Cavity QED effects in low dimensional systems

Arnardottir, Kristin Bjorg

2018

Arnardottir, K. B. (2018). Cavity QED effects in low dimensional systems. Doctoral thesis, Nanyang Technological University, Singapore.

<http://hdl.handle.net/10356/75866>

<https://doi.org/10.32657/10356/75866>

**NANYANG
TECHNOLOGICAL
UNIVERSITY**

SINGAPORE

CAVITY QED EFFECTS IN LOW DIMENSIONAL SYSTEMS

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A thesis submitted to the Nanyang Technological University in partial fulfilment
of the requirements for the degree of Doctor of Philosophy.

2018

Acknowledgements

First, I would like to thank my supervisor Prof. Timothy Liew who has been very helpful and supportive. I also want to thank my former supervisor Prof. Ivan Shelykh for all his help and for giving me so many opportunities. I would not be in Singapore without him.

I would also like to thank all my colleagues and fellow students during my time in both Shelykh group and the group of Prof. Liew. In particular, I would like to thank Dr. Tania Espinosa-Ortega, Dr. Helgi Sigurðsson, Dr. Skender Morina, Dr. Kristinn Kristinsson, Dr. Ivan Savenko and Dr. Oleksandr Kyriienko for many insightful talks, both relating to physics and not. We shared a lot of time and memorable moments which I will not forget.

I am grateful for all the friends I have made in Singapore, who have enriched my life and made this time wonderful, even though it was hard at times.

Lastly, I want like to thank my my whole family, especially my parents. Without their unwavering support and belief in me, I would not be where I am today. They push me to be the best I can be and I hope I have made them proud.

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Abstract

Cavity quantum electrodynamics (QED) is the study of the interaction between light confined in a microcavity and material modes (such as single atoms or electrons in quantum wells), where the quantum nature of light is crucial. This thesis theoretically examines four different systems where light becomes strongly coupled with different material modes leading to different effects stemming from the different dimensionality and natures of the systems. The first system consists of a semiconductor slab embedded into a zero-dimensional microcavity, leading to vacuum band gap opening of the electron dispersion caused by the interactions with the cavity photon mode. The second system is a quantum ring coupled to polarized light, calculated non-perturbatively. Since the polarization breaks time reversal symmetry, the particles in the ring will exhibit an energy difference of particles with the same magnitude of angular momentum but different directions. The viability of these resulting in Aharonov-Bohm oscillations in large rings is considered. The third system consists of an array of quantum wires embedded into a planar microcavity. The inhomogeneity of the two directions along and perpendicular to the wires, coupled with the homogeneous two-dimensional photon mode, leads to an emergence of a hyperbolic region and many peculiar effects. The fourth and last work considers an algorithm to modify systems, leading to a ladder of equidistant energies. To illustrate, a radially symmetric nanoparticle with a hydrogenic impurity is analysed. The material composition along the radius can be altered to achieve a cascade of transitions in the Terahertz range. When multiple such particles are embedded into a resonant microcavity, one can convert photons of a higher energy to multiple Terahertz photons.

1. Introduction

The interaction between light and matter is a fundamental aspect of physics that leads to many interesting phenomena. These interactions are all around us and its many forms fall under different aspects of physics. The light from our sun is a consequence of concepts described by nuclear and thermal physics, the formation of rainbows is expressed in classical optics, and solid state physics gave us the light emitting diodes we see making up screens and billboards.

From a quantum viewpoint, the basis of the coupling between light and matter are the two processes of the absorption of a photon by the matter system leaving the latter in an excited state, and the time-reversed process when the matter system relaxes from an excited state to a state of lower energy emitting a photon. The coupling between a matter mode and light is generally relatively small, meaning that the time between photon absorption and emission is really short compared to other relevant time scales. It is however possible to engineer systems where the rate of coupling becomes larger.

The most straightforward way of engineering a system where this can happen is to make sure that the light in question is confined. This can greatly enhance the strength of the light present in a given space, which greatly increases the likelihood of both absorption and stimulated emission of a photon, called the *Purcell effect*[1]. This is often done by embedding the matter system inside a *microcavity*. Inside the microcavity, a photon has the opportunity to interact multiple times with the same matter mode. When the rate of the interactions becomes fast enough the two modes representing the light and matter, respectively, become mixed in a way where it is not possible to separate the two, resulting in two new modes that are a linear combi-

nation of both of the preceding modes. This phenomena is called *strong coupling* and the resulting modes are described as belonging to a quasi-particle called *polariton* which can be considered as a composite of the cavity photon and the matter mode. Weisbuch *et al.* were the first to observe polaritons experimentally in 1992, where the coupling took place between light and excitons (diples formed by electron-hole pairs) in a quantum well [2]. In the over two decades since, the regime of strong coupling has been extensively investigated both theoretically and experimentally in a variety of systems, including optical planar microcavities with semiconductor [3–5] and organic [6–8] quantum wells, intersubband transitions [9–12], zero- dimensional cavities with individual quantum dots [13–15] and others. Effects of strong coupling has been proposed for a variety of technological applications [16], including novel types of lasers [17, 18], electrical spin switches [19], optical switches and logic gates [20–22], all- optical integrated circuits [23], sources of entangled photon pairs [24, 25], light-mediated superconductivity [26], weak lasing [27], frequency combs [28], photon guns [29] and many more.

The focus of this thesis revolves around looking at different systems where light matter coupling takes place, where either or both of the two modes being coupled are of lower than three dimensions. In this first chapter I will start by introducing the concepts vital to understanding the works that follow. I will start by giving a brief introduction to the different types of microcavities, then give a background on light-matter interactions, mainly focusing on the *strong light-matter coupling* regime. Afterwards, I will give some perspective on how the dimensionality of a system can influence the behaviours of the particles enclosed. Last, I will introduce a theoretical framework based on Feynman diagrams and Green’s functions, describing how it can be used and what approximations can be made using it.

The second chapter demonstrates the use of the Green’s functions approach for a system consisting of a slab of semiconductor in a closed cavity. The dimensions of the systems are chosen so that the electrons in the semiconductor can be treated as continuous (i.e. 3 dimensional) while the cavity light modes are discrete (i.e. 0 dimensional). When the electron dispersion relation is renormalized, taking into

account the interaction between the electrons and the cavity photon ground modes, a band splitting occurs. The number of gaps opened will depend on the relative direction between the electron momentum and the wave vector of the ground photonic mode.

The third chapter contains another example of the use of the Green's functions approach, where the coupling between circularly polarized light and particles in a one dimensional ring is studied. Since the circular polarization of the light breaks time reversal symmetry, two states of equal but opposite angular momentum will no longer have the same energy. This energy shift is studied for different parameter ranges. The results are compared with results previously published using a perturbative approach, while the current method based on Green's functions is non-perturbative.

The fourth chapter begins by introducing concepts used later in the chapter, such as Bose-Einstein condensation and hyperbolic metamaterials. The latter part of the chapter is then devoted to the study of excitons in a periodic array of quantum wires embedded into a planar microcavity. The effect of the coupling between the one dimensional excitons and the two dimensional photons is calculated using the formalism of second quantization. The periodicity of the wires leads to an emergence of a quasi-momentum of the excitons perpendicular to the direction of the wires, similar to what is seen for electrons in crystalline structures. When the 2D photonic mode couples with the quasi-2D exciton mode, the resulting dispersion relation exhibits some peculiar effects owing to the inhomogeneity of the two directions (perpendicular and parallel to the wires).

The fifth chapter describes a new approach for engineering systems for producing terahertz (THz) radiation. The concept revolves around looking at systems where energy levels can be modified by tuning an external parameter, such as material composition. Using variational calculus we can make an algorithm that searches for solutions which lead to a ladder of equidistant transitions in the THz range. To demonstrate this we consider a $\text{GaAl}_x\text{As}_{1-x}$ nanoparticle, where we engineer the energy levels by varying the aluminium concentration x along the radius of the particle.

1.1 Microcavities

A microcavity is an optical resonator that confines light in at least one direction. The simplest way to achieve this confinement is using a Fabry-Pérot cavity. Microcavities consist of two mirrors separated by a distance on the scale of the wavelength of the light to be confined. These mirrors can make use of single interface reflection (such as metallic interfaces) or by using periodically modulated microstructures optimized to reflect a fixed, resonant wavelength. A widely used example of the latter is the *distributed Bragg reflector* (DBR). The DBR is made by alternating layers of two different refractive indices, n_1 and n_2 , of differing thickness, d_1 and d_2 . When a light beam is shone onto the stack it will be partially reflected at each interface, as depicted in figure 1.1. The parameters of the system can be chosen such that the outgoing beams will have the same phase, resulting in a constructive interference and a near perfect reflection. This condition is $n_1 d_1 = n_2 d_2 = \lambda/4$, where λ is the wavelength of the light. The reflectivity of the DBR will always have high reflectivity for the resonant frequency but by increasing the number of alternating layers light of proximate wavelengths will also experience high reflectivity. The reflectivity spectrum of a DBR is shown in figure 1.1(b). There we can see a high reflectivity band (often called stop band) around the resonant frequency.

Even though the reflectivity of DBRs can be high, it can not reach unity with a finite number of layers. This means that realistically there will always be some loss of photons through the mirrors, as there will also be for single interfaces. In other words, when one composes a cavity, the cavity photons will have a finite lifetime τ_{ph} , which leads to a broadening of the spectral lines of the confined modes. The relationship between the lifetime and the linewidth $\delta\omega_{ph}$ is simply $\delta\omega_{ph} = 1/\tau_{ph}$ [30].

It can be useful to characterize microcavities by their quality. To do that we define the quality factor of a cavity with a resonant cavity frequency ω_{ph} and linewidth $\delta\omega_{ph}$ as

$$Q = \frac{\omega_{ph}}{\delta\omega_{ph}}. \quad (1.1.1)$$

Figure 1.1: (a) A sketch of a distributed Bragg reflector (DBR). It consists of alternating layers of two materials of differing refractive indices (here n_1 and n_2). (b) Reflectivity of a planar microcavity made using DBRs. The resonant wavelength λ can be chosen arbitrarily. Figure taken from *Wikipedia*, <https://en.wikipedia.org/wiki/File:DBRREFL.png>

We can consider Q^{-1} as the fraction of energy a photon loses while making a single round-trip of the cavity. In state of the art experiments, the Q -factor of a planar microcavity can reach 320,000 [31].

A commonly used Fabry-Pérot cavity used for cavity QED purposes is a *planar microcavity*, where the two mirrors are two planar DBRs. There are other types, such as the *Spherical mirror microcavities* that function similarly to the planar ones, but exchange one of the mirrors out for a mirror with spherical curvature [32], or have both mirrors with positive curvature [33].

It is also possible to reach extremely high Q values using the same principle as the acoustic whispering galleries, most famously found in St Paul's Cathedral in London, where a whisper close to the edges of a dome shaped structure can be heard clearly from the other end. These are called *whispering gallery modes* (WGM) and the concept behind them depends on reaching total internal reflection, where a light beam is completely reflected when it encounters an interface to a lower refractive index material under a large enough incident angle. The WGM resonators are made

Figure 1.2: A sketch of a spherical whispering gallery mode resonator. The inset shows a photograph of a such a resonator made from silica and doped with rare earth. Figure taken from Ref. [34]

out of disks or spheres, where light is shone along the edge leading it to bounce of the walls ending up trapped on the circular path. A sketch of a silica microsphere WGM resonator can be seen in figure 1.2. The inset shows a silica sphere doped with rare earth atoms [34]. Resonators of this type can reach ultra-high values (*e.g.* $Q = 8 \times 10^8$ reported in [35]), but since the mechanism at play is bound by the light bouncing multiple times along the edge, these resonators have a limit to how small they can be made. The number of bounces also means that the light will be of a high optical mode, which is not always preferable. That is not to say that it is impossible to do cavity QED with a WGM, strong coupling of atoms and a WGM was reported in 1998 [36] and they continue to stay relevant [37–39].

Photonic crystals are made from periodic dielectric structures where the light travelling inside is analogous to electrons travelling in a periodic lattice of atoms or molecules, the periodic potential felt by the electrons being replaced by a periodic dielectric function. By engineering the dielectric function of the system it is possible to recreate similar phenomena for the photons, as electrons experience in the varying crystal types [40]. One of those phenomena is the opening of complete band gaps, where propagation is thwarted. By opening up a photonic band gap in a low-loss material it is possible to confine light. In principal this confinement could be in one, two, or three dimensions.

Figure 1.3: A high Q -factor cavity in a 2D photonic crystal (PC). The crystal consists of a triangular lattice of air holes with lattice constant a . The cavity consists of a defect, where three holes in a line are missing. (a) The Q -factor can be enhanced by displacing the two air holes at each end. (b) an even greater enhancement is achieved by shifting three holes on each side, instead of only one. Figure taken from Ref. [41].

