Single Photon Emitters

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Abstract:

In this report one of the fundamental ingredients needed to make quantum cryptography and other quantum information applications work, single photon emitters, is discussed. The ideal single photon emitter should produce light pulses on demand which contain exactly a single photon. Besides this, they should preferably work at room temperature and be easy to produce. In this report first a short introduction is given to why these sources are important in what is perhaps the first quantum information application which will be commercially used: quantum cryptography. Next, an overview is given of several single photon emitters based on different principles utilizing quantum dots, wells, molecules and colour centres. It is then speculated which single photon emitter is most promising to produce on a mass scale for everyday use. It turned out that all structures should in principle be able to produce ideal single photon emitters but there are differences in the difficultly of producing them on a bigger scale. Although quantum dots and wells have been proven to work around 0 K temperature and are easy to produce, it will be difficult to realize single photon emitters this way which work at room temperature due to phonon emission in the bulk. The problem with molecules and colour centres remains that they are not very easy to control on a bigger scale.

Contents

Introduction 3
Introduction to Quantum Cryptography 4
2.1 Modern Cryptography and the Key Distribution Problem 4
2.2 Quantum Key Distribution 5
Single Photon Emitters 7
3.1 Introduction 7
3.2 The Basics 7
3.2.1 Photon States 7
3.2.2 The Hanbury Brown Twist Experiment 8
3.3 Quantum Dot Single Photon Emitters 9
3.3.1 Introduction to Quantum Dots 9
3.3.2 Collection Efficiency Improvement 11
3.3.3 Multiple Photon Suppression 12
3.3.4 Indistinguishability of the Single-Photon Wavepackets 12
3.3.5 Electrically Excited Quantum Dot Single Photon Emitters 13
3.3.6 Future Prospects on Quantum Dot Single Photon Emitters 14
3.4 Mesoscopic Quantum Well Single Photon Emitters 15
3.4.1 Introduction 15
3.4.2 Experiments 16
3.4.3 Future Prospects on Quantum Well Single Photon Emitters 17
3.5 Molecule Single Photon Emitters 18
3.5.1 Introduction 18
3.5.2 Dilute Molecule Solution between Bragg Reflectors 18
3.5.3 Single Photon Emitter Setup using the Stark Effect 19
3.5.4 Room Temperature Stable Single Photon Emitter 19
3.5.5 Future Prospects on Molecule Single Photon Emitters 20
3.6 Colour Centre Single Photon Emitters 21
3.6.1 Introduction 21
3.6.2 Experimental Results 21
3.6.3 Future Prospects on Colour Centre Single Photon Emitters 22
Conclusion 23
References 24

Chapter 1

Introduction

In the last fifteen years Quantum Information Processing (QIP[1]) has evolved from being a purely theoretical science into an experimental science. This branch of science is built around the superposition principle and the concept of entanglement. Although these principles were known for a long time and actually led to the big debate between Einstein and Bohr about the non-locality of quantum mechanics in the beginning of the 20th century[2][3], it was not until Feynman suggested at a conference in 1982 that quantum mechanical systems could be used to simulate and compute other quantum mechanical systems that serious thought has been given to the ability for quantum systems to perform calculations. With this suggestion the first blueprint for the theoretical quantum computer was given by Deutsch in 1985[4] and subsequent theoretical research has been performed in the decade thereafter[5][6]. In the beginning of the nineties the first physical realizations of new applications in QIP also began to arise: experimental quantum teleportation[7] and quantum cryptography[8] were born. In these days up until today more and more experiments are arising giving shape to the theory behind QIP.

In this report one of the fundamental ingredients needed to make quantum cryptography work, single photon emitters, is discussed. The ideal single photon emitter needed for quantum cryptography should emit no more then one photon at the time. Also, for practical use they should be easy to create, easy to control, be fast and should work at room temperature. In order to understand why the first criterion is crucial for, for example, quantum cryptography purposes a short chapter of this report has been devoted to the very basics of quantum cryptography. In the next chapter an overview is given of single photon emitters produced by various techniques including the utilization of quantum dots, quantum wells, molecules and colour centers. The goal of this report is to give an overview of the various techniques to produce single photon emitters and compare them to see which one is most promising for everyday use in practical applications such as quantum cryptography.

Chapter 2

Introduction to Quantum Cryptography

2.1 Modern Cryptography and the Key Distribution Problem

As soon as people could write, people wanted to communicate secretly and thus all kinds of methods were invented to prevent people who were not supposed to read their messages from reading them. In general the original message, in cryptography called the plaintext, is made unreadable by an encryption procedure to produce a so called cryptotext or sometimes called ciphertext. The intention is then to transport this cryptotext to the person who is supposed to receive the message (whether this is done by means of an electronic highway or on foot is irrelevant in this case) and eavesdroppers can do nothing with this text. The person who is now supposed to read the message then converts the cryptotext into the plaintext again with a decryption procedure. The secrecy of the message then depends on the encryption and decryption procedure. The science of these procedures and their secrecy is called cryptography[1].

In the old days of cryptography, the secrecy of a cryptotext depended on the entire encrypting and decrypting procedures. Today however, ciphers are used for which the algorithms for encrypting and decrypting are known, but need a specific set of parameters, called the key, together with the plaintext or cryptotext in order to encrypt or decrypt the message. In today's computer science this key consists of a string of bits.

In order to obtain secrecy, the key needs to be totally random and should only be known by the sender and receiver of the message. In the ideal case a new key of equal size of the message must be made every time a new message is sent. In reality however people settle for less then ideal secrecy for example by renewing a key every once in a while or repeating a smaller key to create the big key for a message. The problem however persists that a key needs to be shared between senders and receivers at least once in a while. The distribution of a secret key between two persons who initially share nothing is known as the *key distribution problem.*

To solve this key distribution problem, there are basically two solutions to the problem. The first one is the mathematical public key cryptography and the second one the physical quantum cryptography.

The public key cryptography method depends on the fact that certain mathematical operations are easier to do in one direction then in the other direction. Although this is a method very easy to work with it relies on unproven mathematical assumptions about the difficulty of certain algorithms. In fact, it has actually been shown that as soon as the first quantum computer is put into operation most public key cryptography methods immediately become insecure[6]. This creates the need for a new cryptography method which does not have this vulnerability.

The quantum cryptography method promises us completely secure key distribution as long as quantum mechanics as a theory holds. Since this theory has been tested extensively for almost a century now and any additional theory to be discovered will most likely have quantum mechanics as an approximation of that theory this can be regarded as almost completely reliable in contrary to the mathematical case. The principles behind this method of distributing keys are explained next.

2.2 Quantum Key Distribution

In order to establish a key secretly quantum key distribution can be used which gives the promise of being absolutely secure. This works by utilising a quantum channel between a sender and receiver in which either entangled quanta are distributed or single quanta are sent from the sender to the receiver. The measuring of these quanta by any eavesdropper then unavoidably changes the state of these quanta by the von Neuman principle and this change can be detected by the sender and the receiver and provides the certainty that a key is not detected by any eavesdropper and therefore the distributed key is perfectly secure. Besides the quantum channel a public channel is also assumed to be present which is 'ideal' in the sense that anyone is able to see the communication in this channel but no one is able to forge for example identities or messages. Forging problems of these kind are called authentication problems and can in principle be exactly solved by means of an authentication key which needs to be distributed once between two persons by meeting face to face[1].

There exists two possibilities of performing quantum key distribution. The first uses single particle states with a prescribed set of non orthogonal quantum states while the second uses entangled particles with a prescribed set of measurements performed on them. Since the first possibility is the most easiest to explain and gives insight into the necessity of single photon emitters for quantum key distribution this one will be explained. For a treatment of the second the literature[1] may be consulted.

