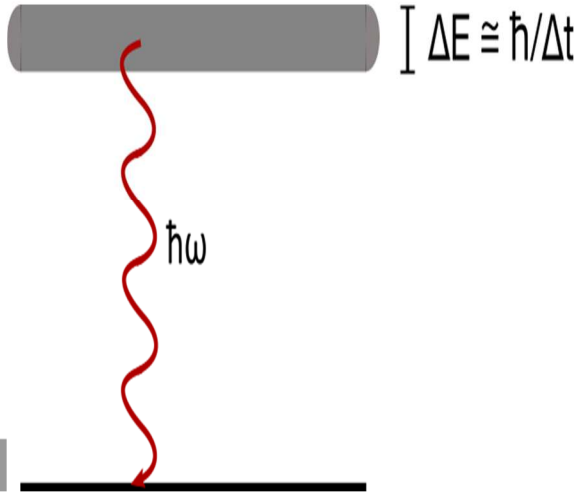

Figure 6: Emission of a single due to the decay [14].

2.5 Color Centers

Color centers are crystal defects which introduce additional light absorption and/or emission features in crystalline materials. At a certain lattice position, an electrically negative ion is missing, and electrons fill the vacancy, while being trapped by surrounding positive ions. In other cases, ions migrate to an unusual interstitial position [13]. The color center acts as an emitting dipole at room temperature with strong damping coherence and modeled by a two-level system.

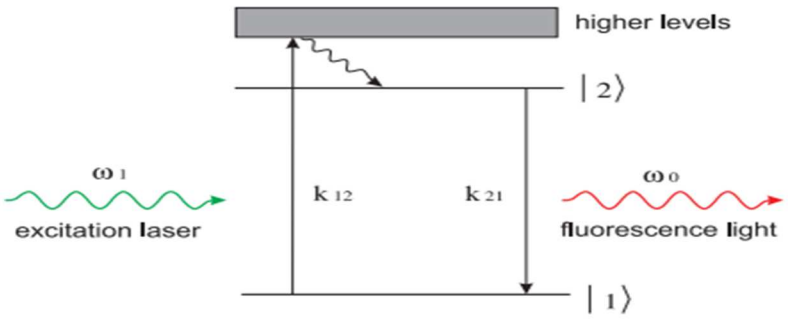

Figure 7: Energy Levels for Color Centers [13].

The system with an energy difference of hw_0 is illuminated by a laser with a frequency w_1 ($w_1 > w_0$). When a photon is absorbed, the system is excited from state 1 to a higher level. From the higher level the system relaxes to state 2, then decays to state 1 by emitting a photon with frequency w_0.

Hence, single color center cannot emit more than one photon at one time. The photons emitted are separated by a time constant, therefore this system generates a single photon source.

3. Surface Plasmon Polaritons (SPPs)

3.1 Background of Plasmonic

Surface plasmons were first predicted in 1957 by Rufus Ritchie. In the 1960s and 1970s surface plasmons (SPs) were studied extensively especially the excitation techniques specifically prism coupling which was developed by Kretschmann and Otto [8].

In general, surface plasmons (SPs) can be thought of as a type of surface wave, that is guided along the interface. The plasmonic field embodies the interaction between free electrons in a metal and electromagnetic field. Specially, free electrons in a conductive medium or a dielectric can be induced to oscillate by the electric component of a wave [11]. Classically, the plasmon is described as the oscillation of free electron density with respect to fixed positive ions in a metal. Electrons surrounding the positive nucleus inside a bulk metal will be displaced to one side of the core until the field inside the metal is canceled in the presence of a uniform electric field. When a shift or change in electric field is made, electrons move back to the center of the core and then to the opposite side [8]. The electrons will keep oscillating until the energy is lost

due to damping or resistance. The bulk plasma frequency is known as the natural harmonic frequency of this oscillating motion and a plasmon is a quantization of this oscillation. In order to excite plasmons, the free electrons within the bulk plasma should be coupled to an electromagnetic wave with a driving frequency near the plasma frequency [6]. SPs have energy lower than bulk plasmons which quantize the longitudinal electron oscillations surrounding the positive ion cores inside the bulk electron plasma. Since they have shorter wavelength than the incident light exciting them, SPPs have smaller spatial confinement and higher field intensity at the interface.