Using photonic crystals as 2D microcavities has also shown promise, where a Q -factor of 10^5 was reported by Ref. [41] (see figure 1.3). The main drawbacks of this procedure are however the large surface area of such crystals compared to the active region, as well as their difficulty in fabrication [30].

1.2 Coupling strength

Once we have sufficiently confined light, we want to study how it couples to matter. To fully study systems of light-matter coupling, and to distinguish weak and strong coupling, we need to be able to quantify the strength of the coupling. This is done by a parameter called *coupling strength*, g , which has the unit of energy. It is calculated by simply taking the inner product of the dipole moment of the matter mode, $\mathbf{d}$, and the electric field making up the light mode,

$$g = \hat{\mathbf{d}} \cdot \mathcal{E}. \quad (1.2.1)$$

The amplitude of the electric field $\mathcal{E}$ will depend on the photon energy ε_{ph} and the cavity volume V in the following way [42]

$$|\mathcal{E}| = \sqrt{\frac{\varepsilon_{ph}}{2\epsilon\epsilon_0 V}}, \quad (1.2.2)$$

where ϵ and ϵ_0 are the relative and vacuum permittivity inside the cavity, and V is the mode volume.

In a quantum system, the dipole matrix elements between two eigenstates i and j is calculated as follows

$$\mathbf{d}_{if} = \langle f | e\hat{\mathbf{r}} | i \rangle, \quad (1.2.3)$$

where e is the elementary charge and $\hat{\mathbf{r}}$ is the real space position operator. To calculate this further we need to know the wave function of the states in question.

It is important to note that even though the dispersion relation of light is normally linear, here we are dealing with light that is confined in at least one direction. That means that we need to take into account that there will be some quantization of the momentum and the light dispersion of the first mode for a 2D photon (where the confinement length L_c will take the form

$$\begin{aligned} \varepsilon_{ph}(\mathbf{k}) &= \hbar c |\mathbf{k}| = \hbar c \sqrt{\mathbf{k}_\perp^2 + \left(\frac{\pi}{L_c}\right)^2}, \\ &\approx \hbar c \frac{\pi}{L_c} \left[1 + \frac{k_\perp^2}{2} \left(\frac{L_c}{\pi}\right)^2 - \frac{k_\perp^4}{8} \left(\frac{L_c}{\pi}\right)^4 + \dots \right] \end{aligned} \quad (1.2.4)$$

where $\mathbf{k}_\perp$ is the 2D wave vector perpendicular to the confinement direction. Looking at this formula we see that when $\mathbf{k}_\perp$ is small compared to $\frac{\pi}{L_c}$ the dispersion will be parabolic. When we are working in this range it can be useful to define the effective mass of the photon m_{ph} as well as the detuning of the photon mode δ . This is done

by writing the photon energy as

$$\varepsilon_{ph}(\mathbf{k}_\perp) \approx \frac{\hbar^2 k_\perp^2}{2m_{ph}} + \varepsilon + \delta, \quad (1.2.5)$$

where ε is the excitation energy of the matter mode that the light is coupling to. By comparing equation 1.2.5 with the Taylor series of equation 1.2.4, we find that

$$m_{ph} = \frac{\pi}{cL_c}, \quad \delta = \frac{c\hbar\pi}{L_c} - E_g. \quad (1.2.6)$$

When the light is confined in two directions the photon will be effectively 1D instead of 2D, but the results are similar. The spectrum will remain parabolic but when calculating the effective mass and detuning the term $\left(\frac{\pi}{L_c}\right)^2$ will be more complicated, and will depend on the geometry of the cross-section (whether the cavity will be rectangular or cylindrical, for example). Effective photon masses are typically engineered to be around $10^{-4} - 10^{-5}$ times the free electron mass.

The case of the 0D photon will however be qualitatively different. With confinement in all directions the photon momentum will be fully quantized, resulting in discrete energy levels. The momentum of such a photon will be

$$\mathbf{k} = \left(\frac{n_x\pi}{L_x}, \frac{n_y\pi}{L_y}, \frac{n_z\pi}{L_z} \right) \quad (1.2.7)$$

where $n_i = \pm 1, \pm 2, \dots$ and L_i is the confinement length along i axis, $i = x, y, z$. Therefore, the ground state energy of a 0D photon will be

$$\varepsilon_{ph}(\mathbf{k}) = \hbar c \sqrt{\left(\pm \frac{\pi}{L_x}\right)^2 + \left(\pm \frac{\pi}{L_y}\right)^2 + \left(\pm \frac{\pi}{L_z}\right)^2}. \quad (1.2.8)$$

If we assume that $L_x = L_y = L_z \equiv L$ this can be simplified to

$$\varepsilon_{ph}(\mathbf{k}) = \frac{\sqrt{3}\pi\hbar c}{L}. \quad (1.2.9)$$

Figure 1.4: The structure consists of a quantum well layer sandwiched between two distributed Bragg reflectors (DBRs).

1.2.1 Exciton-Polariton

To demonstrate this we will look at a specific, well studied system consisting of a semiconductor quantum well (QW) embedded into a microcavity. The QW will have both electrons and holes, which under the right circumstances can form bound pairs called *excitons*. The frequency of cavity light mode is tuned to be close to the exciton mode of the QW.

To study the system we choose to write down its Hamiltonian density using the formalism of second quantization,

$$\mathcal{H}_k = \varepsilon_{ph}(k)\phi_k^\dagger\phi_k + \varepsilon_X(k)\chi_k^\dagger\chi_k + g\left(\phi_k^\dagger\chi + \chi_k^\dagger\phi_k\right) \quad (1.2.10)$$

where ε_{ph} and $\phi^{(\dagger)}$ are the photon energy and annihilation (creation) operator, respectively, and ε_X and $\chi^{(\dagger)}$ are the same for the exciton. The coupling constant, discussed in the previous chapter, is denoted by g .

This Hamiltonian can be diagonalized by transforming the operators to

$$\psi_{L,k} = A_k \phi_k + B_k \chi_k, \quad (1.2.11)$$

$$\psi_{U,k} = B_k \phi_k + A_k \chi_k. \quad (1.2.12)$$

Equation (1.2.10) can then be rewritten as

$$\mathcal{H}_k = \varepsilon_U(k) \psi_{U,k}^\dagger \psi_{U,k} + \varepsilon_L(k) \psi_{L,k}^\dagger \psi_{L,k} \quad (1.2.13)$$

the energy spectrums ε_U and ε_L satisfying

$$\varepsilon_{U,L}(k) = \frac{\varepsilon_X(k) + \varepsilon_{ph}(k)}{2} \pm \frac{1}{2} \sqrt{(\varepsilon_X(k) - \varepsilon_{ph}(k))^2 + 4g^2} \quad (1.2.14)$$

where the + solution corresponds to an upper polariton branch (U) while the - solution corresponds to a lower polariton branch (L). A qualitative picture of these dispersions can be seen in figure 1.5 where the detuning of the bare photon mode is set to negative, zero and positive, going from left to right respectively. The bare modes are plotted in blue while the coupled modes are in red.

Typical Rabi splittings in GaAs and CdTe based materials lie in the range of 5-20 meV, below the thermal energy at room temperature, which is why many experiments are performed under cryogenic conditions. ZnO, GaN, and perovskite materials have been used to achieve higher Rabi splittings. The latter are particularly promising for applications (*e.g.* lasing [43]) given the relatively low cost in their fabrication.

It can be of interest to know the photon and exciton fractions of the two polariton modes. We can study this by calculating the so called Hopfield coefficients A_k and B_k , named after J. J. Hopfield who is known for predicting the exciton-polariton [44]. These coefficients can be seen as the exciton or photon fraction of the exciton branches. We find that they are given by the expressions

$$A_k^2 = \frac{g^2}{(\varepsilon(k) - \varepsilon_X(k))^2 + g^2} \quad B_k^2 = \frac{g^2}{(\varepsilon(k) - \varepsilon_{ph}(k))^2 + g^2}. \quad (1.2.15)$$

Figure 1.5: The bare photon and exciton dispersion are shown in blue while the coupled mode is seen in red. From left to right the graphs show the dispersion for when the detuning, $\delta = \varepsilon_{ph}(0) - \varepsilon_X(0)$, is negative, zero and positive detuning ($\delta < 0, \delta = 0$ and $\delta > 0$, respectively).

As we might expect, when a polariton mode lies close to either uncoupled mode, the coefficient corresponding to that mode will be close to unity (and the other close to zero, since $A_k^2 + B_k^2 = 1$ holds) but around the anti-crossing of the modes the polariton will be roughly half-photon and half-exciton.

The fact that the polariton dispersion is non-parabolic can have some interesting effects [45]. For example, when the lower polariton branch is pumped close to the inflection point of the dispersion, a pair of polaritons can scatter off each other into a signal and idler mode of lower and higher energy, respectively, all while conserving both momentum and energy. This is called parametric scattering, and can be seen as a polariton four wave mixing process [46, 47]. This four wave mixing leads to a solution of spatial solitons that are localized in the direction perpendicular to the pump momentum, called parametric polariton solitons [48].

1.3 Low dimensional systems

The dimensionality of a system can have a large effect on the physics that govern it. The first thing that comes to mind is how the dimensionality of a system controls how its volume scales, or $V \sim L^d$ if V is the volume, L is characteristic length and d the dimensionality. Because of this, the integral measure will depend on the dimensionality which can mean that different interactions between two points can have vastly different behaviours in different dimensions. The best known case of this is how the Coulomb potential becomes a non-integrable singularity in 1D, as has been much studied by mathematicians [49].

There are also fundamental phenomena that are restricted to certain dimension. For example how the Ising model does not have a phase transition in 1D but does for dimensions 2 and higher [50]. Similarly, in continuous systems the Mermin–Wagner theorem, which states that continuous symmetries cannot be spontaneously broken at finite temperature in systems with sufficiently short-range interactions, holds for dimensions ≤ 2 [51]. Taking this further leads to the result that it is not possible to achieve superconductivity in a purely 1D system [52]. (Note that superconductivity has been achieved in quasi-1D systems, where a bundle of 1D rods can interact

between each other [53]).

Another thing that is heavily influenced by the dimensionality is the density of states. The narrow peak caused by the density of states in a 1D system, called the Van Hove singularity, has even been predicted to couple strongly with light leading to so called Van Hove polaritons [54].

1.3.1 Density of states

The density of states (DoS) is a measure of how many available states there are for a given energy. It depends on two factors: The dispersion relation $\varepsilon(k)$ of said system and the dimensionality. The DoS is defined as

$$D(\varepsilon) = \frac{d\Omega(k)}{d\varepsilon} = \frac{d\Omega(k)}{dk} \left(\frac{d\varepsilon}{dk} \right)^{-1}, \quad (1.3.1)$$

where $\Omega(k)$ is the volume of the reciprocal space for wave vectors less than or equal to k , which depends on the dimensionality. The derivative is

$$\frac{d\Omega(k)}{dk} = \begin{cases} 2, & \text{in 1D} \\ 2\pi k, & \text{in 2D} \\ 4\pi k^2 & \text{in 3D.} \end{cases} \quad (1.3.2)$$

The concept of the DoS is generally only used for one or more dimensions since the volume of the reciprocal space is not well defined in zero dimensions. Since energy levels in 0D systems are discrete they are by extension countable and we can define the number of states of that energy as the DoS.

Consider the parabolic dispersion, $\varepsilon = \varepsilon_0 + \alpha k^2$. Here $\frac{d\varepsilon}{dk} = 2\alpha k$ and $k = \sqrt{\frac{\varepsilon - \varepsilon_0}{\alpha}}$,

Table 1.1: The density of states for two common dispersion relation, the parabolic ($\varepsilon = \varepsilon_0 + \alpha k^2$) and linear ($\varepsilon = \alpha k$). A plot of these functions can be seen in figure 1.6.

DoS	Parabolic $\varepsilon = \varepsilon_0 + \alpha k^2$	Linear $\varepsilon = \alpha k$
1D	$\frac{1}{\sqrt{\alpha(\varepsilon - \varepsilon_0)}}$	$\frac{1}{\alpha}$
2D	$\frac{\pi}{\alpha}$	$\frac{2\pi}{\alpha^2} \varepsilon$
3D	$\frac{2\pi}{\alpha^{3/2}} \sqrt{\varepsilon - \varepsilon_0}$	$\frac{4\pi}{\alpha^3} \varepsilon^2$

so in 3D

$$\begin{aligned}
D(\varepsilon) &= \frac{d\Omega(k)}{dk} \left(\frac{d\varepsilon}{dk} \right)^{-1} \\
&= 4\pi k^2 / 2\alpha k \\
&= \frac{2\pi k}{\alpha} \\
&= \frac{2\pi}{\alpha^{3/2}} \sqrt{\varepsilon - \varepsilon_0}.
\end{aligned} \tag{1.3.3}$$

Table 1.1 shows the resulting DoS for 1D, 2D and 3D for both the case of parabolic and linear ($\varepsilon = \alpha k$) dispersion, and their plots can be seen in figure 1.6.