The quantum key distribution setup using single particle states is outlined below:

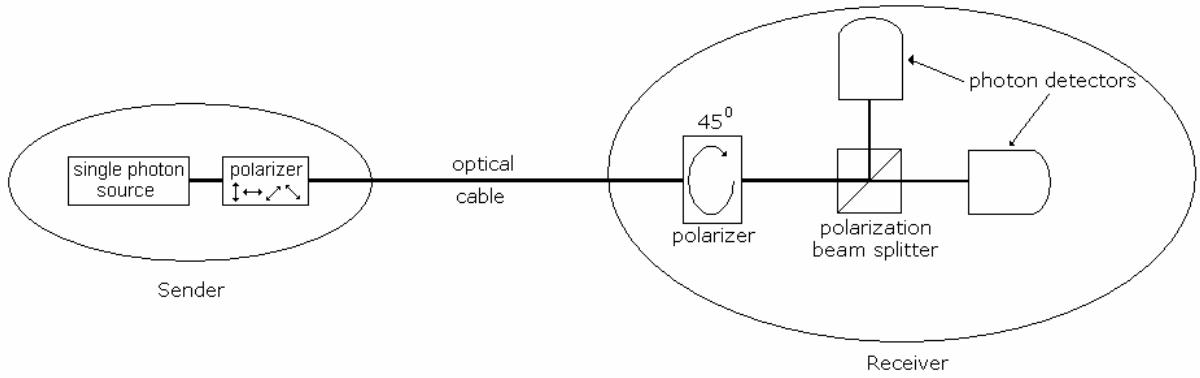

Figure 1: The polarization scheme for quantum key distribution

In this case a sender has a single photon source at its disposal with a fast switching polarizer. The classical bits, '0' and '1', are now encoded into two non orthogonal bases. Let's say that the first basis is the horizontal-vertical polarization basis $\oplus = \left(\left|\updownarrow\right\rangle, \left|\leftrightarrow\right\rangle\right)$ where $\left|\updownarrow\right\rangle$ and $\left|\leftrightarrow\right\rangle$ denote the classical '0' and '1' bit respectively. The second basis is rotated by 45 degrees $\otimes = \left(\left|\nwarrow\!\!\searrow\right\rangle, \left|\swarrow\!\!\nearrow\right\rangle\right)$ with $\left|\nwarrow\!\!\searrow\right\rangle$ and $\left|\swarrow\!\!\nearrow\right\rangle$ denoting again the classical '0' and '1' bits. The setup to the right shows how the receiver is planning to distinguish between all the polarizations send. It consists of a fast switching rotation device capable of rotating the polarization of the incoming photon by 45 degrees or not and a polarization beam splitter, transmitting the vertical polarization and deflecting the horizontal polarization. Two detectors are placed behind this beam splitter in order to distinguish between the elements of the $\oplus$ polarization basis if the rotation device is set to 0 degrees or between the elements of the $\otimes$ basis if the rotation device is set to 45 degrees. If the sender now sends a bit in for example the $\oplus$ basis and the receiver measures this bit in the $\otimes$ basis there exists a probability of ½ that the detectors will measure a '0' and another ½ probability of measuring '1'.

The protocol for generating random keys goes now as follows:

- The sender first sends a randomly chosen binary number with each bit encoded in a randomly chosen basis. The receiver chooses to detect the polarization for each bit in a randomly chosen basis.
- Once sufficient bits are sent the sender and receiver reveal through a public channel which basis they chose for each separate bit. They discard the bits in which they have chosen the wrong basis with respect to each other. Furthermore, if a bit has not been detected at the receiver it is also discarded.

Now in order to show that eavesdropping will unavoidably lead to errors between the string of bits of the receiver and sender, say that both accidentally chose the right basis for any bit in the protocol above. Any eavesdropper, not knowing their basis, detects the bit in any of the two basis and resends it in the same basis. If the eavesdropper does this trick in the wrong basis with respect to the sender and receiver, he has a 50% probability of recording either bit and when resending it, the receiver also has a 50% probability of recording the wrong bit. If the eavesdropper now applies this trick to every bit the string of bits of the sender and receiver then have an average error rate of 25%. By choosing now a subset of the string of bits received with the right basis and comparing the results publicly any eavesdropper can be detected because he inevitably induces errors. If no eavesdropper has been detected the bits not used for the testing for an eavesdropper now becomes the key. A perfectly secure key is then established.

This polarization scheme in principle works but in practice is pretty difficult to realize due to depolarization and birefringence present in optical fibres. Therefore in practice phase encoding is used which relies on interference effects in Mach Zehnder interferometers. To see how this works the literature[1][9] may be consulted.

In this scheme single photons must be used because if pulses of multiple photons are send over the quantum channel eavesdropping can theoretically be performed. This can be seen from the discussion above. If an eavesdropper detects photon counts in both detectors of his receiver he knows he has chosen the wrong basis and can just choose not to resend any photon. The sender and receiver will just consider this as a lost photon pulse and they lost their ability of detecting eavesdropping.

Chapter 3

Single Photon Emitters

3.1 Introduction

As has been discussed in chapter 2, single photon states play a central role in quantum cryptography. It is therefore not surprising that much effort has been put in trying to create these states in an efficient way. Here, various single photon emitters all created using different techniques and sometimes working on different principles will be discussed.

In order to characterise a single photon emitter as being good, several criteria are needed. In the ideal case, the single photon emitter emits pulses at high speed which can only contain one photon at a time all with identical wavepackets and should be able to work at room temperature. Furthermore, the single photon emitter should preferably also be easy to create and easy to control.

In order to understand most of the experiments performed in the field some basics must first be explained. This will be done in the next section.

3.2 The Basics

3.2.1 Photon States

To begin the discussion about the basics needed to understand several single photon emitters let's first consider the fundamental quantum mechanical modes of light. When the electromagnetic field in quantum optics is quantized fundamental modes of the electromagnetic field arise defined by their photon number *n*. They are the so called number states denoted by $|n\rangle$. These fundamental excitations of the electromagnetic field have the property that their electric field magnitude is precisely determined according to:

$$E_0 = \sqrt{n + 1/2} \tag{1}$$

In units of in units of $2\sqrt{\frac{\hbar\omega}{2\varepsilon_0 V}}$ where *V* is the volume of the cavity in which the excitation takes place and ω the frequency of the elementary excitation[9]. The phase however is undetermined due to the number-phase Heisenberg uncertainty principle. Although this set of fundamental modes is complete and orthogonal, excitations of this field we observe in every day life are superpositions of these fundamental modes. This basically means that in the everyday case the electric field magnitude becomes less known, while the phase gets a more defined value according to the Heisenberg uncertainty

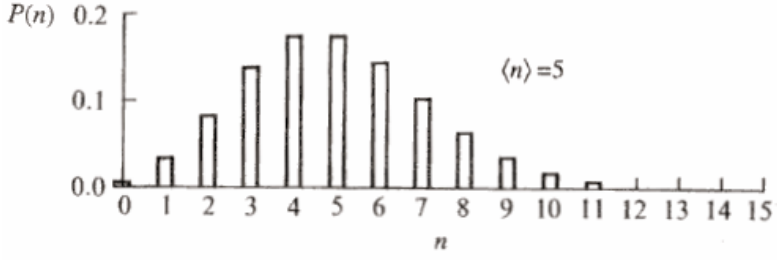

Figure 2: The Poisson photon-number distribution of a coherent state with average photon number 5. P(n) stands for the probability of finding n photons in this state.

principle. For example the light coming from a laser operating above threshold is called coherent light and has a Poissonian distribution in it's number states shown in figure 2. In this Poisson distribution the average of the distribution is $\langle n \rangle$ and the variance $\sigma_n^2 = \langle n \rangle$ as is normal for a Poisson distribution. Everyday light has the property that it is either Poissonian or so called super-Poissonian, that is the variance of the distribution is bigger then in the case of a Poissonian distribution. This light is referred to as classical light. It might also be instructive to say that all states of light are normally confined to a region of space defined by a wavepacket. In this case a Heisenberg uncertainty relation is again at work: the relation between the position and the momentum of the photon state.

In the case of our single photon emitters we want to create the nonclassical single photon state $|1\rangle$ which contains only one photon in a controlled way. No states may be created where more then one photon is present. However, a superposition state between the $|0\rangle$ and $|1\rangle$ photon states can still be used to realise for example quantum cryptography because the physics as described in chapter 2 does not change. The only extra thing which occurs is that there is then a probability of measuring $|0\rangle$ i.e. no photon at all by the receiver if such a superposition state is sent.

If a laser pulse is tuned to be very weak (defined by a very low average photon number) then the probability of detecting one photon in the pulse is low, but the probability of detecting two photons per pulse is even lower. The laser pulse can then approximately be described as a superposition between the $|0\rangle$ and $|1\rangle$ photon states. However, due to the Poissonian distribution a very high probability of detecting no photon is present and this translates into a very low emission rate.

As an example Bennet and Brassard[8], the people who first showed experimental quantum cryptography, originally produced single photons at a rate of 0.l photons per pulse of their laser with a 5% probability of creating more then one photon in each pulse. The goal of creating single photon emitting devices is thus to increase the rate at which single photons can be emitted while at the same time suppressing the rate at which multiple photon states are produced.