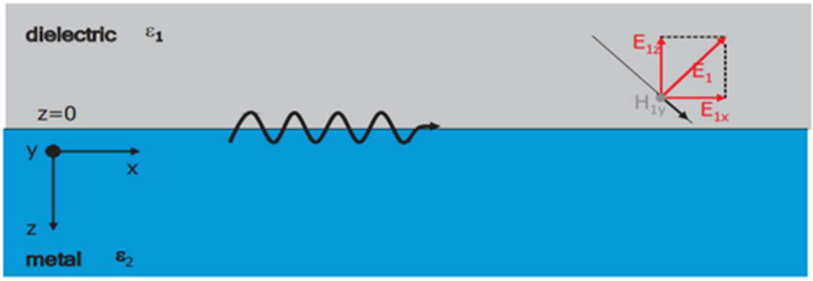

Figure 8: Interface between a dielectric and a metal in the x-y plane [8].

3.2 Geometry of SPPs

To derive these magnetic excitations along the interface we start using Maxwell's equations solving separately for the metal and the dielectric.

Maxwell's equations [6]:

$$\nabla . D = \rho$$

$$\nabla . B = 0$$

$$\nabla \, x \, E = -\frac{\partial B}{\partial t}$$

$$\nabla \, x \, H = J + \frac{\partial D}{\partial t}$$

These equations connect the charge density ρ and current density J with the electric field E, the dielectric displacement D, the magnetic induction B and the magnetic field H. Where $D =$

$\epsilon_0 \epsilon E,\ B = \mu_0 \mu H,\ J = \sigma E$ for homogeneous, linear, nonmagnetic, and isotropic medium (ϵ_0, $\mu_0\ and\ \sigma$ are the permittivity, permeability and conductivity of free space respectively) [6].

ϵ is the dielectric constant which is dependent on the frequency $\epsilon = \epsilon(w)$ and is a complex function $\epsilon = \epsilon' + i\epsilon''$ that is related to the complex index of refraction $n = n + i\kappa = \sqrt{\epsilon}$.

From the following we can obtain these relations [8]:

$$\epsilon' = n^2 - \kappa^2 \rightarrow n^2 = \frac{\epsilon'}{2} + \frac{1}{2}\sqrt{\epsilon'^2 + \epsilon''^2}$$

$$\epsilon'' = 2n\kappa \rightarrow \kappa = \frac{\epsilon''}{2n}$$

The real part of the index of refraction $n(w)$ determines the dispersion and the imaginary part $\kappa(w)$ is responsible for the absorption.

Since we are treating the SPPs at a flat interface, both the charge density and current density are not present, and the wave equation becomes [6]: $\nabla\left(-\frac{1}{\epsilon}E.\nabla_E\right) - \nabla^2 E = \epsilon_0\mu_0\epsilon(\frac{\partial^2 E}{\partial t})$

The wave equation simplifies into $\nabla^2 - \frac{\epsilon}{C}\frac{\partial^2 E}{\partial t} = 0$ since we have a homogeneous material. Assuming harmonic time dependence in the electric field, the equation is reduced to:

For the transverse electric modes (TE), the governing field components are derived from the wave equation $\frac{\partial^2 E_y}{\partial z^2} + ({k_0}^2 - \beta)E_y = 0$ [6]:

For z > 0:

$$E_y(z) = A_2 e^{i\beta x} e^{-k_2 z}$$

$$H_x(z) = -iA_2 \frac{1}{w\mu_0} k_2 e^{i\beta x} e^{-k_2 z}$$

$$H_z(z) = A_2 \frac{\beta}{w\mu_0} e^{i\beta x} e^{-k_2 z}$$

For z < 0:

$$E_y(z) = A_1 e^{i\beta x} e^{k_1 z}$$

$$H_x(z) = iA_1 \frac{1}{w\mu_0} k_1 e^{i\beta x} e^{k_1 z}$$

$$H_z(z) = A_1 \frac{\beta}{w\mu_0} e^{i\beta x} e^{k_1 z}$$

At the interface, E_y and H_x are continuous. Hence it leads to the following condition:

$$A_1(k_1 + k_2) = 0$$

However, in order to satisfy the confinement at the surface, it requires the Re(k_1) < 0 and Re(k_2) > 0. This condition can only be true if $A_1 = 0$ implying that $A_1 = A_2 = 0$. As a result, this disproves the existence of TE modes at the surface, thus surface plasmon polaritons can only exist for transverse electric modes (TM).