For a more detailed demonstration, let us consider the DoS for the exciton polaritons in the previous subsection. The dimensionality is 2D and the dispersion relation is given by equation 1.2.14. For simplicity we will take the exciton-photon detuning $\delta = 0$ and neglect terms of the order $\mathcal{O}\left(\frac{m_{ph}}{m_X}\right)$, since $m_{ph} \ll m_X$. First, we calculate the derivative of the upper polariton branch with respect to the momentum k

$$\begin{aligned}
\frac{\partial \varepsilon_U}{\partial k} &= \frac{1}{2} \frac{\partial(\varepsilon_X + \varepsilon_{ph})}{\partial k} + \frac{1}{2} \frac{2(\varepsilon_X - \varepsilon_{ph})}{2\sqrt{(\varepsilon_X - \varepsilon_{ph})^2 + 4g^2}} \frac{\partial(\varepsilon_X - \varepsilon_{ph})}{\partial k} \\
&= \frac{\hbar^2}{2m_{ph}} k + \frac{1}{\sqrt{\left(\frac{\hbar^2 k^2}{2m_{ph}}\right)^2 + 4g^2}} \left(\frac{\hbar^2}{2m_{ph}} \right)^2 k^3.
\end{aligned} \tag{1.3.4}$$

Figure 1.6: The density of states for parabolic dispersion (left hand column) and linear dispersion (right hand column). The first row shows the DoS for 1D, the middle row shows 2D and the bottom row 3D.

The DoS, as a function of the momentum, then becomes

$$\begin{aligned}
D(k) &= \frac{d\Omega(k)}{dk} \left(\frac{d\varepsilon}{dk} \right)^{-1} \\
&= 2\pi k \left[\frac{\hbar^2}{2m_{ph}} k + \frac{1}{\sqrt{\left(\frac{\hbar^2 k^2}{2m_{ph}} \right)^2 + 4g^2}} \left(\frac{\hbar^2}{2m_{ph}} \right)^2 k^3 \right]^{-1} \\
&= \frac{4\pi m_{ph}}{\hbar^2} \left[1 + \frac{1}{\sqrt{\left(\frac{\hbar^2 k^2}{2m_{ph}} \right)^2 + 4g^2}} \frac{\hbar^2 k^2}{2m_{ph}} \right]^{-1}. \tag{1.3.5}
\end{aligned}$$

Now we need to rewrite the DoS to be a function of the energy ε instead of k . Using the photon dispersion defined in 1.2.5 we get the upper polariton branch

$$\begin{aligned}
\varepsilon_U &= \varepsilon_g + \frac{1}{2} \frac{\hbar^2 k^2}{2m_{ph}} + \frac{1}{2} \sqrt{\left(\frac{\hbar^2 k^2}{2m_{ph}} \right)^2 + 4g^2} \\
\frac{\hbar^2 k^2}{2m_{ph}} &= \varepsilon_U - \varepsilon_g - \frac{g^2}{\varepsilon_U - \varepsilon_g}. \tag{1.3.6}
\end{aligned}$$

After combining equations (1.3.5) and (1.3.6) and rearranging the terms, we find that the density of states for the upper branch of the exciton-polariton is

$$D_U(\varepsilon) = \frac{2\pi m_{ph}}{\hbar^2} \left(1 + \frac{g^2}{\varepsilon^2} \right). \tag{1.3.7}$$

The momentum k is a physical, real quantity that must obey the condition that equation (1.3.6), which is proportional to k^2 , must be greater or equal to 0. This means that equation (1.3.7) only holds when $\varepsilon - \varepsilon_g \geq g$. Outside of that range, the DoS is 0.

Repeating these calculations for the lower polariton branch we arrive at the same formula

$$D_L(\varepsilon) = \frac{2\pi m_{ph}}{\hbar^2} \left(1 + \frac{g^2}{\varepsilon^2} \right), \tag{1.3.8}$$

but now the validity range is $-g \leq \varepsilon - \varepsilon_g < 0$. The DoS diverges at $\varepsilon = \varepsilon_g$, which is to be expected when the exciton mass is neglected. In that case the lower polariton branch will be asymptotic to the flat line $\varepsilon = \varepsilon_g$ as $|k| \rightarrow \infty$ leading to infinitely many states for that energy. When the exciton mass is kept finite, the equations for the DoS become quite unwieldy so they will not be shown here. Those results can however be seen in the center panel of figure 1.7, along with the functions described in equations (1.3.7) and (1.3.8). The left panel shows the exciton polariton dispersion for comparison. The dark lines correspond to the analytical case, where the exciton mass is neglected, which results in the DoS of the lower branch to diverge at $\varepsilon = \varepsilon_g$. The light, solid lines show the DoS when the exciton mass is kept finite, and set to $\frac{m_{ph}}{m_X} = 0.1$. In that case, the DoS saturates at a constant $\frac{1}{2} \frac{m_X}{m_{ph}} = 5$, on this scale, which is what we would get for a quadratic dispersion with mass m_X in 2D. The upper branch is quite similar in both cases, starting at around 1 (or $1 - \frac{m_{ph}}{m_X}$ in the later case) but staying a constant $\frac{1}{2}$ for higher energies, as the dispersion becomes more parabolic. Realistically, the fraction of the masses is generally $\frac{m_{ph}}{m_X} \lesssim 10^{-3}$ [30]. The resulting DoS for $\frac{m_{ph}}{m_X} = 10^{-3}$ can be seen plotted in a light, dashed line. In the scale used here it is almost identical to the results from equation (1.3.8), except that the DoS does not diverge and is well defined for all $\varepsilon > \varepsilon_g - g$.

The rightmost panel of figure 1.7 shows the DoS for exciton-polaritons in 1D, with the same parameters as in the 2D case. The upper branch resembles the Von Hove singularity in all cases, where it diverges at the bottom of the branch and then tapers off towards zero for increasing energy. The lower polariton branch exhibits some interesting behaviours though. The bottom of the branch behaves similar to in the case of the upper branch, where it diverges and then decays rapidly. Here, however it will increase again. How much will depend on the relative mass between the exciton and photon. The solid, light line shows how the DOS will reach a local maxima before starting to decrease again.

The local minima in the 1D DOS is especially interesting since it is a contributing factor to something called *the bottleneck effect* which can happen during polariton condensation. For polariton condensation to occur, higher energy excitons must relax their energy, converting into polaritons during the process, and these polaritons

often further relax in energy toward the ground state. A requirement is that these relaxation processes occur faster than the decay rate of polaritons. If not, polaritons may accumulate in an uncondensed (incoherent) state near the inflection point of the polariton dispersion [55].

This bottleneck effect is influenced significantly by the dependence of energy relaxation processes on the shape of the polariton dispersion. In general, both polariton-exciton scattering and polariton-phonon scattering should be considered and their efficiency depends on the density of states and whether or not phase matching conditions can be fulfilled. In particular, near the inflection point of the polariton dispersion (also known as the bottleneck region of the dispersion), the acoustic phonon dispersion is too flat to connect different polariton wavevectors and consequently polariton-phonon scattering is inefficient. This effect is however enhanced in 1D by the nature of the DoS. The minima in the DoS of the lower branch coincides with this inflection point, leading to fewer available states to scatter into.

Polariton condensation was first observed in 2D systems [3] but turned out to be harder to achieve in 1D. The lower density of states and the consequently weaker energy relaxation processes needed higher quality factor microcavities to provide a longer lifetime to compensate and achieve polariton condensation [56]. Theoretical calculations show that in 1D, the influence of polariton-phonon scattering is very small and that the condensation here is dominated by other factors such as polariton-exciton scattering [57].

1.4 Green's function approach and Feynman diagrams

In the 1830's the mathematician George Green developed a formalism to study inhomogeneous linear differential equations. He found that instead of solving the equation

$$\mathcal{L}u(x) = f(x), \tag{1.4.1}$$

Figure 1.7: The density of states for the 2D exciton-polaritons discussed in the previous section is plotted in blue in the middle panel while the dispersion is plotted in red on the left. The darker lines correspond to the results from the analytical calculations resulting in equations (1.3.7) and (1.3.7). For comparison, the lighter solid lines show the results for the case where the exciton mass is not ignored and set at $m_X = 10m_{ph}$. For realistic parameters it is however a lot larger. The light dashed line shows the DoS when $m_X = 10^3m_{ph}$, which is a more realistic ratio. The dark solid line and the light dashed line look identical on this scale. The rightmost panel shows the DoS for exciton-polaritons in 1D, with the same parameters as in the 2D case. The upper branch resembles the Von Hove singularity in all cases, where it diverges at the bottom of the branch and then tapers off towards zero for increasing energy. The lower polariton branch exhibits some interesting behaviours though. The bottom of the branch behaves similar to in the case of the upper branch, where it diverges and then decays rapidly. Here, however it will increase again. How much will depend on the relative mass between the exciton and photon. The solid, light line shows how the DOS will reach a local maxima before starting to decrease again. The energy scale has been scaled by the coupling energy g as well as the unitless momentum $\kappa \equiv \frac{\hbar k}{g\sqrt{2m_{ph}}}$.

where $\mathcal{L}$ is a linear differential operator, he could rather solve

$$\mathcal{L}G(x, x') = \delta(x - x') \quad (1.4.2)$$

for multiple values of x' . Then, since the function $f(x)$ can be written as a sum of delta functions, the solution $u(x)$ can be written as a sum of the functions G , which are called Green's functions. These Green's functions turned out to be a powerful tool, since they describe the response of the system and they make it possible to extract important information about the system's behaviour without having to solve equation (1.4.1) directly.

In physics this approach has found broad application, since time dynamics of physical systems are typically described by partial differential equations.

Let us consider the Green's function for the Schrödinger equation for a free, massive particle. We then need to solve the following equation

$$\left(i\hbar\frac{\partial}{\partial t} - H\right) G(t, \mathbf{r}; t', \mathbf{r}') = \delta(t - t')\delta(\mathbf{r} - \mathbf{r}') \quad (1.4.3)$$

where the Hamiltonian for our case is simply $H = -\hbar^2\nabla^2/2m$. We can solve this by taking the Fourier transform of the equation in both space and time. This lands us in the reciprocal space of momentum and frequency. The equation in reciprocal space is

$$\left(E - \frac{\hbar^2\mathbf{k}^2}{2m}\right) G(E, \mathbf{k}) = 1, \quad (1.4.4)$$

so clearly the Green's function is simply

$$G(E, \mathbf{k}) = \frac{1}{E - \frac{\hbar^2\mathbf{k}^2}{2m}}. \quad (1.4.5)$$

We see that the propagator has a pole at $E = \frac{\hbar^2k^2}{2m}$, which is exactly the dispersion for a free, non-relativistic, massive particle. This is a universal quality of propagators, the eigenenergies of the system considered can be found by finding the poles of the Green's function. Particles whose energy-momentum relation follows the dispersion

are called *on-shell* and can be considered physical or real, while the calculations still include *off-shell* particles which are, on the other hand, called *virtual*.

Now, the real advantage of using the Green's functions approach in studying quantum particles lies in the generalization of the approach to cover interacting many-body systems [58–61]. The building blocks of such an approach are the Feynman propagators, vacuum expectation values of pairs of field operators

$$iG(t, \mathbf{r}; t', \mathbf{r}') = \langle \Omega | T \{ \psi(t, \mathbf{r}) \psi^\dagger(t', \mathbf{r}') \} | \Omega \rangle, \quad (1.4.6)$$

where T is the time-ordering operator, which permutes its arguments so that the time coordinate of the operators grows from right to left. If the arguments correspond to fermionic fields each permutation will introduce a minus sign. $|\Omega\rangle$ denotes the vacuum (ground) state of the system.

We can consider equation 1.4.6 as a description of the propagation of a particle created at time t' and point $\mathbf{r}'$ to a point $\mathbf{r}$ at time t , or its antiparticle going the opposite direction if $t' > t$.

Let us now write out the time-ordering of equation 1.4.6 explicitly in the case of an electron field:

$$\begin{aligned} iG(t, \mathbf{r}; t', \mathbf{r}') = & \theta(t - t') \langle \Omega | \psi(t, \mathbf{r}) \psi^\dagger(t', \mathbf{r}') | \Omega \rangle \\ & - \theta(t' - t) \langle \Omega | \psi^\dagger(t', \mathbf{r}') \psi(t, \mathbf{r}) | \Omega \rangle, \end{aligned} \quad (1.4.7)$$

$\theta(t)$ being the Heaviside step function which is zero if t is negative but unity otherwise.