3.2.2 The Hanbury Brown Twist Experiment

An experiment to determine at which rate multiple photons are emitted with respect to the rate at which single photons are emitted is a Hanbury Brown Twist (HBT) experiment. In this experiment the autocorrelation function is determined. It is defined by:

$$g^{(2)}(\tau) = \frac{\langle I_1(t) I_2(t+\tau) \rangle}{I^2} \tag{2}$$

where $I_i(t)$ is the intensity at time *t* at place *i* and the brackets denote the average over time. *I* is here the average intensity detected and acts as normalization. The setup of this experiment is shown in figure 3. It consists of a 50/50 beam splitter with two photon detectors placed at equal distances behind the beam splitter. In the case of a HBT setup the first intensity in the auto correlation function is the intensity in the upper photon detector while the second intensity is the intensity detected in the other detector. It can be seen that in the ideal case when single photons are

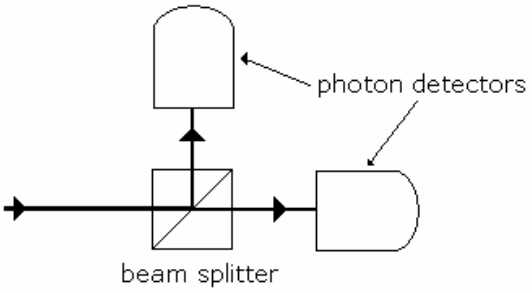

Figure 3: The Hanbury Brown Twist setup consisting of two photon detectors and a beam splitter

pulsed towards the beam splitter that a photon detected at time *t* can either be detected in detector 1 or 2 but not in both. This means that in this case the correlation function at $\tau = 0$ is equal to zero. However, because ideally the single photons are pulsed towards the setup correlation will exist between the intensities measured at detector 1 and 2 at multiple values of the times between pulses. In the ideal case when this autocorrelation is measured therefore a series of spikes will be seen at multiple values of the times between pulses. In practice, due to the finite extend of the photons in the form of wavepackets these spikes are a bit more smeared and often a small peak will still be observed at $\tau = 0$ because of the in general non ideal two photon suppression. If the autocorrelation function in experiments is properly normalized $g^{(2)} \equiv g^{(2)}(0)$ can then be regarded as the probability of finding two or more photons in the same pulse. This is one of the parameters used to estimate the quality of the single photon source.

3.3 Quantum Dot Single Photon Emitters

3.3.1 Introduction to Quantum Dots

The first single photon emitters to be discussed are based on the utilisation of quantum dots. Quantum dots are defined as being structures so confined in 3 dimensional space that particles in this quantum dot start to experience quantum mechanical effects due to their confinement. This is the so called quantum confinement effect. An easy estimate to see when these quantum confinement effects become important is given by considering the Heisenberg uncertainty principle when we confine particles in a region with dimension Δx:

$$\Delta p_x \sim \frac{\hbar}{\Delta x} \tag{3}$$

where Δp_x is momentum uncertainty introduced in the *x* direction. The confinement energy introduced is then:

$$E_{confined} = \frac{(\Delta p_x)^2}{2m} \sim \frac{\hbar^2}{2m(\Delta x)^2} \tag{4}$$

It will become important when it is bigger or equal then the thermal motion energy in the *x* direction:

$$E_{confined} \sim \frac{\hbar^2}{2m(\Delta x)^2} > \frac{1}{2} k_B T \tag{5}$$

For a typical semiconductor with *m* = 0.1 m_0 at room temperature this happens when the confinement has dimensions below 5 nm. The levels introduced by quantum confinement in the upper and lower part of the quantum dots is similar to the levels given for example in the finite potential cubic box problem for cubic quantum dots or of a finite potential spherical well for spherical quantum dots. The method of how to calculate these levels is treated in most quantum mechanics books[3]. For a readable

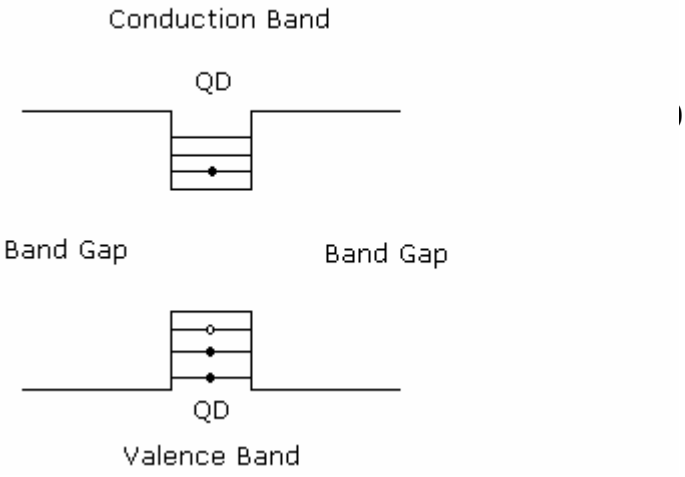

Figure 4: A semiconductor quantum dot band picture in 1 dimension. Note however that the levels inside the quantum dot are discrete and localized. An electron-hole pair can also be observed in the quantum dot.

discussion about quantum confinement, the general term for quantum dots, wires and wells, the literature[10] may be considered.

In the case of the articles discussed in this section, a quantum dot is defined as a piece of semiconductor with a bandgap smaller then the bandgap of the semiconductor in which it is embedded. A one dimensional band picture of this is shown in figure 4. It is noted however that a band picture is a little bit misleading since the whole quantum dot only has a few electronic levels and are not continuously located into bands.

Due to their design, the electronic states in the quantum dots are strongly localised and interact very little with the surroundings giving rise to sharp transition lines within the dot. In addition, because of the Coulomb interaction among the strongly confined electron-hole pairs,

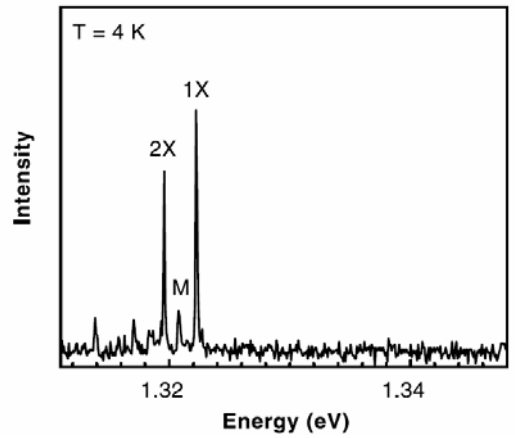

Figure 5: Photoluminescence spectrum of a single InAs quantum dot embedded in a GaAs disc shown in figure 6. 1X is the single exciton level, 2X is the biexciton level and M is a level due to the Purcell effect which enhances the background noise at that energy. Picture taken from reference [11].

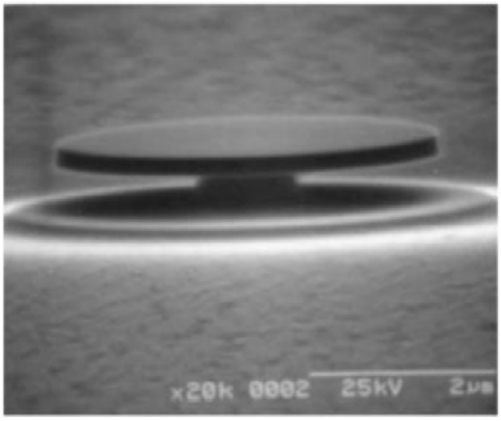

Figure 6: Microdisc structure consisting of a GaAs disc with in it a InAs quantum dot standing on a pole of a AlGaAs mixture. The GaAs-air boundaries provide a weak mirror effect introducing a weak Purcell effect which can be seen in figure 5. Picture again taken from reference [11].

the emission frequency of a single exciton level transition within this quantum dot depends strongly on the total number of carriers in both the valence and conduction band part of the quantum dot separately. This fact makes sure that if two exciton states are present within the quantum dot they can never emit a single photon at the same frequency because if say an exciton in the lowest exciton state decays and a second higher lying exciton goes to this lowest exciton state it still cannot emit another photon at the same frequency because all the energy levels in both parts of the quantum dot are shifted because in both parts a different amount of carriers are present. When the quantum dot is now excited by a laser pulse, in the spontaneous decay of the exciton levels a single transition frequency can then be selected by a monochromator to obtain single photon pulses.

In the single-photon structures the quantum dots lie within a semiconducting host material. As an example a structure is shown in figure 6. When a very short intense laser pulse is pointed at the structure this mainly produces electron-hole pairs in the host material. These electron-hole pairs are then efficiently captured by the quantum dot because this is energetically favourable. In order for the quantum dot to emit single photons the lifetime of the carriers in the host material must be small compared to the lifetime of the carriers in the quantum dot so the quantum dot can only be excited once. However, their lifetime must be large enough for the quantum dot to capture one or more carriers. When the quantum dot decays a relative long time (~ns) later it then emits one or more single photon states each of different frequency which can be filtered by a monochromator.

In order to show the coulomb effect between excitons, a photoluminescence spectrum is shown in figure 5. In this figure the levels 1X and 2X stand for the presence of one and two excitons in the quantum dot before the spontaneous decay of a *single* level. A level M is also shown in the figure illustrating the Purcell effect discussed next.