The TM solutions for the field components are derived from the wave equation $\frac{\partial^2 H_y}{\partial z^2} + ({k_0}^2 - \beta)H_y = 0$ [6]:

For z > 0:

$$H_y(z) = A_2 e^{i\beta x} e^{-k_2 z}$$

$$E_x(z) = iA_2 \frac{1}{w\varepsilon_0 \varepsilon_2} k_2 e^{i\beta x} e^{-k_2 z}$$

$$E_z(z) = -A_1 \frac{\beta}{w\varepsilon_0 \varepsilon_2} e^{i\beta x} e^{-k_2 z}$$

For z < 0:

$$H_y(z) = A_1 e^{i\beta x} e^{k_1 z}$$

$$E_x(z) = -iA_1 \frac{1}{w\varepsilon_0 \varepsilon_1} k_1 e^{i\beta x} e^{k_1 z}$$

$$E_z(z) = -A_1 \frac{\beta}{w\varepsilon_0 \varepsilon_1} e^{i\beta x} e^{k_1 z}$$

The wave vector perpendicular to the interface is $k_i = k_{z,i}(i = 1,2)$ and the continuity at the interface of H_y and $\varepsilon_i E_z$ requires $A_1 = A_2$ and yields $\frac{k_2}{k_1} = -\frac{\varepsilon_2}{\varepsilon_1}$.

For a solution bound to the interface, the components of $k_{z,i}$ should be imaginary and opposite signs. This yields to an exponential decaying field satisfied only when $\varepsilon_1 = -\varepsilon_2$.

Hence surface plasmon polaritons can only exist at the interface between a dielectric ($\varepsilon_1 > 0$) and a metal ($\varepsilon_2 > 0$) [8].

The figure below depicts the geometry of the electric field at the interface between the dielectric and a metal which shows an exponential decay in both directions.

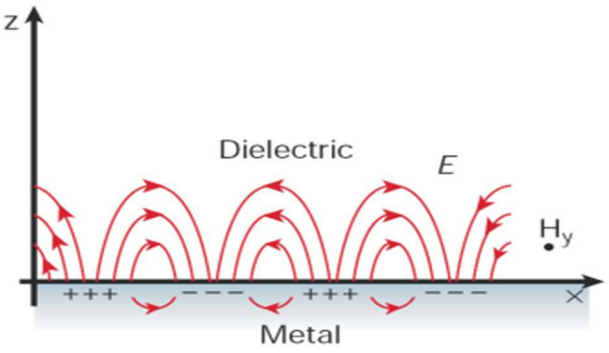

Figure 9: Geometry of electric field at the interface [8].

3.3 Dispersion Relationship for Surface Plasmons

Using the above TM solutions and the condition for continuity $\frac{k_2}{k_1} = -\frac{\varepsilon_2}{\varepsilon_1}$, the wave vector should obey the following relationship [8]:

$$k_1^2 = \beta^2 - k_0^2\varepsilon_1$$

$$k_2^2 = \beta^2 - k_0^2\varepsilon_2$$

From the wave vector relation, we obtain the SPP dispersion relation: $\beta = k_0\sqrt{\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1+\varepsilon_2}}$

Where $k_0 = \frac{w}{c}$ is the wave vector of light in vacuum and $\varepsilon_1, \varepsilon_2$ are the frequency dependent relative permittivity of the two medium.

For the sake of this paper, we will only take two special cases to study their dispersion relation. We will consider a surface plasmon polariton propagating on an air-gold interface and on an air-silver interface. The permittivity values for both the gold and silver metals were given as real and imaginary parts along with the values of the wavelength. Using these values and the dispersion relation, a figure on MATLAB was generated.