The field operator can be written out as

$$\psi(t, \mathbf{r}) = \int \frac{d^3k}{(2\pi)^3} e^{i\mathbf{k}\cdot\mathbf{r}} e^{-iE_k t/\hbar} a_{\mathbf{k}}, \quad (1.4.8)$$

where $a_{\mathbf{k}}$ is the operator associated with an electron with momentum $\mathbf{k}$ and E_k is the energy of a single such electron. Using that we can rewrite the first term in equation 1.4.7 as

$$iG_{t>t'} = \theta(t-t') \iint \frac{d^3k}{(2\pi)^3} \frac{d^3k'}{(2\pi)^3} \langle \Omega | a_{\mathbf{k}} a_{\mathbf{k}'}^\dagger | \Omega \rangle e^{i\mathbf{k}\cdot\mathbf{r} - i\mathbf{k}'\cdot\mathbf{r}'} e^{-iE_k t/\hbar + iE_{k'} t'/\hbar}, \quad (1.4.9)$$

and the second term naturally takes a similar form. Looking at the term $\langle \Omega | a_{\mathbf{k}} a_{\mathbf{k}'}^\dagger | \Omega \rangle$ we see that it will disappear unless $\mathbf{k} = \mathbf{k}'$ and since in the vacuum state $|\Omega\rangle$ all states below the Fermi level are full; the term also disappears unless $k > k_F$. Similarly the term $\langle \Omega | a_{\mathbf{k}'}^\dagger a_{\mathbf{k}} | \Omega \rangle$ disappears unless $k < k_F$, and corresponds to the creation and annihilation of a hole instead of the electron described in the former term.

Another thing we can make use of is writing the Heaviside step function in its integral form

$$\theta(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{ie^{-i\omega t}}{\omega + i\delta}, \quad (1.4.10)$$

where $\delta > 0$ is an infinitesimal parameter. Now,

$$G = \int \frac{d^3k}{(2\pi)^3} \frac{d\omega}{2\pi} \left[\frac{\theta(k - k_F)}{\omega + i\delta} + \frac{\theta(k_F - k)}{\omega - i\delta} \right] e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} e^{-i(E_k + \hbar\omega)(t-t')/\hbar}, \quad (1.4.11)$$

shifting the ω integral by E_k and rewriting the terms inside the brackets in a more compact form we have

$$G(t, \mathbf{r}; t', \mathbf{r}') = \int \frac{d^3k}{(2\pi)^3} \frac{d\omega}{2\pi} \frac{1}{\omega - E_k/\hbar + i\delta \text{sgn}(k - k_F)} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} e^{-i\omega(t-t')}, \quad (1.4.12)$$

where sgn is the sign function. From there we can see that the propagator in reciprocal space is

$$G(\omega, \mathbf{k}) = \frac{1}{\omega - E_k/\hbar + i\delta \text{sgn}(k - k_F)}, \quad (1.4.13)$$

which is similar to the Green's function in equation (1.4.5), except here we have the infinitesimal parameter $\pm i\delta$ where the $+$ refers to an electron and $-$ to a hole.

Next, let us proceed with calculating the propagator for a cavity photon. Now,

the role of field operator is taken by the vector potential $A_{\mathbf{k}}(t) = a_{\mathbf{k}}(t) + a_{-\mathbf{k}}^\dagger(t)$ and we take note that here we are working with bosons. The operators $a_{\mathbf{k}}^{(\dagger)}$ are the annihilation (creation) operators of a cavity photon with wave vector $\mathbf{k}$. In this case the ground state $|0\rangle$ is empty space, with no photons present. The propagator is then

$$\begin{aligned} iD(t - t', \mathbf{k}) &= \langle 0 | T \{ A_{\mathbf{k}}(t) A_{\mathbf{k}}^\dagger(t') \} | 0 \rangle \\ &= \theta(t - t') \langle 0 | a_{\mathbf{k}}(t) a_{\mathbf{k}}^\dagger(t') | 0 \rangle + \theta(t' - t) \langle 0 | a_{-\mathbf{k}}(t') a_{-\mathbf{k}}^\dagger(t) | 0 \rangle, \end{aligned} \quad (1.4.14)$$

where we have used that since the vacuum state has no photons, $a_{\mathbf{k}}|0\rangle = 0$ for any $\mathbf{k}$ and $\langle 0 | a_{\mathbf{k}} a_{-\mathbf{k}}^\dagger | 0 \rangle = 0$. Since a photon with a wave vector $\mathbf{k}$ has a well defined energy ω_k , the time dependence of the operators in the Heisenberg picture is simply $a_{\mathbf{k}}(t) \propto e^{-i\omega_k t}$. The energy only depends on the magnitude of the wave vector so $\omega_k = \omega_{-k}$. Now we can take the Fourier transform in the time domain $\tau = t - t'$, resulting in

$$\begin{aligned} iD(\omega, \mathbf{k}) &= \int_{-\infty}^{\infty} d\tau e^{-i\omega\tau} [\theta(\tau) e^{-i\omega_k\tau} + \theta(-\tau) e^{i\omega_k\tau}] \\ &= \int_0^{\infty} d\tau e^{-i(\omega + \omega_k)\tau} + \int_{-\infty}^0 d\tau e^{-i(\omega - \omega_k)\tau} \\ &= -\frac{i}{\omega + \omega_k - i\delta} + \frac{i}{\omega - \omega_k + i\delta} \\ &= \frac{2\omega_k - 2i\delta}{\omega^2 - \omega_k^2 + 2i\omega_k\delta + \delta^2}, \end{aligned} \quad (1.4.15)$$

where the infinitesimal parameter $\delta > 0$ has been added to allow the integrals to converge. Since δ is small we can neglect higher order terms and the cavity photon propagator then becomes

$$D(\omega, \mathbf{k}) = \frac{2\omega_k}{\omega^2 - \omega_k^2 + 2i\omega_k\delta}. \quad (1.4.16)$$

1.4.1 Feynman Diagrams

Now that we have calculated the propagators for electrons, holes and cavity photons we need to know how to combine them to calculate interesting effects. For this we make use of Feynman diagrams, a pictorial description of the behaviour of particles, named after their inventor, Richard Feynman. The diagrams offer a way to calculate the probability amplitude of a given process from simply writing down the diagram for the process and then using the accompanying rules to translate it to a mathematical form.

Feynman diagrams can be a powerful tool. Before defining the rules for the diagrams we need to define our theory. In this thesis we restrict ourselves to cavity quantum electro dynamics (cQED), which is non-relativistic and describes electrons, holes and photons. The only interaction we consider is one where these three particles participate. From this we can draw diagrams describing all interactions and propagations in a cavity. The time flow in our diagrams will be from left to right.

Now, we can start looking into the physical meaning of diagrams. The simplest diagrams depict bare propagation of the three fundamental particles of the theory. We choose a solid line to represent fermions, an arrow along the flow of time meaning an electron while an arrow in the opposite direction represents the hole. This is natural, since in this theory the hole can be seen as the antiparticle of the electron and an anti-particle can formally be considered mathematically as the particle going backwards in time with negative energy. The photon is depicted by a dashed line, with no arrow since a photon is massless and therefore its own antiparticle. In this thesis we will be neglecting the spin component of the photon in our calculations and will therefore consider it as a scalar. The theory can be generalized to include the spin of both fermions and bosons by considering them as tensor objects. These three propagators can be seen in figure 1.8(a), (b) and (c), respectively.

The last element of the theory is the interaction between particles. First we can have Coulomb interaction between electrons and/or holes, which is depicted with a squiggly line, as shown in figure 1.8(d). Lastly we need to take into account an interaction which includes an electron, a hole and a photon, which describes a

Figure 1.8: Pictorial representation of propagators. (a) the conduction band electron, (b) the valence band hole, (c) photon, (d) Coulomb interaction and (e) the electron-hole-photon vertex.

process such as an electron and hole combining and emitting a photon. This we mark by a dot in the diagram, like in figure 1.8(e), and is mathematically described by the coupling constant g (which was the subject of §1.2) accompanied by a delta function to account for the conservation of momentum.

Diagrams are now classified into orders depending on how many vertices are in each one. A bare propagator can be considered a zeroth order diagram while *e.g.* an electron and hole combining to make a photon would be a first order diagram. Each diagram has an *amplitude* associated with it, which can be calculated using the Feynman rules. This amplitude is related to the probability of the process depicted in the diagram taking place. The usefulness of the Feynman diagram approach relies on the coupling constant being smaller than other energy scales in the system so that the amplitude of each order is smaller than the one before. The Feynman rules followed in this thesis are as follows:

- Write down all diagrams of the desired order
- For every line or vertex write down the corresponding propagator:
 - For each electron (hole) line, write a factor of $iG_c^0(\varepsilon, \mathbf{k})$ ($G_v^0(\varepsilon, \mathbf{k})$)
 - For each photon line, write a factor of $iD_0(\omega, \mathbf{k})$
 - For each squiggly line, write a factor of the Coulomb potential $V(\mathbf{r})$
 - For each photon-electron-hole vertex, include a factor $g\delta(k_{in} - k_{out})$
- Integrate over each internal line

- For every closed fermion loop, include a factor of -1.

Note that these Feynman rules are not universal and may differ between sources. The biggest differences can be found when looking at sources studying particle physics. There you will see many more types of particles (such as muons, the Higgs boson and the family of quarks) and the propagators for the current particles will not be the same. The current electron propagator is not Lorentz invariant and will therefore only hold at low velocities. In high energy physics the propagator needs to be able to describe propagation at relativistic speeds [59]. We have also ignored the polarization of the light. When that is included the photon propagator becomes a tensor.

Other, more specific differences in the Feynman formalism can be seen while considering sources on similar topics to the ones considered here. One example is how Keldysh formalism can be integrated into the Green's function approach to study non-equilibrium occupation of states [62]. This means that the propagators are not calculated by integrating over the normal Feynman contour (which we introduced through the Heaviside function $\theta(t - t')$) but instead a special Keldysh contour is used. Another interesting example is how the diagrammatic approach is used in Ref. [63] in connection with the second quantization Hamiltonian and related operators, but not the Green's functions which are normally associated with these diagrams.

1.4.2 Approximations

Although Feynman diagrams can be a powerful tool for calculation there is always a need to make approximations when it comes to physical systems. We have even already made a few approximations when setting up our model. When we decide to only consider photons, electrons and holes we have neglected the account of other high energy particles, such as positrons, muons and gluons. The amplitudes of diagrams including these particles are next to zero in the energy range considered in condensed matter systems, so we can safely disregard them.

We have also assumed that all momentum is conserved, which implies spatial homogeneity. This does not always hold true in condensed matter systems and we

need to be careful to not impose this formalism blindly.

After accepting the approximations made when forming the model we use, we run into another problem. We find that when writing down all possible diagrams that contribute to a process, the number and complexity grows quickly. Realistically we do need to make some approximations and restrict what diagrams to include in our calculations. When the coupling constant is small, the Feynman diagram approach fits perturbation theory very well. Since the amplitude will decrease with growing orders of diagrams, it is natural to calculate only lower order terms. Let us go over the most common approximations used in regards to cQED systems.

Self-Energy and the Hartree-Fock approximation

A particle propagating in a given system can go through any number of interactions when going from one point in space-time to another. To account for this we can renormalize the propagator by summing up all diagrams where we start and end with a single particle. The resulting propagator is said to be *dressed* while the propagator that includes no interactions is called *bare*. Figure 1.9(a) Shows some examples of terms we need to account for to calculate the dressed propagator. Note that even though we might conceive of diagrams where two aspects of the diagram are not connected, like depicted in 1.10(a), they are not included in the renormalization. It turns out that these so called *vacuum bubbles* are a property of the vacuum and can be factored out (see *e.g.* §4.4 in reference [59]). In the end they do not have any effect on physical quantities except in special cases, like in the Casimir effect [64].

Let us distinguish between reducible and irreducible diagrams. An irreducible diagram can not be made disconnected by cutting a single fermion line, while a reducible diagram can. In other words a reducible diagram can be written as a multiplication of one or more irreducible ones. Looking at term III in figure 1.9(a) we see that it can be considered a combination of the terms I and II. We can reduce the number of diagrams we need to calculate for the renormalization by defining the self-energy operator Σ which is a sum over all irreducible diagrams. Using this operator we can write the infinite sum for the renormalization on a simpler form,

Figure 1.9: (a) The renormalized electron propagator (thick line) is in principle calculated by summing up all possible self interaction diagrams. The terms marked *I* and *II* the Hartree-Fock terms, *III* is an example of a reducible term (which can be made by splicing terms *I* and *II* together) and *IV* is an example of a higher-order term that because cumbersome to calculate. (b) The renormalized propagator can be rewritten in the form shown, where Σ represents the self energy and holds all irreducible terms. The equation in (b) is called the Dyson equation and can be written in the recursive form depicted in (c).