3.3.2 Collection Efficiency Improvement

In the experiments performed with quantum dots the collection efficiency has often been enhanced by utilising the Purcell effect[12]. If a light source is placed between two mirrors and the wavelength of the light source fits precisely one or multiple times between the mirrors, the spontaneous emission of the light source is enhanced in the directions parallel to the mirror's normal. If then two parallel mirrors are very carefully placed around the quantum dot the emission of photons in the direction of a detector placed perpendicular to the direction of the mirrors is then increased by a factor called the Purcell factor.

Since all of the structures treated[11][13-18] have been created by molecular beam epitaxy[19] this technique has also been used to create the mirrors: in these cases distributed Bragg mirrors. In order to work with these quantum dots all the work has been performed around 0 K temperature in order to suppress all the non radiative carrier losses such as optical phonon emission in the bulk. It has been stated[11] that room temperature operation should in principle be possible by using materials which have higher confinement potentials for electrons and holes reducing the coupling of the quantum dot states with the states of the host material although this might destroy the advantage of easy creation of single photon emitters by semiconductor technology.

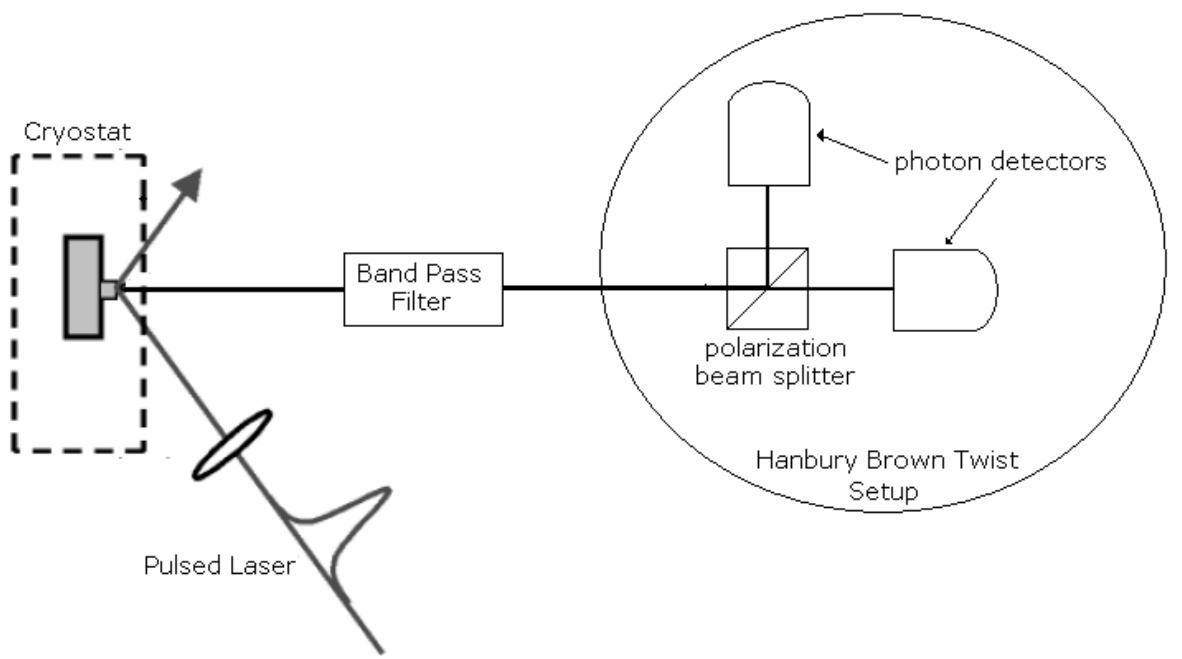

Figure 7: General setup for measuring the auto correlation function of the single photons coming from the quantum dot structures with a Hanbury-Brown-Twist interferometer. Parts of the picture borrowed from [15].

3.3.3 Multiple Photon Suppression

A general HBT setup for measuring the two photon suppression in quantum dots is shown in figure 7. In this setup a pulsed laser is fired from an angle onto the sample containing the quantum dot. The quantum dot is then excited with one or more excitons radiating in succession in either an arbitrary direction or sometimes with a preferred but still somewhat uncertain direction. The band pass filter then only accepts one single transition frequency after which it is sent to a HBT setup to measure the autocorrelation function and determine the two photon suppression.

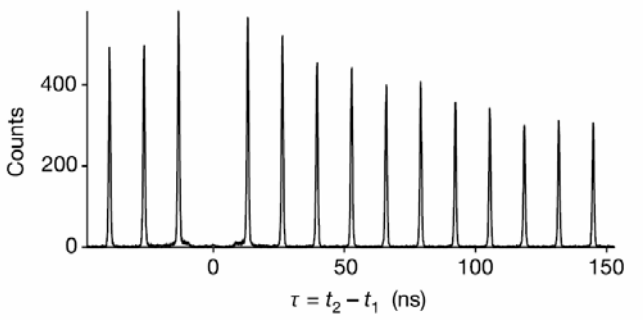

Figure 8: Almost ideal unnormalised autocorrelation function. Picture taken from reference [13].

The results for the autocorrelation function are in general pretty good ranging from $g^2(0) = 0.05$ to 0.3. Recall that this can be regarded as an upper limit to the probability of finding two or more photons in the same pulse. One exception was[16] an experiment in which the biexciton emission line was probably not properly filtered from the single exciton emission line spoiling the experiment and 0.74 was obtained for the autocorrelation function. An almost ideal unnormalised correlation function is shown in figure 8.

3.3.4 Indistinguishability of the Single-Photon Wavepackets

Since most proposed applications for single photon emitters in the field of quantum computing[20] makes use of experiments in which it is necessary that two consecutive photons are indistinguishable this is also tested by one of the authors[13]. Two consecutive photons are indistinguishable if their number state is equal (in this case equal to one) and they are emitted with identical wavepackets. The experiment to test this has an experimental setup often used in theoretical quantum computers.

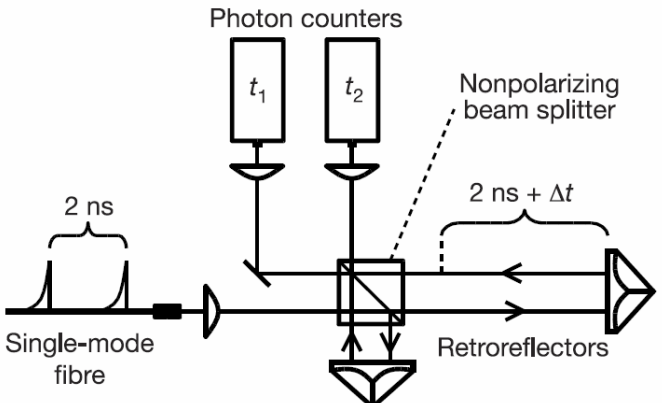

Figure 9: Two Photon Interference Setup. Every 13 ns two pulses are sent containing0 or 1 photons from the single photon emitter. The long arm on the right is 2 ns longer then the short arm on the bottom. Picture taken from [13].

It consists of a Michelson interferometer shown in figure 9. It works on the quantum mechanical prediction that if two indistinguishable photons enter a 50/50 beam splitter from opposite sides both photons need to leave the beam splitter together. This effect has been treated in [9] and [21] and is referred to as the 'bunching' of photons. The experiments using this effect are additionally called bunching experiments in contrast to for example the HBT setup for measuring $g^2(0)$ in which photons are supposed to leave the single photon emitters one by one. The HBT experiment is often referred to as an antibunching experiment.

An autocorrelation result between the two photon counters in the setup is obtained and shown in figure 10. Five peaks appear corresponding to three types of coincidence. For peaks 1 and 5 at $\tau = \mp 4$ ns the first photon follows the short arm of the interferometer while the second follows the long arm. Peaks 2 and 4 at $\tau = \mp 2$ ns correspond to both photons following

the same arm. In these four peaks no two photon interference is observed because obviously although they lead to coincidence effects, they never enter the beam splitter from opposite sides at the same moment. Also note that peaks 2 and 4 are larger then peaks 1 and 5 because in the former case there are two 'paths' in which the photons can travel while in the latter case the photons can only follow one path.

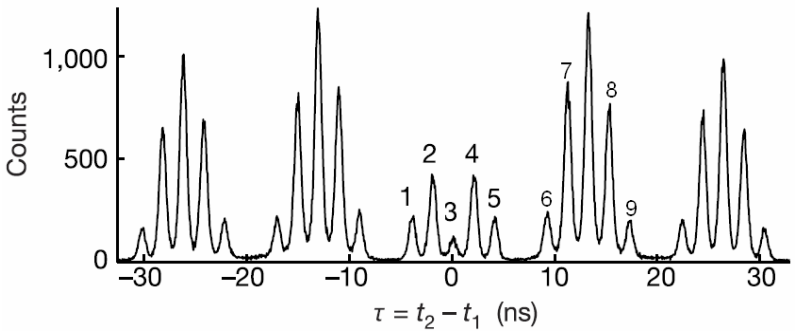

Figure 10: Unnormalised autocorrelation result from the two photon interference experiment outlined in figure 9. Picture taken from [13].