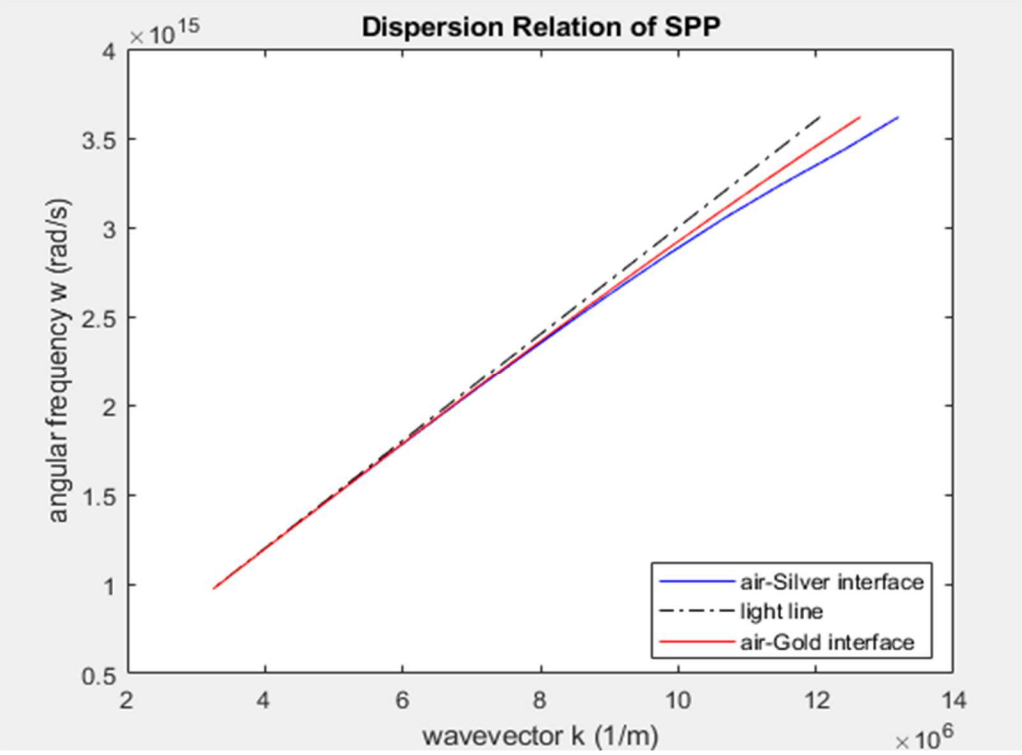

Figure 10: Dispersion relation of SPP compared to light.

From the figure, we realize that the dispersion relation of SPP propagating on both the air-gold and air-silver interface lie below the free space light line such that β > k. Thus, SPPs cannot be excited by the direct light illumination since the conservation of energy and momentum cannot be achieved at the same time. Hence, in order to excite SPPs, a transfer of momentum has to occur through special techniques such as prism coupling.

3.4 Prism Coupling

It is a phase matching technique which excites the plasmon at a metal/dielectric interface since there exists no angle such that the momentum condition $k_x = ksin\Theta$ that matches the SPP constant of propagation β [6]. The technique is a three-layer system of two dielectric medium with different dielectric constants and a thin metal film in between. This film is evaporated on a glass prism and illuminated at an angle greater than the angle of total internal reflection. When the photon wavevector coincides with the SPP wavevector in the prism on air-metal surface, light tunneling resonance occurs in the metal film and the surface polaritons are coupled with light $\beta = \frac{w}{c}\sqrt{\varepsilon_{prism}}sin\Theta$. As the thickness of the film is increased, the efficiency of the

excitation of SPP decreases because the tunnelling distance increases. Illustrated in the figure are the two configurations Kretschmann and Otto that are able to achieve the coupling. In the kretschmann configuration, a thin film is placed on the hypotenuse of the prism. Photons from the glass side tunnel at a larger angle than the critical angle of the internal reflection through the metal film and then excite the SPPs at the interface. Whereas in the Otto configuration, there is an air gap between the prism and the metal and total internal reflection occurs at the interface of the air/prism and excites the SPP at the metal/ air interface through tunneling.

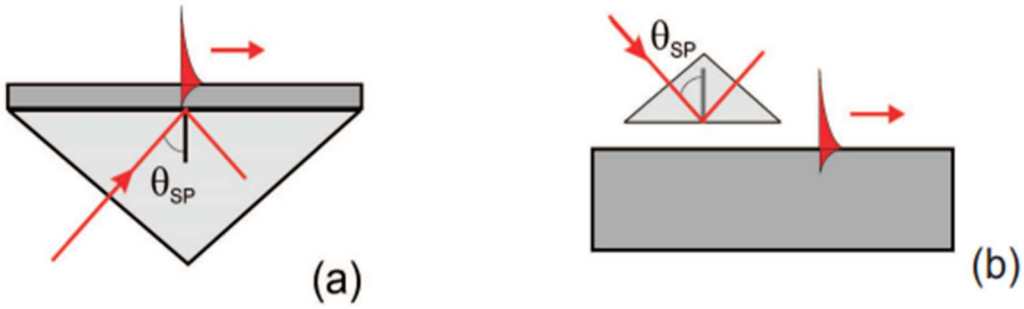

Figure 11: (a) Kretschmann configuration, (b) Otto configuration [8].

4. Photon Counting

4.1 Second Order Correlation Function

The second order correlation function is important for the characterization of quantum states as it describes the correlation between the intensities of the field between two points. Thus, this function is used for testing how good the single photon source is. For stationary fields and temporal domain, the second order correlation function in the classical theory is given by [9]:

$$G^{(2)}(\tau) = \langle E^{(*)}(0)E^{(*)}(\tau)E(\tau)E(0)\rangle$$

And the normalized form [13]:

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

Where $\langle . \rangle$ is the average over a long-time interval, $E(t), E^{(*)}(t)$ are the amplitude of electronic field at a time t and the conjugate respectively and $I(t)$ is the intensity time dependent.