Figure 1.10: (a) An example of a diagram including a vacuum bubble. These types of diagrams, where a part of the diagram is disconnected from the rest, do not change the physical properties of the propagator. They can therefore be excluded from the self-energy calculations. (b) The polarisation operator Π_0 is described by an electron-hole bubble, which portrays the event when an electron-hole pair is created and at a later time combines.

shown in 1.9(b). This can formally be written as

$$\mathbf{G} = G_0 + G_0 \Sigma G_0 + G_0 \Sigma G_0 \Sigma G_0 + \dots \quad (1.4.17)$$

where $\mathbf{G}$ and G_0 are the dressed and bare propagators, respectively. We see that this is a simple geometric series, which can be written in a recursive form shown pictorially in 1.9(c) and has the solution

$$\begin{aligned} \mathbf{G} &= G_0 + G_0 \Sigma \mathbf{G} \\ &= \frac{G_0}{1 - G_0 \Sigma}. \end{aligned} \quad (1.4.18)$$

Taking the bare propagator from equation (1.4.13) we see that the dressed propagator becomes

$$\mathbf{G}(\omega, \mathbf{k}) = \frac{1}{\omega - (E(\mathbf{k}) + \Sigma) \pm i\delta}. \quad (1.4.19)$$

Here it becomes clear why Σ is dubbed self-energy. It simply shifts the energy of the particle in question and is a result of the particle interacting with itself or the vacuum.

Looking back at the terms included in the self-energy we see that the higher order terms can become complicated and are daunting to calculate, like for example the term marked IV in figure 1.9. Calculating every term is an insurmountable task

so we need to find fitting approximations.

More often than not the coupling constant turns out to be small, which means that perturbation theory in the order of diagrams is a natural choice. A common approximation is taking only the first order terms (the ones marked *I* and *II* in figure 1.9a) into account. This is called the Hartree-Fock approximation. This is often a good approximation for the energy shift caused by self interactions of a particle.

Random Phase Approximation

In many cases it becomes important to estimate contributions caused by interaction between particles. The random phase approximation (RPA) is commonly used in systems such as a highly concentrated electron gas. It was first introduced using the formalism of classical electrodynamics [65–67] but was later also derived using an infinite series of Feynman diagrams, in a similar form to the Dyson series [68]. Here we will give a brief description of the latter approach.

The RPA was introduced to study plasma oscillations, which are density modulations in a dense Fermi gas. The energy spectrum of these oscillations can be calculated by finding where the dielectric function of the electron gas is zero. The dielectric function is given by

$$\varepsilon(\omega, \mathbf{k}) = 1 - V(\mathbf{k})\Pi(\omega, \mathbf{k}) \quad (1.4.20)$$

where $V(\mathbf{k})$ is the Coulomb potential in reciprocal space and Π is the polarization operator. The polarization operator describes the process of a photon exciting an electron to form an electron-hole pair that can then interact before it recombines, reemitting a photon. The bare propagator Π_0 is represented with a simple fermion bubble, like shown in figure 1.10(b). In the RPA the polarization operator is renormalized by accounting for Coulomb interactions between different such bubbles, like shown in figure 1.11. The infinite sum has the same form as the Dyson equation in the previous chapter, so the solution becomes

Figure 1.11: A pictorial representation of the random phase approximation (RPA). The approximation revolves around renormalizing the polarization operator (made up of an electron-hole bubble) by summing up the bare operator interacting any number of times with another bubble. The infinite series depicted in (a) can be rewritten as a recursive form, seen in (b), similar to the Dyson equation shown in figure 1.9 (b) and (c).

$$\Pi(\omega, \mathbf{k}) = \frac{\Pi_0(\omega, \mathbf{k})}{1 - V(\mathbf{k})\Pi_0(\omega, \mathbf{k})} \quad (1.4.21)$$

The form of both V and Π_0 will depend on the dimensions of the system under consideration. Details about the calculations can be found in textbooks, for example Refs. [42, 69], so here we will make due with stating the results. For example, the resulting plasmon frequency spectrum for three dimensional Fermi gas shows a quadratic behaviour

$$\omega^{3D}(\mathbf{k}) = \omega_p + \frac{3}{10} \frac{v_f^2}{\omega_p} k^2, \quad (1.4.22)$$

where ω_p is the classical plasmon frequency and v_f is the Fermi velocity.

The name Random Phase approximation stems from the fact that the approximation is based on the assumption that the quantum phase carried by the particles changes quickly enough in the relevant time scale that terms that include different wave vectors (more complicated Fermion loops than the ones shown in figure 1.11(a)) average out.

Excitons and ladder diagrams

Although the Feynman diagram formalism can be a powerful tool, it is not always the best choice. An example of a system where this formalism can become more cumbersome than other known methods is the description of two-particle bound states such as excitons. To study such a system we need to consider a two-particle propagator, which has two incoming states and two outgoing states compared to the single in- and out coming states of a single particle function. In the case of excitons the incoming and outgoing states both consist of an electron and hole pair. The exciton propagator (describing an incoming electron and hole, and the same outgoing states) is renormalized in a similar manner as before. The self-interaction of the electron/hole is taken into account by using the dressed single particle propagator, so we only need to account for interactions between the two.

Figure 1.12 depicts the renormalization in the ladder approximation, where the infinite sum corresponds to a continuous interaction between the electron and hole. The sum can be written in a recursive form similar to the Dyson equation, but in this case the right hand side of the equation will include an integral over the operator Γ , therefore it can not be separated as before. This equation is called the Bethe-Salpeter equation and can successfully be used to describe excitons [70] as well as other two-body bound states such as Cooper pairs (two electrons) and mesons (quark-antiquark). Solving this equation is not trivial however, and typically it is more feasible to use a Hamiltonian approach where the excitons are treated like bosons.

Figure 1.12: Pictorial representation of the ladder approximation. This series describes the renormalization of a two-particle propagator which includes an electron and a hole. The series describes interactions between the electron-hole pair, which results in a bound state called an exciton. This equation is called the Bethe-Salpeter equation.

2. Band gaps induced by vacuum photons in closed semiconductor cavities

Like mentioned in the previous chapter, the coupling between light and matter needs to exceed a certain threshold to enter the strong coupling regime, where the behaviour of the electron will be affected in a qualitative way. Strong coupling can be achieved by embedding a system of material modes inside a microcavity. These cavities quantize the light modes in one or more directions, leading to a finite vacuum electric field amplitude. This opens up the possibility of electrons interacting with the vacuum fluctuations of the electromagnetic field. These vacuum fluctuations are characterized by the zero-point energy, which can be made reasonably large inside a microcavity. In the diagrammatic approach the zero-point energy manifests as photons in the ground state. These photons are called virtual since the ground state cannot radiate. Interestingly, recent theoretical work has however showed that in certain circumstances these virtual photons can be converted to real ones, showing that the distinction between the two might not be as fundamental as often thought [71].

The work in this chapter is inspired by the fact that there is no formal physical difference between these so called virtual photons (quanta of vacuum fluctuations) and real ones (quanta of electromagnetic wave), so it is theoretically possible to open energy band gaps by coupling electrons to a vacuum photonic mode inside a cavity.

In previous papers on this subject [72–74] the modification to the electron dis-

Figure 2.1: (a) A sketch of the system under consideration. A slab of semiconductor (SC) is embedded in a cavity made out of distributed Bragg reflectors (DBR) with side lengths L_x, L_y and L_z . The lengths are chosen so that the electron dispersion in the SC can be assumed continuous while the light inside the cavity is quantized in all 3 directions.

persion was calculated for various open cavities (one- and two-dimensional) but the resulting vacuum-induced band gap opening turned out to be too small to measure experimentally. In this chapter we demonstrate that for *closed* (zero-dimensional) microcavities this band gap opening is larger, and large enough to be measurable in state-of-the-art experiments.

2.1 Model

In this chapter we will consider a slab of semiconductor embedded in a zero-dimensional (0D) cavity, as depicted in figure 2.1. The lengths of the cavity are denoted by L_x, L_y and L_z and are macroscopically large compared to the de Broglie wavelength of the electron. Under these conditions the light modes are quantized while the electrons can still be considered continuous. The dispersion relations for electrons in the valence (v) and conduction (c) bands are therefore the familiar ones of bulk electrons in a semiconductor:

$$\varepsilon_{v,c}(\mathbf{k}) = \pm \left(\frac{\varepsilon_g}{2} + \frac{\hbar^2 k^2}{2m_{v,c}} \right), \quad (2.1.1)$$

where ε_g is the band gap of the semiconductor and $m_{v(c)}$ is the effective mass in the valence (conduction) band.

The presence of the vacuum photon modes enhances the probability of virtual transitions between the valence and conduction bands, the process seen depicted in figure 2.1(b). These transitions lead to a modification of the bare dispersions in equation (2.1.1). Here we will use the diagrammatic Green's functions approach described in §1.4 to renormalize the electron dispersion. Here we will focus on the renormalization of the valence band and the calculations for the conduction band will be a straightforward generalization.

We begin by writing down the Feynman diagram depicting the process we are considering. Figure 2.2(a) shows the renormalized valence band propagator G_v as an infinite sum of the bare propagator G_v^0 and differing number of transitions to the conduction band (G_c^0) by means of a photon (D_0). The bare propagators are the same as the ones discussed in §1.4:

$$G_v^0(\varepsilon, \mathbf{k}) = \frac{1}{\varepsilon - \varepsilon_v + i0}, \quad G_c^0(\varepsilon, \mathbf{k}) = \frac{1}{\varepsilon - \varepsilon_c - i0}, \quad (2.1.2)$$

$$D^0(\nu, \mathbf{q}) = \frac{2\omega(\mathbf{q})}{(\nu + \omega(\mathbf{q}) + i0)(\nu + \omega(\mathbf{q}) - i0)}, \quad (2.1.3)$$

respectively.

The infinite sum is the same as the Dyson equation, and can therefore be contracted to the recursive form depicted in figure 2.2(b), which in the mathematical form can be written as

$$G_v(\varepsilon, \mathbf{k}) = \frac{G_v^0(\varepsilon, \mathbf{k})}{1 - \Sigma_v(\varepsilon, \mathbf{k})G_v^0(\varepsilon, \mathbf{k})}, \quad (2.1.4)$$

where the self-energy for the valence electrons interacting with the cavity photon mode is defined by the diagram in figure 2.2(c). It corresponds to the expression

$$i\Sigma_v(\varepsilon, \mathbf{k}) = \sum_{\mathbf{k}'} g^2(\mathbf{k}', \mathbf{k}) \int \frac{d\nu}{2\pi} iD^0(\nu) iG_c^0(\varepsilon - \nu, \mathbf{k}'), \quad (2.1.5)$$

(a)

(b)

(c)

Figure 2.2: (a) The Dyson equation for renormalized electron Green's functions corresponding to the valence band. A propagating valence electron can absorb a photon and jump to the conduction band and at a later time re-emit the photon and relax back down to the valence band. This can happen anywhere from never to infinite amount of times, leading to an infinite sum. (b) The same equation as in (a) written in a recursive form. (c) The self-energy operator responsible for photon-induced dressing of the valence band. The dashed line corresponds to a virtual cavity photon and g denotes the electron-photon coupling constant.

with g denoting the light-matter coupling constant.

The modified valence electron dispersion relation $\varepsilon(\mathbf{k})$ can be extracted from the renormalized propagator by finding the poles of the Green's function in equation (2.1.4). In other words, we need to solve

$$1 - \Sigma_v(\varepsilon, \mathbf{k})G_v^0(\varepsilon, \mathbf{k}) = 0. \quad (2.1.6)$$

The solution of this equation depends on the self-energy, which includes the coupling constant g . As previously discussed in §1.2, the coupling constant can be written as

$$g(\mathbf{k}', \mathbf{k}) = |\langle \psi_c(\mathbf{k}'), 0 | \mathbf{d} \mathcal{E} | \psi_v(\mathbf{k}), 1 \rangle|, \quad (2.1.7)$$

where $\psi_{c(v)}(\mathbf{k})$ is the wave function for an electron in the conduction (valence) band, the numbers there after are the photon occupation numbers of the ground state cavity mode, $\mathbf{d}$ is the electric dipole moment operator and $\mathcal{E}$ is the electric field corresponding to the photon mode. We define the wave vector of the ground state photonic modes as

$$\mathbf{q} = \left(\frac{n_x \pi}{L_x}, \frac{n_y \pi}{L_y}, \frac{n_z \pi}{L_z} \right), \quad (2.1.8)$$

where $n_i = \pm 1$ and all permutations of signs is allowed, leading to $2^3 = 8$ degenerate ground state modes. Using the expression for the electric field given in equation (1.2.2) we can then say that the coupling constant becomes

$$g^2(\mathbf{k}', \mathbf{k}) = \frac{|d_{cv}|^2}{16} \frac{\omega}{\epsilon V} \sum_{\mathbf{q}} \delta_{\mathbf{k}', \mathbf{k}-\mathbf{q}}, \quad (2.1.9)$$

where ϵ is the absolute permittivity of the material, as before, and the dipole moment matrix element is given by

$$|d_{cv}|^2 = \frac{e^2 \hbar^2}{2m_c E_g}. \quad (2.1.10)$$

Combining equations (2.1.5) and (2.1.9) we get

$$\begin{aligned}\Sigma_v(\varepsilon, \mathbf{k}) &= i \frac{|d_{cv}|^2}{16} \frac{\omega}{\epsilon V} \sum_{\mathbf{q}} \int \frac{d\nu}{2\pi} D^0(\nu) G_c^0(\varepsilon - \nu, \mathbf{k} - \mathbf{q}) \\ &= i \frac{|d_{cv}|^2}{16} \frac{\omega}{\epsilon V} \sum_{\mathbf{q}} \int \frac{d\nu}{2\pi} \frac{2\omega}{(\nu - \omega + i0)(\nu + \omega - i0)(\varepsilon - \nu - \varepsilon_c(\mathbf{k} - \mathbf{q}) - i0)}.\end{aligned}\quad (2.1.11)$$