When comparing these peaks with the set of peaks of the neighbouring pulse where photons from different pulses lead to coincidences an additional feature can be observed. Peaks 1 and 5 are almost as high in intensity as peaks 6 and 9 while peaks 2 and 4 are approximately two times smaller then peaks 7 and 8. This is due to the fact that with peaks 2 and 4 compared to peaks 7 and 8 in addition to the 'normal' coincidence effects of peaks 2 and 4 (both photons in the same arm) there also exist an additional probability of coincidence effects: the first or second photon of the first pulse can go through the short arm while the 'same' photon of the second pulse goes through the long arm. With peak 3 at $\tau = 0$ ns however the first photon needs to follow the long arm and the second photon needs to follow the short arm of the Michelson interferometer leading to two photon interference. In the ideal case, this peak should vanish since both photons should leave to one of the photon detectors. In other words, they should bunch.

An additional experiment is performed in which the length of the long arm is varied a little bit to see how this influences peak 3 in figure 10. With this experiment it is possible to investigate the wavepacket overlap between two photons emitted because changing the length of the arm means that the two photons at peak 3 do not arrive exactly on time at the beam splitter but still have some overlap in their wavepackets. The photons arriving at the beam splitter in this case then have a reduced probability of showing two photon interference effects and this can be seen by a higher peak 3. By modelling and fitting this to the experimental data it is possible to estimate the average wavepacket overlap between two photons. This has been done by the author[13] and for his single photon emitters an average wavepacket ranging from 0.7 to 0.8 could be found which is good enough for some proposed quantum computation experiments. The remaining imperfection is due to the fact that when a quantum dot is excited it is first in a higher vibrational state of the electronic quantum dot state and needs to relax to the lowest vibrational state inside the quantum dot by phonon emission. This process takes time and also the finite temperature helps to make sure the electron and hole carriers do not always recombine from the same states. This can change the emitted photon wavepacket a little bit resulting in a non perfect indistinguishability between emitted photons.

3.3.5 Electrically Excited Quantum Dot Single Photon Emitters

As a last application of quantum dots to produce single photon emitters it has also been tried to implement quantum dots into the intrinsic region of a p-i-n diode in order to electrically excite single photons by using short electrical pulses instead of the conventional laser pulses[18]. The schematic of the single-photon-emitting diode is shown in figure 11. In a pulse electrons and holes are injected into the intrinsic region of the p-i-n junction where only one or two electron-hole pairs can be captured by the quantum dot due to its finite dimension. If the pulse is much shorter then the recombination time of electron-hole pairs of the quantum

dot and the recombination time of the intrinsic region is also much lower then this electron-hole recombination time it is then possible to create single photons on demand.

By using a HBT setup focussed on the emission aperture the autocorrelation could be determined shown in figure 12. When comparing this to theoretical fits a two photon suppression $g^{(2)}$ of 0.14 could be determined at low currents. This peak is not shown too clearly in the figure though due to a finite time resolution but when the theoretical curve is calculated and fitted (not shown here) this becomes apparent. All experiments in this case were again performed in a cryostat at very low temperatures (4K) in order to suppress the nonradiative carrier losses.

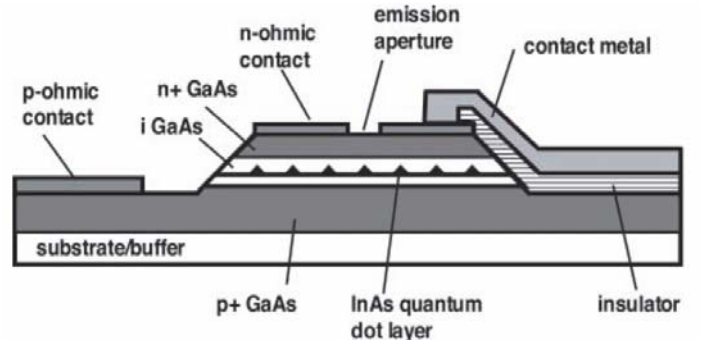

Figure 11: Quantum dots embedded in the intrinsic region of a p-i-n junction. Picture taken from Ref. [18]

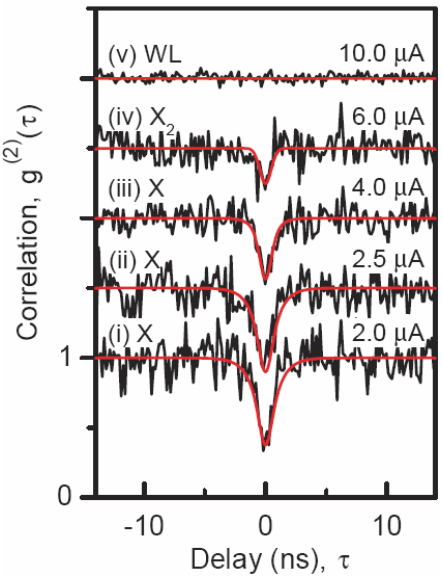

Figure 12: Autocorrelation results from the exciton X_1 and biexciton X_2 line and as a reference the wetting layer WL at different pulse currents. Picture taken from [18].

3.3.6 Future Prospects on Quantum Dot Single Photon Emitters

In order to take a look at the future prospects let's summarize the advantages and disadvantages of quantum dot single photon emitters. The first advantage of these structures is that they are very easy to create by existing proven techniques already implemented on a mass scale such as molecular beam epitaxy[19] and etching techniques. Another advantage is that in principle these structures are able to really produce single photons on demand. This is due to the fact that the efficiency of the source for emitting photons can be brought almost arbitrarily close to 1 by making the laser or electrical pulses strong and short enough and two photon suppression has been shown to get as low as 0.025[13] if the right methods for producing the structures are used. Furthermore it is possible by varying the size of the quantum dots to tune the emission frequency which is useful because in optical fibres a specific frequency is often chosen to minimize attenuation.

It is elegant to utilize electrical pulses to make a laser obsolete. However this is not even strictly necessary since it is already shown that lasers can already be implemented by the same techniques on the substrate of the quantum dot single photon emitters[22][23]. The biggest drawback of using quantum dots is that structures created using easy to use semiconducting materials and techniques need to be cooled to very low temperatures in order to suppress non radiative carrier losses. Room temperature operation should however be possible if other semiconducting materials are used which increase the potential barriers in the quantum dot[11].

It is also noted that the performance of quantum dots as single photon emitters in quantum cryptography applications is in principle good enough since the lifetimes of the quantum dots are all in the order of a few nanoseconds giving a maximum single photon rate of around 100 MHz.

As a conclusion it can be stated that quantum dot single photon emitters are promising candidates for becoming the single photon sources in quantum information applications if the temperature at which they operate can be increased to room temperature by choosing suitable materials.

3.4 Mesoscopic Quantum Well Single Photon Emitters

3.4.1 Introduction

The second possibility of creating single photon emitters is by constructing an intrinsic quantum well, the one dimensional analog of a quantum dot, in the middle of a pn junction.. Next to this central quantum well two additional quantum wells are introduced at the p and n side of the junction separated by tunnel barriers from the central quantum well. This has been proposed in the article of Kim, Benson, Kan and Yamamoto[24].

The band structure of the construction is shown in figure 13A. The lateral side of the structure is confined in order to introduce charging effects and discrete levels in the central quantum well. The construction then effectively behaves as a quantum dot but because the lateral side is around 100 times bigger then the quantum well side it is still named a quantum well structure. The physical process which the authors of this article used is however different from that treated in section 3.3 about quantum dots so it is still handy to make the (somewhat artificial) distinction between quantum dots and this 'quantum well'. If a certain voltage is applied across this construction an electron is able to tunnel through the barrier when an empty energy level within the quantum well lies on resonance with the Fermi energy of the n-type quantum well due to a process called resonance tunneling.