In the quantum theory, the second order correlation function needs quantization of the electromagnetic field. The electromagnetic field with a polarization and a wavevector is described as a superposition of the number states. And the energy of the photon $h\nu$ is directly linked with the number states, so the number states contain n photons. to detect a photon, it is equivalent to the transition between the states of the detector. Hence, the formula of the second order correlation function in the quantum theory becomes [13]:

$$g^{(2)}(\tau) = \frac{\langle E^-(t)E^-(t+\tau)E^+(t+\tau)E^+(t)\rangle}{\langle E^-(t)E^+(t)\rangle\langle E^-(t+\tau)E^+(t+\tau)\rangle} \qquad (2)$$

Where E^+ and E^- are the positive and negative frequency field operators respectively. And the t and $t+\tau$ are the correlated detection time of two consecutive photons corresponding to the matrix element $E^+(t+\tau)E^+(t)$.

A deterministic single photon source should have $g^{(2)}(0) = 0$ since this quantity is directly related to the probability of more than one photon at a time. So, in the classical field, the single photon source quality is measured by the intensity correlation function (1), and in the quantum theory, the quality is measured by the probability coherence function (2).

4.2 Mandel Dip Experiment

The Mandel dip experiment measures the coincidence of photon pairs based on HOM (Hong-Ou-Mandel) interferometer. It is the second order coherence effect caused by the combination of the probability that the two photons are transmitted and reflected by the beam splitter. If both the idler photon pair and the signal arrive at the beam splitter with the same phase on the same time, one detector in the interferometer detects the photons. The coincidence of photon pairs under these conditions is zero. If the phase is changed at the interferometer's path, the probability to detect the photons from different ports is not equal to zero i.e., the coincidence for a pair of photons is no longer zero. This experiment will measure the degree of indistinguishability and interference visibility of photons emitted from single photon sources for the purpose of single photon counting [3].

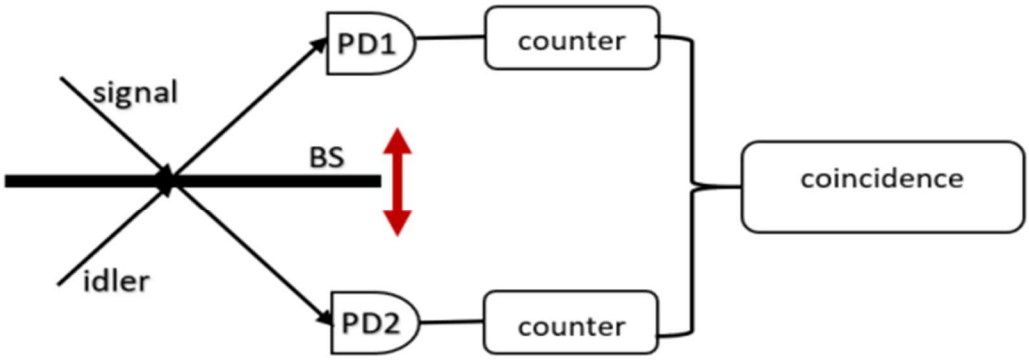

Figure 12: Mandel Dip Experiment set up [3].

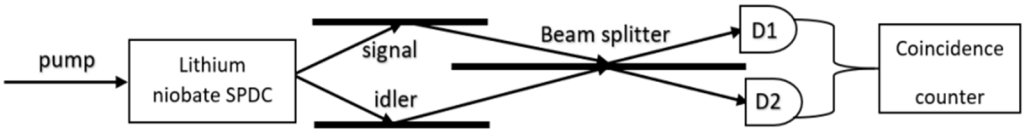

Figure 13: HOM interferometer [3].

To simplify the experiment, we assume that the angle of incidence is 45 degrees, and the photons have identical physical properties. The derived expected photon number recorded by the counter of coincidence is [3]: $N_c = C[R^2 + T^2 - 2RT.\frac{\int_{-\infty}^{\infty} g(\tau)g(\tau-2\Delta\tau)d\tau}{\int_{-\infty}^{\infty} g(\tau)^2 d\tau}]$

Where C is a constant, T is the transmissivity and R is the reflectivity.

Special Case

Taking a special case in which $g(\tau)$ is the Gaussian function $g(\tau) = e^{\frac{-(\Delta w\tau)^2}{2}}$ (Δw is the bandwidth) the equation of photon number reduces to: $N_c = C(R^2 + T^2)[1 - \frac{2RT}{R^2+T^2}.e^{\frac{-(\Delta w\Delta\tau)^2}{2}}]$.

For $\Delta\tau = 0\ and\ R = T = 50\%$, $N_c = 0$. However, if $R + T < 1$, the $\frac{2RT}{R^2+T^2}$ factor will not become equal to 1 and the term $\frac{2RT}{R^2+T^2}.e^{\frac{-(\Delta w\Delta\tau)^2}{2}} \neq 1$ even if $\Delta\tau = 0$.