Using Cauchy's residue theorem we can calculate the integral and find that the self energy is

$$\Sigma_v(\varepsilon, \mathbf{k}) = \sum_{\mathbf{q}} \frac{\omega |d_{cv}|^2}{16\epsilon V (\varepsilon + \omega - \varepsilon_c(\mathbf{k} - \mathbf{q}))}. \quad (2.1.12)$$

and equation (2.1.6) becomes

$$1 - \frac{\omega |d_{cv}|^2}{16\epsilon V} \sum_{\mathbf{q}} \frac{1}{(\varepsilon + \omega - \varepsilon_c(\mathbf{k} - \mathbf{q}))(\varepsilon - \varepsilon_v(\mathbf{k}))} = 0. \quad (2.1.13)$$

This is equivalent to

$$(\varepsilon - \varepsilon_v) \prod_{\mathbf{q}} (\varepsilon + \omega - \varepsilon_c(\mathbf{k} - \mathbf{q})) - \frac{\omega |d_{cv}|^2}{16\epsilon V} \sum_{\mathbf{q}} \prod_{\mathbf{Q} \neq \mathbf{q}} (\varepsilon + \omega - \varepsilon_c(\mathbf{k} - \mathbf{Q})) = 0. \quad (2.1.14)$$

Since the first term is a product of nine brackets containing ε (since $\mathbf{q}$ takes eight different values) the resulting equation is a 9th order polynomial.

2.2 Results

Solving 9th order polynomials is not trivial. According to the Abel-Ruffini theorem, such a polynomial does not, in general, have analytical solutions. We therefore need to either resort to making approximations to simplify the equation or to solve the equation numerically. Let us consider both cases.

2.2.1 Analytical approximation

First, let us assume that the wave vector of the photon mode, $\mathbf{q}$, can be neglected, since we know that the electron wave vector $\mathbf{k}$ is generally much larger. This is quite a naive approximation and we will discuss its validity later in the chapter, but it is common and can give good insight into the behaviour of the system.¹ So, under this approximation, equation (2.1.13) can then be rewritten as

$$1 - \frac{\omega |d_{cv}|^2}{2\epsilon V} \frac{1}{(\epsilon + \omega - \epsilon_c(\mathbf{k}))(\epsilon - \epsilon_v(\mathbf{k}))} = 0, \quad (2.2.1)$$

or

$$(\epsilon + \omega - \epsilon_c(\mathbf{k}))(\epsilon - \epsilon_v(\mathbf{k})) - \frac{\omega |d_{cv}|^2}{2\epsilon V} = 0, \quad (2.2.2)$$

if we take $\epsilon \neq \epsilon_v(\mathbf{k})$ and $\epsilon \neq \epsilon_c(\mathbf{k}) + \omega$. Since this is a second order polynomial there will be two solutions, which correspond to the mixed modes of the system. The modes that are being mixed are the one corresponding to an electron in the valence band, ϵ_v , and the one corresponding to the electron in the conduction band and an absorbed photon, $\epsilon_c - \omega$. We know that being in the strong coupling regime means that the modes will anti-cross when they mix, but when we talk about renormalizing the valence dispersion we talk about the solution closest to the uncoupled dispersion. When solving equation (2.2.2) we therefore always choose the solution closest to the bare valence solution. This solution can be written as

$$\epsilon = \frac{\epsilon_c(\mathbf{k}) + \epsilon_v(\mathbf{k}) - \omega}{2} - \frac{\eta(\mathbf{k})}{2} \sqrt{(\omega - \epsilon_c(\mathbf{k}) + \epsilon_v(\mathbf{k}))^2 + 4\hbar^2 \Omega_R^2}, \quad (2.2.3)$$

where

$$\Omega_R = |d_{cv}| \sqrt{\frac{\omega}{2\hbar^2 \epsilon V}} \quad (2.2.4)$$

¹If one expands equation (2.1.13) one finds that this approximation holds as long as $\frac{\hbar^2}{2m_c} \frac{q^2 - 2\mathbf{k} \cdot \mathbf{q}}{\epsilon + \omega - \epsilon_c(\mathbf{k})} \ll 1$. Later in this chapter we will explore what that means physically.

Table 2.1: Vacuum-induced band gaps, $\Delta\varepsilon$, calculated for various semiconductor cavities based on the analytical calculations where we assume that the wave vector of the photonic modes can be neglected. L_z is chosen so that the photon mode is in resonance with a transition away from the band gap. Other dimensions are then assumed to be $L_x = L_y = 5L_z$.

Semiconductor	CdTe	ZnO	GaN	GaAs
L_z (nm)	100	85	88	100
$\Delta\varepsilon$ (meV)	0.36	0.28	0.27	0.38

is the Rabi frequency and

$$\eta(\mathbf{k}) = \begin{cases} -1, & \omega > (\varepsilon_c(\mathbf{k}) - \varepsilon_v(\mathbf{k})) \\ 1, & \omega < (\varepsilon_c(\mathbf{k}) - \varepsilon_v(\mathbf{k})) \end{cases} \quad (2.2.5)$$

makes sure that the correct solution is chosen.

A qualitative graph of this dispersion can be seen in figure 2.3(a). We see that the dispersion of the renormalized valence band electron exhibits a band gap opening where the transition between the valence and conduction band resonates with the cavity photon mode, or where the wave vector satisfies

$$\begin{aligned} \varepsilon_v(\mathbf{k}) - \varepsilon_c(\mathbf{k}) &= \omega \\ k^2 &= \frac{2\mu(\omega - E_g)}{\hbar^2}, \end{aligned} \quad (2.2.6)$$

where $\mu^{-1} = m_c^{-1} + m_v^{-1}$ is the reduced mass. The magnitude of the band gap opening will be twice the Rabi frequency, or

$$\Delta\varepsilon = 2\hbar\Omega_R = |d_{cv}| \sqrt{\frac{2\omega}{\epsilon V}}. \quad (2.2.7)$$

The values of $\Delta\varepsilon$ for various semiconductors can be found in table 2.1. The dimensions of the system are chosen such that $L_x = L_y = 5L_z$ and so that the cavity photon mode is in resonance with a transition between the valence and conduction band for a finite wave vector.

Figure 2.3: (a) Schematic energy spectrum of dressed electrons within the approximation of a negligible small photon wave vector. (b) Exact energy spectrum of dressed valence electrons, calculated numerically for GaAs microcavity with dimensions $L_z = 200$ nm and $L_x = L_y = 1\mu\text{m}$. The components of the electron wave vector $\mathbf{k} = (k_x, k_y, k_z)$ were chosen to satisfy $k_x = k_y$ and $k_z = 0$. The solid blue line shows the full solution of 2.1.13. The multiple gaps arise from the fact that the electron is interacting with 8 different photonic modes which all have the same energy but different wave vectors. This will lead to a gap opening for every value of $\mathbf{k}$ that satisfies equation (2.2.8). The direction chosen here is perpendicular to 4 of them (contributing to the central gap). Two of the photon ground modes give the same positive value when projected on the direction of the chosen $\mathbf{k}$ while the last two give the same value, but negative. These lead to band gaps with slightly higher and lower wave vectors than the central one. We need to be careful though, since the assumption that the electrons are continuous is not strictly correct. The red dots show the allowed values for the electron wavevector. We see that only a few states exist in the gap, making it hard to measure the exact structure.

2.2.2 Numerical calculations

The calculations in the previous section do not, however, show the full picture. Before it was assumed that the wave vector of the ground state of the photonic mode could be neglected since it was small, and therefore all 8 ground state modes could simply be added up. To get a more accurate view of the spectrum we need to solve equation (2.1.13) directly using a numerical approach.

Looking at equation (2.1.13) we see that here the coupling is taking place between multiple modes. Before we assumed they all coincided leading to a coupling between simply $\varepsilon_v(\mathbf{k})$ and $\varepsilon_c(\mathbf{k})$ while in this case the second part is replaced by $\varepsilon_c(\mathbf{k} - \mathbf{q})$, where $\mathbf{q}$ can take all 8 possible values expressed in equation (2.1.8). It is clear that this means that the solution of equation (2.1.13) will depend on the direction of $\mathbf{k}$ relative to $\mathbf{q}$. To find out how, we can solve the conditions analogous to those calculated in equation (2.2.6):

$$\begin{aligned}\varepsilon_v(\mathbf{k}) - \varepsilon_c(\mathbf{k} - \mathbf{q}) &= \omega \\ k^2 - 2\frac{\mu}{m_c}\mathbf{k} \cdot \mathbf{q} &= \frac{2\mu\omega}{\hbar^2} - \frac{\mu}{m_c}q^2.\end{aligned}\tag{2.2.8}$$

Solving for k we get

$$k = \pm k_0 \sqrt{1 - \frac{\mu}{m_c} \frac{q^2}{k_0^2} \left(1 - \frac{\mu}{m_c} \sin^2 \theta\right)} + \frac{\mu}{m_c} q \cos \theta \tag{2.2.9}$$

where $k_0 = \sqrt{\frac{2\mu(\omega - E_g)}{\hbar}}$ and θ angle between k and q . Comparing this to the results of equation (2.2.6) we get from the right hand side a shift of the resonant energy ω by a term proportional to q^2 , which leads only to a small, quantitative correction. The left side does however exhibit a qualitative change. Through the term $\mathbf{k} \cdot \mathbf{q}$ the direction of $\mathbf{k}$ relative to $\mathbf{q}$ can start to matter. Before we assumed that all 8 photonic modes contributed to band gap opening at the same wave vector while here we see that this isn't the case, and find out that the value of the wave vector is shifted by roughly $\Delta k = \frac{\mu}{m_c} q \cos \theta$, where θ is the angle between $\mathbf{k}$ and $\mathbf{q}$. Even though this shift might not be large compared to the value of k we can still end up

with multiple gaps. What happens is the small shift in wave vector leads to a small shift in energy, $\Delta\omega = \varepsilon_v(k \pm \Delta k) - \varepsilon_v(k)$. Since $\Delta k/k \ll 1$ we get that $\Delta\omega \ll \omega$, but that does not mean that it can be neglected. What matters is how $\Delta\omega$ compares to the band splitting $\Delta\varepsilon$.

Looking at the structure of the photon wave vectors $\mathbf{q}$ described by equation (2.1.8) we see that for every one of the different $\mathbf{q}$ s the set also includes $-\mathbf{q}$ so the multiple solutions of equation (2.2.8) will always be symmetric around the solution for the case when $\sin(\theta) = 0$. We also see that if we can take one direction as much smaller than the other, e.g. $L_z \ll L_x = L_y$ we find that four of the vectors $\mathbf{q}$ will lie close to the positive z -axis while the other four will lie close to the negative z -axis, leading to effectively only two vectors where both are fourfold degenerate. We are however very restricted in our choice of L 's since our first assumption in this chapter was that the electrons can be treated as 3 dimensional while the photons are zero dimensional.

For a concrete example let us consider a slab of GaAs in a cavity where the resonant energy of the photon mode is slightly higher than the band gap energy, and the dimensions fulfil $L_x = L_y = 50L_z$. In this case we find that $\Delta\omega$ is larger than $\Delta\varepsilon$ when $\sin\theta$ is large. Figure 2.3(b) shows the band gaps that open at positive k when the electron wave vector lies in the xy plane and fulfils $\mathbf{k} = \frac{1}{\sqrt{2}}(k, k, 0)$. In this case we find that $\sin\theta$ becomes 0 for half of the ground state photon wave vectors while the other half gives $\sin\theta = \pm\frac{\sqrt{2}}{5}$. In this scenario we therefore end up with 3 gaps opening. Note however that on the scale of the gap we cannot ignore the quantization of the electrons. The red dots in figure 2.3(b) show the real dispersion, when discreteness is not ignored. In this case, we see only 5 states inside the gap and would therefore not be able to distinguish the details of the multiple gaps. We however do see a clear band gap opening with some extra states in the center.

2.3 Conclusion

After calculating the renormalized propagator of bulk valence band electrons coupled with 0D vacuum photons in a closed cavity we find that the dispersion exhibits band

gap opening. The size of the band gap opening is of the order of hundreds of μeV , which is larger than both other vacuum effects in the absence of a cavity (*e.g.* the Lamb shift) as well as similar effects that happen in open (two- or one-dimensional cavities [74], and even large enough to be measured. We also find that the band gap has fine structure that is dependent on the direction of the bulk electron, though this structure becomes hard to distinguish because of the discreteness of the electrons.