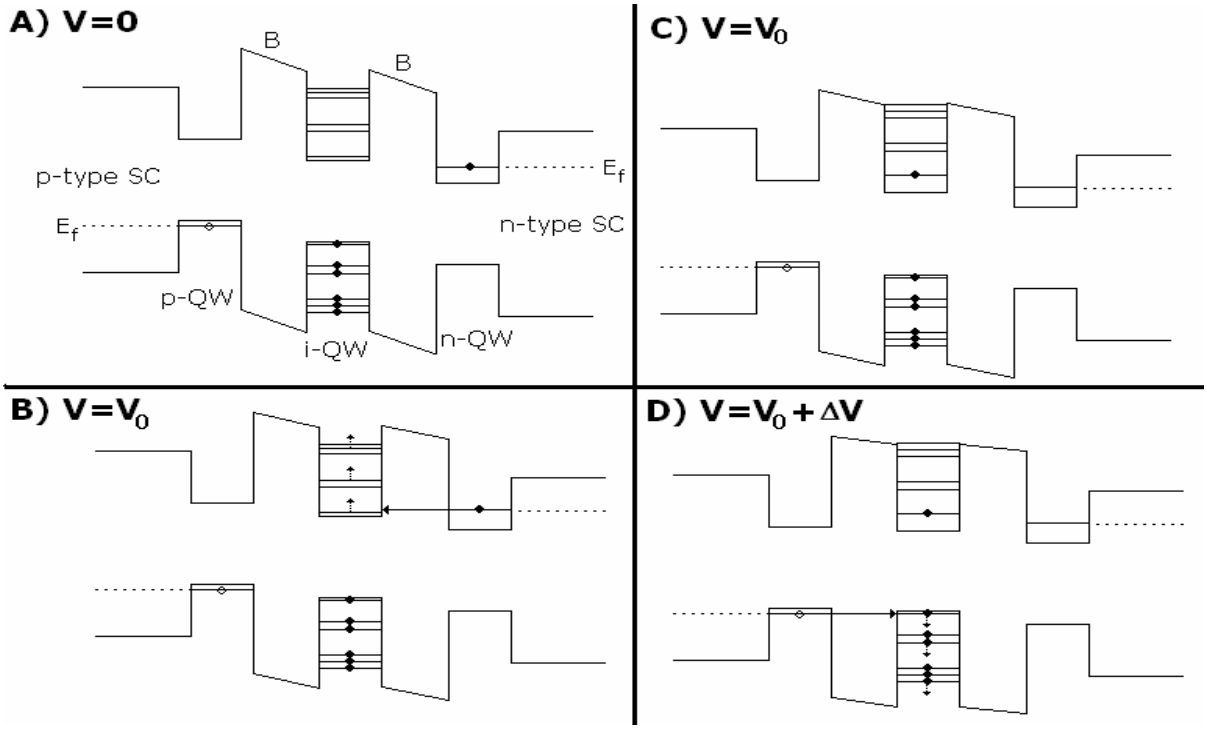

Figure 13: A) The scheme of the mesoscopic quantum well SPE when no voltage is applied, The intrinsic quantum well contains a few electrons and due to the disc like shape of the central quantum well the levels are spaced as indicated in a single 'S level', two closely spaced 'P levels' and three closely spaced 'D levels'. B) When a voltage is applied one electron can inject in the central quantum well. C) The coulomb effect makes the upper quantum well levels go up so no further electron can be injected. D) If an additional voltage is applied a hole can also inject in the central quantum well after which the energy levels are shifted downward again so no further hole can be injected. The electron hole pair recombine and a hole is again injected if the voltage stays as in D). However, because this voltage is too high for electron resonance tunneling to occur no additional electron will be injected in the valence band.

Immediately after the first electron is injected into the quantum well the coulomb interaction between the electrons causes the levels in the upper part of the central quantum well to shift upwards thus preventing additional electrons tunneling in the central quantum well since the new energy level of the quantum well is no longer on resonance with the Fermi level of the n type quantum well. If now an additional small voltage is introduced the Fermi energy of the p-type quantum well can be made equal to the upper energy level of the bottom part of the central quantum well and with this resonance tunneling can occur in which a hole is transported to the central quantum well. The energy levels are then shifted downwards again preventing more then one hole tunneling. After this tunneling no new resonances occur for holes or electrons since the Fermi energies on both sides are not on resonance with one of the energy levels of the central quantum well. The whole process is shown in figure 13.

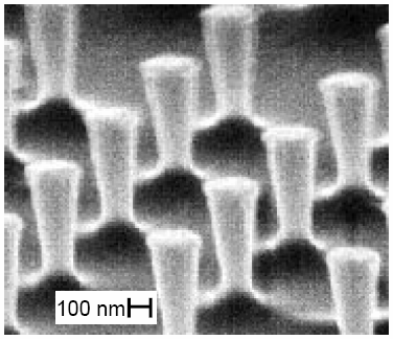

Figure 14: the 'single photon turnstile' devices from [24] consisting of a GaAs/AlGaAs three quantum well structure between n and p-type AlGaAs bulk layers. Figure from [24].

By modulating the voltage now between the two voltages V_0 and $V_0 + \Delta V$ it is possible to inject periodically one electron and one hole and given that the tunnel time and recombination time are smaller then the period of the voltage oscillation only one photon is emitted every oscillation.

From figure 13 it is evident that the structure must be carefully designed by choosing materials with the right bands and right sizes for the quantum wells in order to make this operation possible. This has been done by the group of Kim, Benson, Kan and Yamamoto and a picture of their pillars containing the quantum wells is shown in figure 14. The pillars have been produced by molecular beam epitaxy, electron beam lithography and subsequent plasma etching.

3.4.2 Experiments

In order to make the device work the authors applied simultaneously a DC and AC block voltage. What they found was that the current through the device was a linear function of the

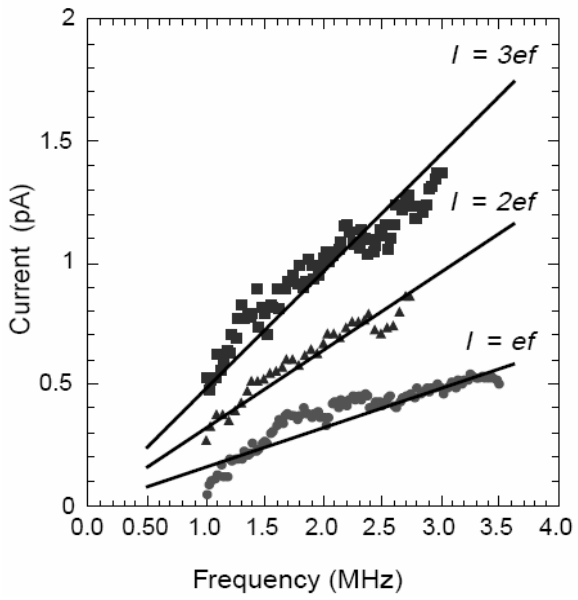

Figure 15: Current through the device versus the applied frequency at different DC voltages where 1, 2 and 3 electrons and holes are injected periodically. The data fits the theoretically expected curve I=3ef well. Figure from [24].

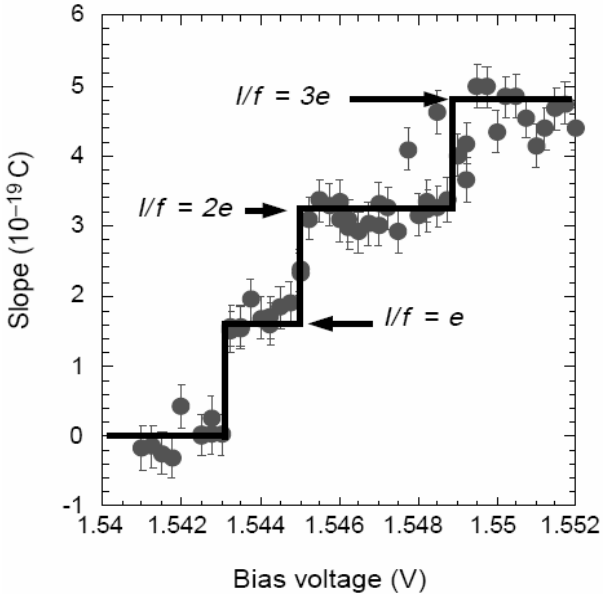

Figure 16: I/f slope versus the bias voltage in the device. Different plateaus are observed corresponding to 1, 2 and 3 levels simultaneously addressed by a constant AC voltage. Figure from [24].

frequency applied which is off course logical since in each cycle one electron and one hole are transported from the n and p-type regions to the central quantum well. The effect is shown in figure 15. Furthermore, the DC voltage V_0 of the whole experiment has also been varied to see an effect called *conductance quantization.* This effect is shown in figure 16 where the slope I / f is plotted against the bias voltage. The AC voltage is held constant in this case. This effect is very clearly explained in [25] and is not going to be repeated in detail here. It is due to the fact that as the DC voltage goes up other more closely spaced higher levels can be addressed (see figure 13 for the approximate level spread for disc like quantum dots) between V_0 and $V_0 + \Delta V$. Although a block pulse is used in the case of Kim, Benson, Kan and Yamamoto the relative inhomogeneous linewidth of the Fermi levels on the n and p-type quantum well causes these levels to be addressed. Evidence lies with the fact that the block pulse sweeps between two values faster then the measured electron and hole tunneling time.

3.4.3 Future Prospects on Quantum Well Single Photon Emitters

In order to look at the future prospects of this device being used as a single photon emitter in everyday applications let's summarize the advantages and disadvantages and see how some elements of this single photon emitter may be improved.