We will consider several values for T and R and calculate the value of

$N_c = C(R^2 + T^2)[1 - \frac{2RT}{R^2+T^2}.e^{\frac{-(\Delta w\Delta\tau)^2}{2}}]$ for $\Delta\tau = 0$.

The visibility of the HOM dip measures the degree of single photon interference. So, for nonideal single photon sources, the HOM interference visibility is reduced. Then we will plot the photon number count and the photon coincidence distribution for different values of $R + T$ to see how the visibility changes.

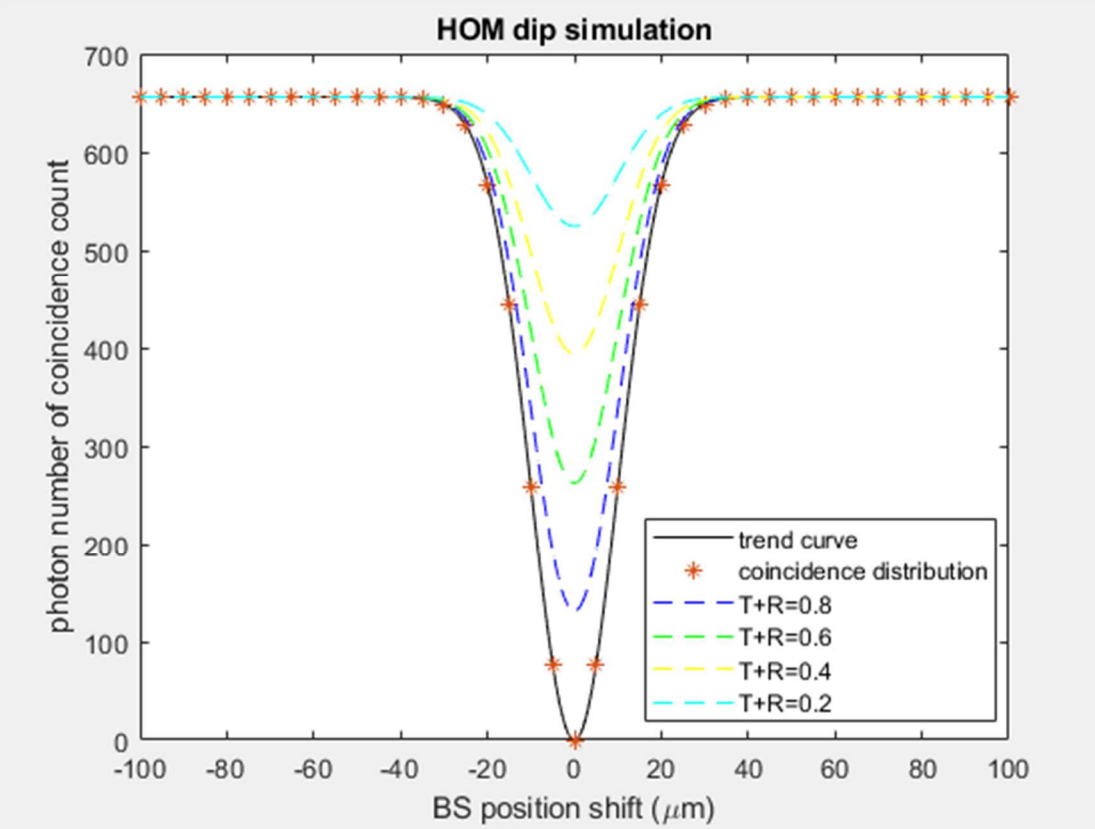

Figure 14: Plot of photon distribution for different values of T+R

The figure shows that as we decrease the values for T+R in other words we are increasing the loss of the transmittivity and reflectivity, the coincidence increases for the two detectors to detect the photons. Such coincidental detection reduces the HOM interference visibility which is directly related to the single photon indistinguishability. Consequently, as the loss increases from the transmittivity and reflectivity, the visibility of photon decreases. Therefore, there is an inverse proportionality between the visibility of the photon and the losses in the reflectivity and transmittivity, proving that as the losses increase, we are less likely to detect a single photon emitted (The dip decreases with the losses). And as the losses decrease, it is more likely to detect a single photon (steeper dip).

4.3 Lifetime and Efficiency

After studying thoroughly, the single photon sources or SPEs and surface plasmons separately, we will examine the effect when combining the two and explore the benefits of coupling the surface plasmon polaritons to the single photon emitters for the aim of the study. In order to

discuss the effects of the coupling, we will first consider exciting a single photon emitter on its own by an external pulse and record its lifetime. We will then couple an emitter with the single photon source and examine what will change [12].