This effect is interesting since the coupling takes place between electrons and “virtual” photons (quanta of vacuum fluctuations) instead of “real” photons (quanta of electromagnetic wave). This is a way to show that the former is a physical quantity, similar to the Casimir effect where conducting plates feel a real force from virtual photons in an asymmetric system.[64]

The experimental procedure angle-resolved photoemission spectroscopy (ARPS) seems to be the most appropriate. Ultraviolet laser-based ARPS [75, 76] provides sub-meV resolution, which is sufficient to measure the gaps in Table 2.1. Given the fine structure of the gap, and how unclear it gets when taking into account the electron quantization, a very high resolution would be needed to measure this effect.

3. Diagrammatic theory of the optically-induced Aharonov-Bohm effect in quantum rings

Modern day nanotechnologies have advanced immensely in the past decade with the development of high-precision lithography methods in fabrication of nanostructures. Such quantum structures have been a subject of intense research due to a plethora of interesting physical properties naturally arising in them with both fundamental discoveries and important application in modern nanoelectric devices. One such structure is the quantum ring which allowed observation of the famous Aharonov-Bohm (AB) effect [77, 78]: a unique property highlighting the importance of structural topology and the influence of a vector potential on electrons due to an external magnetic field going through the ring structure. Electrons traveling in opposite directions along the ring will accumulate phase difference and cause oscillations in the ring conductance as a function of magnetic flux [79–83]. The AB effect is not exclusive to single particle states as it was shown that it can induce phase difference in many systems consisting of composite particles [84] such as excitons in semiconductor nanostructures [85–89] which had not been experimentally confirmed until the turn of the century [90–92].

Recently it was predicted that electrons can be strongly coupled to a circularly polarized external electromagnetic field resulting in persistent currents in ring shaped quantum wires [93] due to broken time reversal symmetry. Namely, the circular polarization of the incoming light creates a non-equivalent behavior in elec-

trons having clockwise and counterclockwise rotation inside a ring-like structure. This new field-dressed electron dispersion is said to lead to an analogue of the AB effect, resulting in oscillations of the ring current [94] and also to observable splitting between exciton states of mutually opposite angular momenta [95] due to the different effective masses of the coupled electron-hole pair. The phenomenon can be described by introducing a stationary artificial $U(1)$ gauge field generated by the strong coupling between electrons and circularly polarized photons [94].

This chapter is dedicated to the extension of Refs. [94] and [95] by using the nonperturbative diagrammatic approach based on Green's functions. First, a short introduction of the AB effect is given. After that we will introduce the model used to study a quantum ring irradiated by circularly polarized light, based on the Green's functions approach discussed in chapter 1.4. This approach is different then the one used in the previously mentioned references, where the approach was more in line with the more established way of considering the AB effect: using the vector potential to describe the light. That approach resulted in only perturbative results.

3.1 Aharonov-Bohm effect

Even though the AB effect is a purely quantum mechanical effect, to understand its nature it is helpful to review the basics of classical electromagnetism. There, the magnetic ($\mathbf{B}$) and electric ($\mathbf{E}$) fields can be written as

$$\mathbf{B} = \nabla \times \mathbf{A} \qquad \mathbf{E} = -\nabla\phi - \frac{\partial \mathbf{A}}{\partial t}, \qquad (3.1.1)$$

where $\mathbf{A}$ and ϕ are called the vector and scalar potential, respectively. We know that in classical physics we only need to know the magnetic and electric fields in a system to be able to fully understand the electrodynamics of a system, but these potentials are not uniquely defined by these equations. In fact if we know one possible set of $\mathbf{A}$ and ϕ , for every continuously differentiable function Λ we can define a new set of

potentials that result in the same fields:

$$\mathbf{A}' = \mathbf{A} + \nabla\Lambda \qquad \phi' = \phi - \frac{\partial\Lambda}{\partial t}. \qquad (3.1.2)$$

This phenomena is called *gauge transformation*, and when the physics of a system does not depend on the chosen gauge it is called *gauge invariant*.

To see how a quantum system is affected by this, we first need to calculate the canonical momentum of a particle subjected to electric and magnetic fields [96]. First we write down the total force a particle of charge q and velocity $\mathbf{v}$ feels when in the presence of the fields $\mathbf{B}$ and $\mathbf{E}$: $\mathbf{F} = q(\mathbf{E} + \mathbf{v} \times \mathbf{B})$. Then we calculate the Lagrangian of such a system, $L = K - V$ where K is the kinetic energy and V is the potential corresponding to the force, defined as $\mathbf{F} = -\nabla V$. Using equations 3.1.1 and some algebra, we arrive at $L = \frac{1}{2}m\mathbf{v}^2 - q\phi + q\mathbf{v} \cdot \mathbf{A}$. The canonical momentum is then $\mathbf{p} = \frac{\partial L}{\partial \mathbf{v}} = m\mathbf{v} + q\mathbf{A}$. Finally, the Hamiltonian can then be calculated as $H = \mathbf{p} \cdot \mathbf{v} - L = \frac{(\mathbf{p} - q\mathbf{A})^2}{2m} - q\phi$. To quantize the Hamiltonian in real space we can finally write

$$H = \frac{1}{2M} (-i\hbar\nabla - q\mathbf{A})^2 + q\phi. \qquad (3.1.3)$$

However by studying this equation we find out that it is *not* gauge invariant. When we apply the transformation detailed in equations 3.1.2 we find that the phase of the eigenvectors of equation 3.1.3 shifts by $q\Lambda/\hbar$. Naively one might think that for classical and quantum theory to be consistent, they would both need to be gauge invariant, but that is not the case. When reproducing classical calculations in the quantum mechanical approach, it happens that the value of the phase of the wave function doesn't influence the result. However, these results suggest that there might be some purely quantum effects that arise due to this phase shift.

In 1959 Y. Aharonov and D. Bohm published a paper predicting a peculiar effect based on this [77]. They proposed a system consisting of a long solenoid where a beam of electrons flow in the plane perpendicular to the solenoid. The beam is split, so part of it travels on either side of the solenoid and is then recombined. A sketch of the system can be seen in figure 3.1. If the solenoid is considered infinite (or

effectively so, when its length is much larger than other length scales considered) the magnetic field will be negligible outside it, but the vector potential won't be. The vector potential can affect the two paths differently, leading to difference in the phase of the two currents. This phase difference leads to an interference effect.

It can be shown that a particle subjected to a vector potential $\mathbf{A}$ travelling along path $\mathcal{P}$ will acquire a phase $\phi = \frac{q}{\hbar} \int_{\mathcal{P}} \mathbf{A} \cdot d\mathbf{s}$. To calculate the phase difference of two paths with the same beginning and end point, the integral becomes an integral over the closed contour formed by adding the two different paths. Calling this closed contour $\mathcal{O}$ we can write the phase difference as $\Delta\phi = \frac{q}{\hbar} \oint_{\mathcal{O}} \mathbf{A} \cdot d\mathbf{s}$. By using Stokes theorem and the definition of the magnetic flux through the solenoid, Φ_B , we find that the phase difference becomes $\Delta\phi = \frac{q\Phi_B}{\hbar}$, regardless of the paths taken.

The magnetic flux of the solenoid can be tuned by changing the current running through it. The right side of figure 3.1 shows results from Ref. [81], where Webb *et al.* measured the AB oscillations in large metallic rings. We see that the resistance oscillates with changing magnetic flux as predicted.

In a more fundamental sense, we can consider the AB effect as a result of broken time reversal symmetry. It is well known that a magnetic field breaks time reversal symmetry, and its presence in the system breaks the equivalence between clockwise counter-clockwise movement. We can then postulate that if we can break this symmetry by other means we can arrive at a similar result.

3.2 Quantum ring irradiated by circularly polarized light

3.2.1 Model

The system we consider consists of a quantum ring of Radius R illuminated by circularly polarized light with frequency ω_0 . Because of the finite circumference of the ring, any particle confined in the wire will have quantized momentum and therefore a discrete energy spectrum. If we assume a quadratic dispersion, we can

Figure 3.1: Left: The Aharonov-Bohm effect can for example be seen when an electric current is split to go around a solenoid on either side, and then recombined. The solenoid is assumed long enough so that the magnetic field $\mathbf{B}$ can be considered 0 outside of it. The vector potential $\mathbf{A}$ will however not be 0. To be consistent with the magnetic field, The $\hat{\varphi}$ component will be a finite constant, leading to a vector field like shown in green. We can see that an electron travelling along the upper path will be moving parallel with the vector potential, while an electron in the lower path travels antiparallel to it. This leads to a difference in phase when the two electrons reunite, resulting in interference of the two currents. Right: Aharonov-Bohm oscillations measured by Webb *et al.* in 1985 [81]. The inset shows a photograph of the ring used in the experiment. (a) shows how the resistance of the ring oscillates with increasing magnetic flux through the ring. The Fourier transform of the oscillating resistance is plotted in (b), showing a large peak at a frequency of e/h , or the ratio of the electron charge and Planck constant.

Figure 3.2: The system under consideration consists of a quantum ring with a particle that can freely move along it, with a quantized momentum k_m . The ring is subjected to light of circular polarization $\sigma^\pm$.

write it as

$$\varepsilon_m = \frac{\hbar^2 m^2}{2m_e R^2}, \quad m = 0, \pm 1, \pm 2, \dots, \quad (3.2.1)$$

with m_e as the effective mass of the particle.

We can study the effect the light-matter coupling has on an arbitrary particle with the dispersion ε_m travelling in this system by using Green's function formalism. First we write the Green's functions describing the bare propagation of the particle and a photon as

$$G_0(\omega, m) = \frac{1}{\omega - \varepsilon_m}, \quad (3.2.2)$$

$$D_0(\omega) = \frac{2\omega_0}{(\omega - \omega_0)(\omega + \omega_0)},$$

respectively.

The dispersion of the coupled system is determined by the poles of the renormalized Green's functions. Fig. 3.3 describes the renormalization of the particle propagator when accounting for processes that include the absorption of a single

Figure 3.3: The pictorial representation of the renormalization of the Green's function of a particle with momentum m . This renormalization takes into account processes where the particle transitions between the states m and $m + 1$, described by the self-energy Σ_+ , and states m and $m - 1$, described by Σ_- .

photon. When we solve for the renormalized Green's function G we get

$$G(\omega, m) = \frac{G_0(\omega, m)}{1 - G_0(\omega, m)(\Sigma_+(\omega, m) + \Sigma_-(\omega, m))}, \quad (3.2.3)$$

where $\Sigma_{\pm}(\omega, m)$ is the self energy related to the process where the particle transitions between the state m and $m \pm 1$, either by absorbing a $\sigma^{\pm}$ photon or emitting a $\sigma^{\mp}$ photon. Combining equations (3.2.2) and (3.2.3) results in

$$G(\omega, m) = \frac{1}{\omega - \varepsilon_m - \Sigma_+(\omega, m) - \Sigma_-(\omega, m)}. \quad (3.2.4)$$

The dispersion of the coupled system can then be found by solving

$$\omega - \varepsilon_m - \Sigma_+(\omega, m) - \Sigma_-(\omega, m) = 0 \quad (3.2.5)$$

for $\omega(m)$.

To do that we first need to determine the self-energies $\Sigma_{\pm}$. For σ^+ light, the

transition between the m and $m + 1$ state will be energy conserving, while the $m \rightarrow m - 1$ will be energy-non-conserving. For a σ^- photon the situation will be reversed. This means that the self-energies $\Sigma_{\pm}$ will depend on the polarization of the light. These processes are described in figure 3.4. From here on we will consider the case of σ^+ light. In this case we can write the self-energies as

$$\Sigma_+^{(+)} = -ig^2 \int \frac{d\nu}{2\pi} iG_0(m+1, \omega - \nu) iD(\nu) \quad (3.2.6)$$

$$\Sigma_-^{(+)} = -ig^2 \int \frac{d\nu}{2\pi} iG_0(m-1, \nu) iD(\omega - \nu) \quad (3.2.7)$$

where g is the coupling constant between the light and the particle. Inserting equation (3.2.2) and performing the integral we arrive at

$$\Sigma_+^{(+)}(\omega, m) = \frac{g^2}{\omega + \omega_0 - \varepsilon_{m+1}} \quad (3.2.8)$$

$$\Sigma_-^{(+)}(\omega, m) = \frac{g^2}{\omega - \omega_0 - \varepsilon_{m-1}}. \quad (3.2.9)$$

Equation (3.2.5) has then become

$$\omega = \varepsilon_m + \frac{g^2}{\omega + \omega_0 - \varepsilon_{m+1}} + \frac{g^2}{\omega - \omega_0 - \varepsilon_{m-1}}. \quad (3.2.10)$$

This gives a 3rd order polynomial in ω , which is straightforward to solve numerically.