Although no HBT experiment has been used in this case there is strong evidence in the current versus frequency graph that only one electron and hole recombine with a certain frequency at the right DC voltage so although we don't know the exact two photon expression it can be safely assumed that this will be at least reasonable since lifetimes and an internal efficiency of 33% has been determined in this setup by measuring the correlation between the counts of a photon counter and the electrical setup. A positive point in this case is that a laser is again obsolete although this is not an essential obstacle.

A bigger problem of the setup is the temperature which had to be kept at mK temperatures in order to use the discreteness of the Fermi levels on both doped quantum wells in the resonance tunneling process. If this temperature is increased electrons and holes are more distributed across the Fermi levels and multiple energy levels are addressed. In this case also the quantum confinement energy of an electron $E = e^2/2C$ becomes invisible due to the distribution. In order to make this device work at room temperature the quantum well has to be very confined to make the charging energy large and their levels very well separated. To see if this is possible let's consider the charging energy of a quantum dot. To obtain a charging energy of 100 meV (large compared to the ~20 meV room temperature) a spherical quantum dot approximated by an isolated charged sphere in a typical dielectric needs a radius of a few nanometer. By this time the energy levels are well separated in the infinite well approximation (see Fox[10] formula 6.13 ΔE ~ 1 eV between the first and second level) so these barriers for room temperature operation can in principle be taken but this is not easy with current semiconducting technology. It also severely limits the ability to tune the frequency of the single photon emitter. Another potential problem is the competition with non radiative carrier losses. However, due to their design with tunnel barriers induced between the three quantum wells it is expected that this may be a lesser problem compared to the quantum dot structures in the previous section.

Performance wise these structures do not really differ from the quantum dot single photon emitters. By coupling a detector to a voltage circuit a combined tunnelling and recombination time of 5 ns was found so again a theoretical maximum frequency of around 100 MHz should be possible which is again enough for most applications.

As a conclusion it is stated that quantum well single photon emitters can also in principle be realised as single photon emitters although this can be pretty difficult due to the very small size needed for the quantum wells. Furthermore the ability to tune the frequency is also severely limited with respect to the quantum dot structures discussed before.

3.5 Molecule Single Photon Emitters

3.5.1 Introduction

The third possibility to be discussed to produce single photon emitters is by using single molecules as excitation centres. Molecules are attractive due to their intrinsic property that they can only emit one photon within a certain period of time. They need a full excitation-emission cycle before they can emit a second photon. To excite molecules in general pulsed lasers are used but continuous mode lasers can also be used due to this excitation-emission cycle. If a laser pulse is so short in time that the excitation-emission cycle of the molecule takes longer then the time for a pulse to pass a molecule and excite it then only one excitation can be induced and only one photon will be emitted after the pulse.

In order to couple the single photon states emitting from the molecule to for example a HBT setup the single photon states must be separated from the laser light. This is achieved by utilising fluorescence of a four level molecule, in which the molecule is first excited to an vibrational state of the first electronic state. This vibrational state then non radiatively decays to the lowest lying vibrational state of the first electronic excited state after which it emits a photon by radiative decay to the highest lying vibrational state of the electronic ground state. This process is shown in figure 17. Because the emitted photon is emitted at a different frequency a simple band pass filter can be used to filter the laser light from the single photon states.

Figure 17: a molecular four level system. Figure taken from [28].

To make the scheme with molecules as single photon emitters work molecules have been searched which needed to be highly fluorescent and very thermally stable because if the molecules are to be excited a very intense short laser pulse needs to be used which can severely heat up the molecule and it's surroundings.

3.5.2 Dilute Molecule Solution between Bragg Reflectors

In the first article to be discussed[26] a dilute Oxazine 720 molecule solution between two Bragg reflectors is used. These Bragg reflectors are highly reflecting at the emission frequency but transparent at the frequency of the laser. They make sure that the photon states coming from the molecules have sharper wavepackets and can be better coupled to a HBT setup they used to determine the autocorrelation of their molecules. This effect is called the Purcell effect and has been discussed previously in section 3.3.2 and can also be found in [12]. In the article a very extensive quantum optical treatment is given for the photon states emerging from a four level molecule excited by pulses of light. In this treatment different states of light for the light pulses passing the molecule have been considered but more importantly it is calculated which photon states emerge from the molecule when it is excited with different duration of the light pulses. It turned out that by carefully manipulating the time duration of the light pulse it should in principle be possible to tune the photon number states $|n\rangle$. This can have applications in for example quantum computing. Furthermore, the effect of the density of the molecules in solution on the autocorrelation function is investigated. It turned out that the autocorrelation was really zero up till a certain molecule density after which the single photon states exponentially become a Poissonian light distribution. However, in order to keep the molecules stable the experiments had to be performed in a cryostat at 10K. It is needless to say that these molecules are not very promising to use for everyday single photon emitters since the temperature can probably not really be improved and also solutions are not very easy applicable on a larger scale.

3.5.3 Single Photon Emitter Setup using the Stark Effect

Another way of creating single photons with the help of molecules can perhaps be most easily explained by showing the schematic experimental setup for this experiment in figure 18. In the experiment performed by Brunel *et al* [27] dibenzanthanthrene (DBATT) molecules are placed in a n-hexadecane matrix. This is placed between two metal plates on a sample which on its turn is connected to a radiofrequency voltage source. This voltage source modulates the transition frequency of the molecules due to the Stark effect. If now a continuous wave laser is directed at the molecules with a frequency equal to the transition frequency of the molecules and the RF source is applied, the molecules can be excited two times every period of the RF source if the frequency of the voltage source is high enough to excite the molecules once when passing the resonance with the laser but low enough to allow the molecules to decay to their ground state between successive excitations. Under these conditions, two times every modulation of the RF voltage source a single photon is emitted which is focussed by the parabolic collecting mirror and sent to a HBT setup to measure the autocorrelation. In this setup the molecules between the two RF plates need to be sufficiently spread across a surface and should not cluster in order to suppress multiple photon emission. After this one molecule should be targeted by the laser.

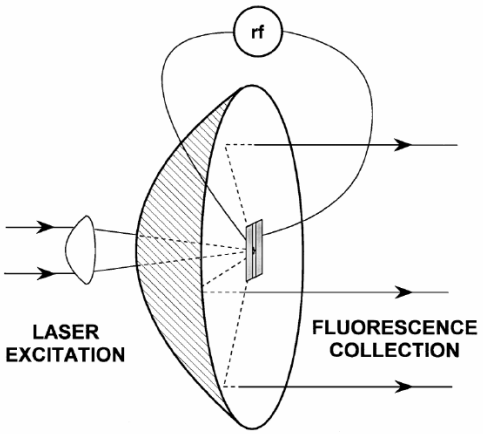

Figure 18: Schematic setup of a single photon emitter based on the Stark effect for the transition of molecules. Figure taken from [27].

In the setup a probability of at most 12% is deduced from the HBT experiment that more then one photon is sent if the amplitude of the RF field is made large enough which is quite reasonable and can already be very useful in quantum cryptography experiments. The internal efficiency of the setup was also almost ideal due to the use of highly fluorescent molecules and a parabolic mirror to collect the single photon states. However, again in this case a cryostat was needed at 1.8K to keep the molecules stable so these molecules are obviously not appropriate for everyday single photon emitters. Other molecules which are more temperature stable might work but an obvious disadvantage of the technique remains that the molecules are heated all the time by a laser so very stable molecules need to be used if this technique is ever going to work. The whole setup however might in principle work if these molecules are found and if these molecules are also easy to process it should not be that difficult to implement this technique on a mass scale since mirrors and continuous wave lasers[23] can already be implemented.

3.5.4 Room Temperature Stable Single Photon Emitter

As a last example a molecular single photon emitter is reported which works at room temperature[28]. In this case a sublimed crystal flake a few micrometers in thickness of p-terphenyl is doped with small amounts (10^{-11} mole per mole) of terrylene. A fluorescence microscope image and the molecular structures of p-terphenyl and terrylene are shown in figure 19. A single terrylene molecule is now selected with the help of fluorescence microscopy. The selected molecule is then excited with a laser producing short and very intense pulses. With the very intense and very short laser pulses used in this article the molecule is excited with a probability of almost one. The pulses can be very intense because the fluorescent terrylene molecules are very stable due to their ability to emit phonons in the p-terphenyl.