The method for photon counting is a time resolved fluorescence spectroscopy based on repetitive, accurate timed registration of single photons upon excitation by a short flash of light or a laser pulse. To register a photon, the difference between the excitation pulse and the photon event is measured. It would count the single photons emitted and the time elapsed between two consecutive emitted single photons. Hence, this technique determines the lifetime of the single photon emitter which is the time the photon emitter spends in its excited state before it returns to the ground state and emit a single photon. The recorded values for the emissions are plotted with respect to time and then fitted exponentially to obtain the graph below for single photon emitters.

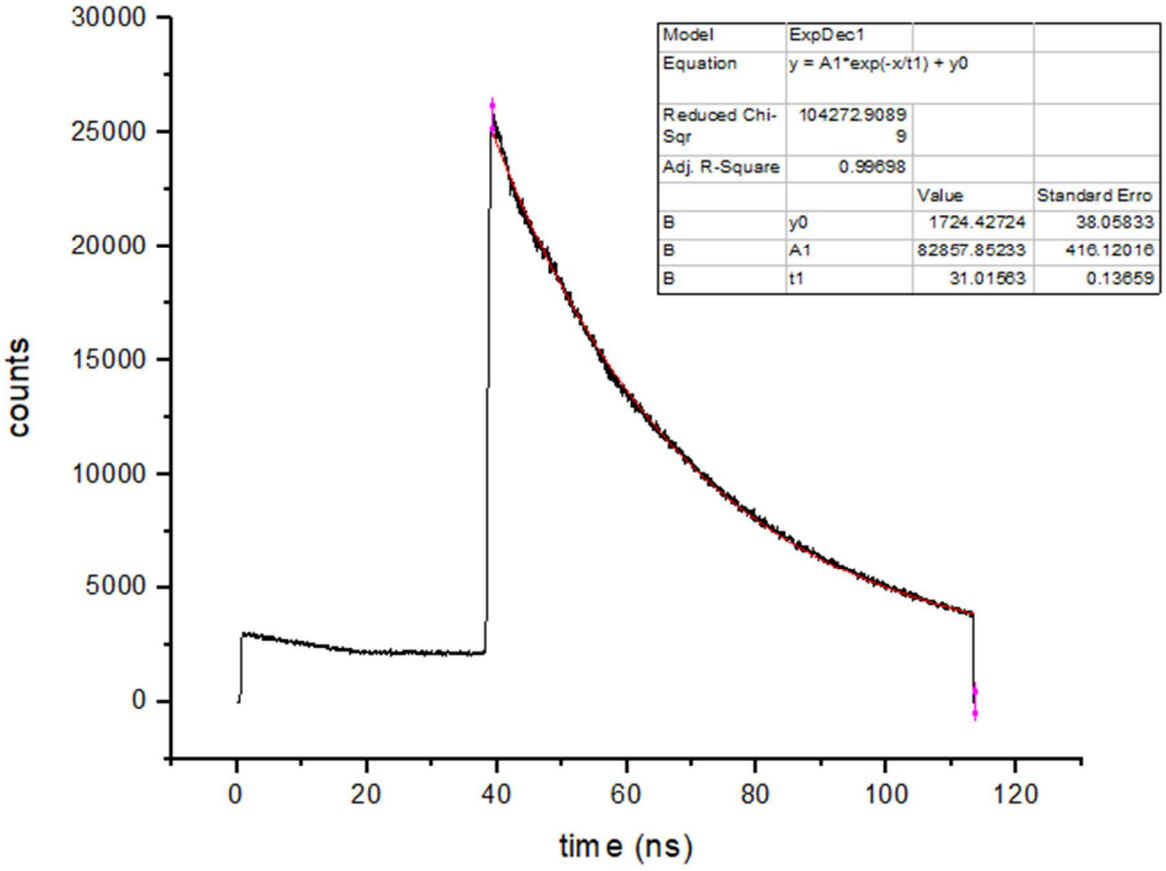

Figure 15: Emission for Single photon emitters without coupling.

The obtained data from the figure above is shown in the table where the lifetime is recorded and is obtained to be 31.01563 ns for the single photon emitter on its own. This lifetime value indicates the time the single photon emitter spends in the transition from its excited state to its ground state to emit a single photon. So, every 31.01563 ns the single photon emitter, emits only one photon.

Now we couple the single photon source with surface plasmon polaritons SPPs and proceed with the same process to extract the lifetime for the emitter after the coupling.