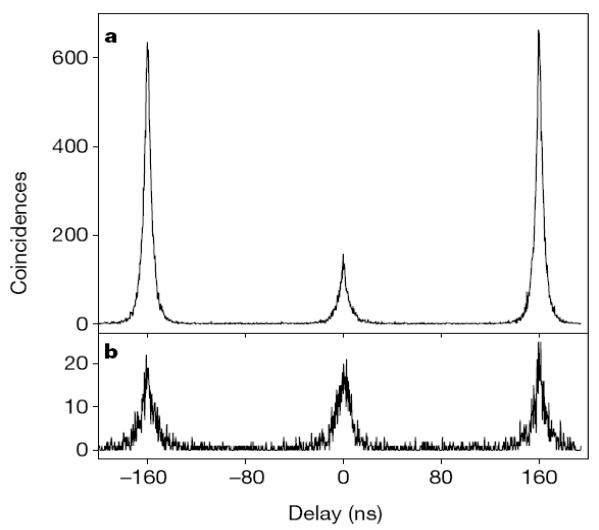

Figure 20: Autocorrelation of a) the pulsed laser focussed on a single terrylene molecule for 120 seconds and b) the pulsed laser not focussed on any terrylene molecule for 300 seconds. Note that when comparing the coincidences the central peak in a) cannot be solely due to the 'background' coincidence. Figure taken from [28].

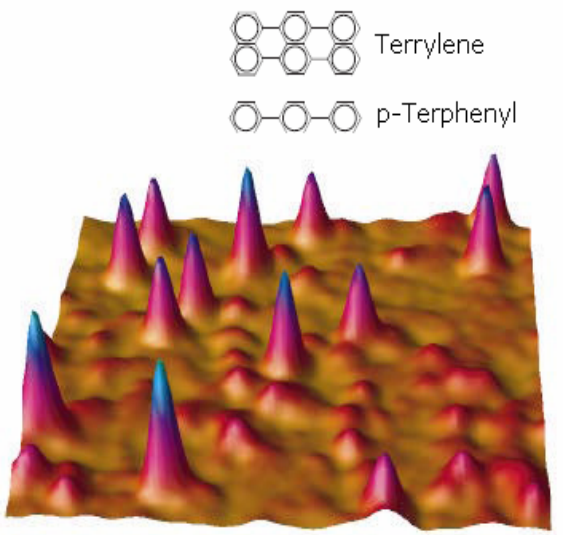

Figure 19: Fluorescent microscope image from the crystal flake. The points indicate the presence of the highly fluorescent terrylene molecules on the surface. The molecular structures of terrylene and p-terphenyl are shown above. Figure taken from [28]

Next, it is argued that the pulse is so short that the theoretical probability of exciting two photons in the same pulse should be around 10^{-3} and an autocorrelation experiment is performed with the laser focussed on a terrylene molecule and on a spot where no terrylene molecule is present. The unnormalised autocorrelation of the terrylene molecule and the background is shown in figure 20a and b. It is then reasoned that the peak at the centre of the autocorrelation of the terrylene molecule is due to the excitation of background molecules.

However, considering that the background autocorrelation has been performed for 300 seconds and the molecule autocorrelation for 120 seconds and by looking at the coincidence rates for both, it can be seen that this cannot be the only reason for the central peak in the molecule autocorrelation. Since only a division has been given of the area of one of the side peaks divided by the area of the central peak of 0.27 this can be taken as the probability of two or more photons being emitted by the setup until off course a better reason is mentioned.

This is a somewhat weak two photon suppression but it has to be noted that this is an upper boundary to two photon suppression and in reality this factor can lay lower. It remains however the most promising and proven example of single photon emitters thus far. The only drawback to these single photon emitters is that they might be difficult to implement on a mass scale since a single terrylene molecule needs to be targeted. It is however not very difficult to imagine that the molecules might be deposited at random and the good single photon emitters can be selected by a simple HBT experiment. It is therefore in principle possible to use these molecules on a mass scale although it might not be the most elegant implementation.

3.5.5 Future Prospects on Molecule Single Photon Emitters

To conclude this section about molecule single photon emitters it is mentioned that in all of the cases reported here it is difficult to harness single molecules on a small scale. Either it works with a sublimed crystal flake or a solution which are difficult to spincoat or deposit using the same semiconductor technology as for the laser which is needed in all cases. Despite this drawback it has been shown that these systems can work at room temperature which has

not been proven yet in the case of quantum dot single photon emitters. An obvious drawback in all these cases is that the light from the molecules is harder to tune although it should in principle be possible to tune the emission of the molecules by creating for example side groups to the molecules[29].

Performance wise molecules are theoretically as good as quantum dots and wells since the lifetime of the molecules, the real fundamental limit to at what speed the single photon emitters can operate, is in the order of nanoseconds.

As a final note it is mentioned that the search for highly fluorescent and temperature stable molecules does not stop and it may be that some day a superior molecule may be found which works at room temperature, is thermally very stable and easy to process making it an excellent single photon emitter.

3.6 Colour Centre Single Photon Emitters

3.6.1 Introduction

As a final example single photon emitters using colour centres are discussed. The principles are the same as in the previous section, only in this case a colour centre instead of a molecule is used as the four level system.

A promising example of the utilisation of colour examples is shown in [30]. Before this article was published it was shown that nitrogen vacancy colour centres soon to be discussed were very stable and highly fluorescent[31].

In order to realise the room temperature stable four level system discussed in [30] diamond nanocrystals have been grown from a synthetic diamond powder. They have typical sizes in the order of 100 nm. In these nanocrystals substitutional nitrogen impurities are naturally present. If they are bombarded by electrons vacancies are introduced in the nanocrystals. By annealing the crystals after this bombardment vacancies tend to move next to the nitrogen impurities in order to create *nitrogen-vacancy* (NV) *colour centres.* These colour centres basically behave as a four level molecule and therefore the same setup as in the previous section can be used to create single photon emitters.

The nanocrystals are first dispersed on a surface and then a single nanocrystal with an average of a few NV colour centres is selected. A continuous wave or pulsed laser is then used to excite the NV colour centres and with a band pass filter the laser frequency can be filtered to obtain the single photons. The NV colour centres are in general still well localised inside the nanocrystal so they theoretically should not emit multiple photon states.

Furthermore they are pretty stable so the whole experiment can be performed at room temperature which can make these sources convenient for everyday use. It is noted again that a continuous wave can also be used to excite single NV colour centres since they need to go through a full excitation-emission cycle before they can emit a second photon. However, this is not convenient if the excitation of single photons needs to be controlled. It can however be used to obtain an autocorrelation function.

In order to use this as a real single photon emitter diamond nanocrystals must be grown which have exactly one NV centre and this diamond crystal needs to be targeted.

3.6.2 Experimental Results

In previous attempts to utilise NV colour centres as single photon emitters the autocorrelation could not be obtained because in these cases bulk diamond has been used[31]. In bulk diamond the single photons emitted from the NV colour centre were strongly attenuated resulting in a detection frequency which was too low to obtain the autocorrelation function. Autocorrelation of the emitted photons in the diamond nanocrystals have however been obtained using both pulsed and continuous wave lasers and these have been shown to lie

around $g^{(2)}(0) = 0.15$, good enough for most applications. The lifetime has been measured to be 25 ns which gives theoretically a performance which lies around 10 times lower then all the other single photon emitters discussed but this is still good enough for most applications.

3.6.3 Future Prospects on Colour Centre Single Photon Emitters

The diamond nanocrystals are easy to create and can be spincoated on a selected surface. They are therefore easy to implement on a large scale which is an advantage they share with quantum dots. The main disadvantage present in molecule single photon emitters that single molecules have to be targeted remains however in this case but might be a lesser problem since targeting has to be done on single crystal scale instead of single molecule scale. Together with the fact that multiple photon suppression is reasonable and operation is possible at room temperature this technique is the most promising example of all the single photon emitters discussed to be created on a larger scale.

Chapter 4

Conclusion

In this report an overview has been given about single photon emitters based on different principles utilizing quantum dots, quantum wells, molecules and colour centres and there has been speculated about how suitable these structures are for production on a bigger scale for quantum information applications such as quantum cryptography.

To summarize the results we can say that all the methods of producing single photon emitters discussed in this report should in principle be capable of producing almost ideal single photon emitters. They are ideal in the sense that only a single photon is produced on demand and that they can work at room temperature. Differences remain however in the difficultly with which the single photon emitters can be produced on a bigger scale. It turned out that although normal quantum dots and wells are easy to implement with current semiconductor technology and have excellent two photon suppression checked by HBT experiments, other semiconductor materials and smaller quantum dots are needed to make the quantum dot and well structures decay radiatively at room temperature. These present challenges to semiconductor technology which makes their implementation difficult. On the other hand molecule and colour centre single photon emitters have already been shown to be able to work at room temperature. Together with the fact that they have reasonable two photon suppression even at room temperature these represent the most probable realization of single photon emitters in the near future until semiconductor technology is able to produce smaller and other material quantum dot and well structures. Molecule and colour centre single photon emitters have the potential drawback however that they might not be easily controllable with current semiconductor technology.

In any case if it is possible to produce single photon emitters on a mass scale which are room temperature stable quantum cryptography and also other quantum information applications such as the linear quantum computer will become feasible.

Chapter 5

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