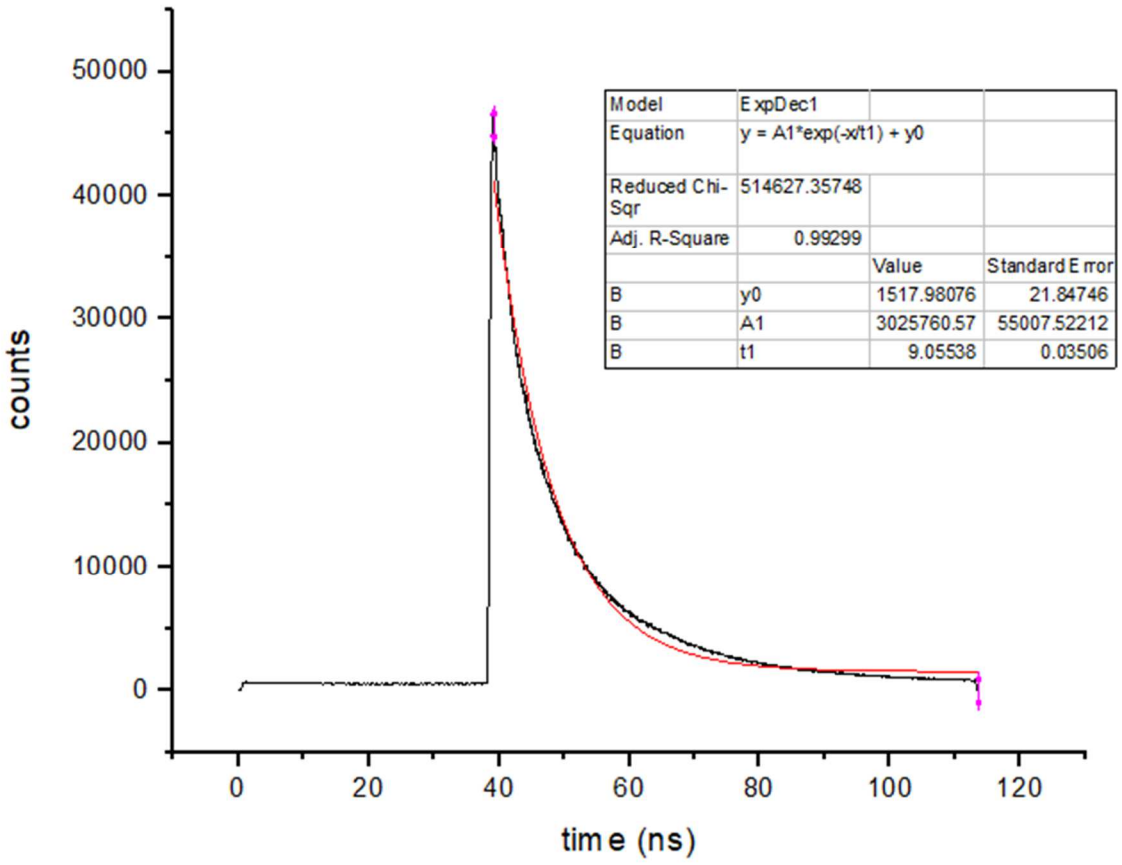

Figure 16: Emission for Single photon emitters with coupling.

The graph obtained, shows a major improvement, and records a value 9.05538 ns for the lifetime of the coupled single photon emitter. The lifetime of the single photon emitter

decreased by a value of $(31.01563 - 9.05538)\ \text{ns} = 21.96025\ \text{ns}$ when coupled by SPPs, which is equivalent to a 70.8% decrease from the initial lifetime value and so this is an indication of improved efficiency. The efficiency of the photon emitter improved by a 70.8% when coupled, which results in a better single photon emitter. This shows how beneficial coupling single photon sources with SPPs is and validates the importance of coupling process for improving the efficiency and the enhancement of these emitters.

5. Conclusion

The subject of single photon sources is important for several applications in developing new technologies nowadays. Although the basics of single photon emitters have been established, but such complex sources still require enhancements. For this paper, the aim was to study single photon sources coupled with surface plasmon polaritons and examine how the efficiency of these emitters improve with the coupling. The single photon systems were described along with its types such as single neutral atoms, single ions, single molecules, quantum dots (QD) and color centers and a brief geometry of surface plasmon polaritons was included. The other part of the study was to prove the existence of such emitters through the second order correlation function and the Mandel dip experiment which showed that as the dip decreases meaning that the losses in the reflectivity and transmittivity increase, the detection of a single photon decreases and vice versa. As for the aim of the study, the research efforts have shown that coupling the surface plasmon polaritons improved the efficiency of the source drastically by more than 70%. This was concluded from the lifetime extracted from the graphs (figure 15-16), plotting the counts of emitting a single photon with respect to time first when uncoupled and then when coupled. The lifetime for the uncoupled single photon emitters was obtained to be 31.01563ns whereas for the coupled it was 9.05538ns. As a result, when the emitter is coupled, it emits a photon every 9.05538ns which is almost 21 times less than uncoupled. This proves the efficient outcome of coupling SPPs to single photon emitters.

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