We are interested in examining the asymmetry of the dispersion as a result of the incident light. We do that by studying the energy splitting which we define as

$$\Delta\varepsilon_m = \omega(m) - \omega(-m). \quad (3.2.11)$$

The controllable parameters are the incident light frequency ω_0 , what kind of particle we consider (e.g. electron or exciton) and what material the ring is made of. The bare particle dispersion ε_m depends on the latter two while the coupling constant g will depend on all three.

Figure 3.4: Description of the processes described by the operators Σ_+ and Σ_- . The first describes the absorption of a photon by a particle in level m , leading it to jump to the higher excited state $m + 1$. The particle later decays again to state m . The latter operator describes the oposite process, where a particle in state m decays to a lower energy state $m - 1$, releasing a photon which is later absorbed again by the particle in state $m - 1$, causing it to get excited back to state m .

3.2.2 Results

First, let us consider the case of a small ring, where the energy levels can be clearly distinguished (compared to possible fluctuations, such as thermal energy) and it is meaningful to look at the lowest energy levels. Figure 3.5 shows the resulting splitting for $m = 1, 2, 3$ as a function of the light frequency ω_0 for an undefined particle. The calculations were done for $g = 0.06\varepsilon_1$, where $\varepsilon_1 = \frac{\hbar^2}{2m_e R^2}$ is the energy of the bare $m = \pm 1$ state and m_e is the effective mass of the particle. We see a change in sign in the splitting when ω_0 crosses the resonant energies $\varepsilon_m - \varepsilon_{m-1}$ and again at $\varepsilon_{m+1} - \varepsilon_m$, which correspond to $\omega_0/\varepsilon_1 = 2m \pm 1$. The magnitude of the difference is bound by $|\Delta\varepsilon_m| \lesssim g$.

It is important to remember that even though Green's functions approach can be considered nonperturbative, it is still bound by approximations. It can only be used if the coupling strength is smaller than the energy of the two particles being coupled (here $g^2 < \varepsilon_1 \omega_0$). If this does not hold the amplitude of the Feynman diagrams will grow with increasing order, which means that the diagrams we have chosen to represent the system will not be sufficient.

Now let us consider a different case, where the radius of the ring is large. By large we mean that we can disregard the quantization of the momentum, for the

Figure 3.5: The energy splitting $\Delta\epsilon_m$ plotted as a function of the incident light frequency ω_0 for different values of m . We see that the splitting $\Delta\epsilon_m$ becomes discontinuous at $\omega_0/\epsilon_1 = 2m \pm 1$. These points correspond to the condition when the energy of the incident light is either resonant to the transition $m \leftrightarrow m+1$ or $m \leftrightarrow m-1$. The coupling constant was taken as $g = 0.06\epsilon_1$. Both axes are scaled by the energy of the first excited state, $\epsilon_1 = \frac{\hbar^2}{2mR^2}$.

parameters considered. This can happen when we consider a finite temperature and the thermal energy is comparable or larger than the separation of the levels. In this case we no longer have the discrete quantum number m as before, but instead a continuous wave vector k . All equations stay the same, except we swap out m for kR . Calculations in this approach are only meaningful when $kR \gg 1$, or else the discrete nature of the real system starts to affect the results. We can still consider $\epsilon_1 = \frac{\hbar^2}{2mR^2}$ as a scale in the system, as a characteristic energy of the particles, but given the large radius this energy will be small. The particle energy $\epsilon_1(kR)^2$ and the photon energy ω_0 will be quite large compared to it.

First, let us look at how the energy difference $\Delta\epsilon$ depends on the particle wave vector for a fixed value of the radiation frequency, as shown in figure 3.6. We find that, for a given ω_0 , $\Delta\epsilon$ exhibits a peak where $\omega_0 = 2kR\epsilon_1$. This is unsurprising, since that is where the energy of the light is resonant with the transition between each level. We however find that the energy difference between the two directions can not exceed ϵ_1 . This can be understood by considering the levels that are being

Figure 3.6: The difference in energy between a particle travelling in a large ring with a momentum k and $k - 1$, for 3 different values of the incident light frequency ω_0 . The light is taken to be positively circularly polarized (σ^+). If the light had negative polarization the results would be of the opposite sign. All curves were calculated with $g = 0.1\omega_0$.

coupled. On the left side of 3.7 we see the levels m and $m \pm 1$ in the absence of light, or when the coupling g is zero. Since the dispersion is parabolic, the level m lies closer to the lower level $m - 1$. When considering the coupling between two levels, like in the case of the exciton-polariton considered in §1.2.1, the two levels exhibit an anti-crossing proportional to the coupling strength, like the two levels push each other apart. In this case the level we consider, m , is coupling to both the $m - 1$ and $m + 1$ at the same time. Since the transitions $m \rightarrow m - 1$ and $m \rightarrow m + 1$ will always be close in energy for large rings (since $m = kR \gg 1$), the light can not be close to resonance for one transition while not to the other. This means that once the coupling increases (by increasing the intensity of the light), the state will feel pushed from both sides, ending up right in the middle of the other two states once the coupling reaches roughly $g \gtrsim 2\varepsilon_1$.¹ At this point, no matter how strong the coupling to the light becomes, the shift can never become greater. The only thing that happens is that the peaks in figure 3.6 will broaden.

¹Note that here the energy of the particles is $\varepsilon_1(kR)^2 \gg \varepsilon_1$, so we do not need to worry about breaking the validity of the approach until $g \approx \varepsilon_1(kR)^2$.

Figure 3.7: To the left we see a schema of an arbitrary energy level m along with the neighbouring states $m \pm 1$, in the absence of coupling to light. In this setup the energy of the transitions will be $(2m \pm 1)\varepsilon_l$. When the coupling to the light is turned on, the state m will shift to a higher energy. The shift will saturate when the coupling constant reaches $G \gtrsim 2\varepsilon_l$, when the state sits equidistant from the two other levels. At that point, the coupling of the two transitions will be equal.

Like noted above, the system of a ring irradiated by a circularly polarized light has been studied before using perturbation theory. Figure 3.8 shows a comparison of the result arrived at by Kibis *et al* [95] and the result of equation 3.2.10 for the same parameters, for calculating the energy difference of the first two excited states ($m = \pm 1$). The radius is set to $R = 9.6$ nm. The perturbative result is plotted in dashed green while the diagrammatic result is in solid blue. As expected, the perturbative approach diverges at the resonant frequency while the diagrammatic one does not. The two approaches are however in good agreement while further away from the resonance.

The same authors also considered the case of excitons in a large ring ($R = 10\mu\text{m}$) in reference [94]. There they predict AB oscillations by tuning either the incident light frequency or its intensity (effectively tuning the strength of the coupling). As mentioned above, the current approach predicts that the shift of energy will not exceed ε_l , which in the case of a large ring will lead to a phase difference of $\Delta\phi \propto \frac{\varepsilon_l}{\varepsilon(k)} \ll 1$ by definition. For the system to exhibit oscillations the phase difference needs to be at least a few multiples of π .

Figure 3.8: Comparison between equation [94] (in dark, solid blue) and the equation from Ref [95] (in dashed, light green). The former was arrived at using the Green's functions approach while the later a perturbative approach for $m = 1$. As expected, the two approaches agree while far away from resonance, while the perturbative approach is inaccurate close to the resonance.

3.3 Conclusion

In this chapter we calculated the energy spectrum of a particle in a quantum ring subjected to circularly polarized light. The circular polarization of the light breaks time reversal symmetry and by extension breaking the symmetry of the dispersion relation. In the case of small rings we found that the energy difference of the clockwise and the counter-clockwise states ($m > 0$ and $m < 0$) changes sign abruptly when crossing the resonant frequencies.

In the case of large rings we find that this effect gets saturated and will not lead to Aharonov-Bohm oscillations, since the maximum phase difference can not exceed unity.

4. An array of quantum wires in a planar cavity

4.1 Bose-Einstein condensate

The most distinct aspects of bosons is the fact that there is no limit on how many particles you can have in a single state, opposite to fermions where the Pauli exclusion principle holds. At sufficiently low temperatures, this leads to peculiar behaviour called Bose-Einstein condensation (BEC), where a macroscopic fraction of particles can accumulate in the ground state[30, 45]. The temperature at which this happens is called the critical temperature T_C and will depend on the system in question. The Bose-Einstein condensate can be described by the order parameter Ψ_0 which is simply the mean value of the condensate wave function ψ :

$$\Psi_0(\mathbf{r}) = \langle \psi(\mathbf{r}) \rangle = \sqrt{n(\mathbf{r})} e^{i\theta(\mathbf{r})}, \quad (4.1.1)$$

where $n(\mathbf{r})$ is the density of the condensate and $\theta(\mathbf{r})$ the phase. A true condensate however experiences phase coherence, meaning that θ will be a constant in real space. At finite temperatures there will always be some fluctuations from this ideal equilibrium state.

4.1.1 Gross-Pitaevskii equation

We go out to seek for an equation that describes the behaviour of this superfluid condensate. We want to be able to introduce an external potential as well as see the

effects of internal interactions. We also want to be able to study the dynamics on time scales larger than the lifetime. To do that we need to have an external pump and take into account the dissipation in the system. To describe this mathematically, we start by writing the Hamiltonian in a second quantization formalism, with terms corresponding to the kinetic energy, potential energy, boson-boson interactions and external pump, respectively:

$$\hat{H} = \sum_k E_k \hat{\psi}_k^\dagger \hat{\psi}_k + \sum_x V_x \hat{\psi}_x^\dagger \hat{\psi}_x + \frac{\alpha}{2} \sum_x \hat{\psi}_x^\dagger \hat{\psi}_x^\dagger \hat{\psi}_x \hat{\psi}_x + \sum_k \left(\hat{\psi}_k^\dagger + \hat{\psi}_k \right) F_k e^{-i\omega t}, \quad (4.1.2)$$

where E_k is the energy of a state with a wave vector k , V_x is the value of the external potential at point x , α is the interaction strength between two bosons, F_k is the profile of the external pump in k -space and ω the pump frequency. The field $\hat{\psi}_x$ is the real space equivalent of $\hat{\psi}_k$, given by a discrete Fourier transform

$$\hat{\psi}_k = \frac{1}{\sqrt{N}} \sum_x \hat{\psi}_x e^{-ikx}, \quad \hat{\psi}_x = \frac{1}{\sqrt{N}} \sum_k \hat{\psi}_k e^{ikx}, \quad (4.1.3)$$

where N is the number of states being summed over.

Since we want to be able to describe dissipation in our system it seems natural to make use of the density matrix formalism, where dissipation can be introduced as a Lindblad term in a generalized Liouville-Von Neumann equation,

$$i\hbar \frac{\partial \rho}{\partial t} = [\hat{H}, \rho] + \frac{i\Gamma}{2} \sum_k \left(2\hat{\psi}_k \rho \hat{\psi}_k^\dagger - \hat{\psi}_k^\dagger \hat{\psi}_k \rho - \rho \hat{\psi}_k^\dagger \hat{\psi}_k \right), \quad (4.1.4)$$

where ρ is the density matrix and Γ is the dissipation rate. This equation, often called the Lindblad master equation, comes from the theory of open quantum systems and is regularly used in the field of quantum optics [97].

Now, the purpose of all this was to be able to describe the dynamics of the order parameter we mentioned in the previous chapter. Remember that the order parameter is the expectation value of the field operator (or the mean-field), and in the density matrix formalism an expectation value is the trace of the operator with

the density matrix:

$$\Psi = \langle \psi \rangle = \text{tr} \left\{ \rho \hat{\psi} \right\}. \quad (4.1.5)$$

We will start by writing out the equation of motion for the mean-field in reciprocal space.

$$i\hbar \frac{\partial \langle \psi_k \rangle}{\partial t} = \left\langle \left[\hat{H}, \hat{\psi}_k \right] \right\rangle + \frac{i\Gamma}{2} \text{tr} \sum_{k'} \left\{ \left(2\hat{\psi}_{k'} \rho \hat{\psi}_{k'}^\dagger - \hat{\psi}_{k'}^\dagger \hat{\psi}_{k'} \rho - \rho \hat{\psi}_{k'}^\dagger \hat{\psi}_{k'} \right) \psi_k \right\}. \quad (4.1.6)$$

After a bit of algebra and defining $V_k = \frac{1}{\sqrt{N}} \sum_x V_x e^{-ikx}$ as the Fourier transform of the potential, we arrive at

$$i\hbar \frac{\partial \langle \psi_k \rangle}{\partial t} = \left(E_k - \frac{i\Gamma}{2} \right) \langle \psi_k \rangle + \frac{1}{\sqrt{N}} \sum_{k'} V_{k'} \langle \psi_{k-k'} \rangle + \frac{\alpha}{N} \sum_{k_1, k_2} \left\langle \hat{\psi}_{k_1}^\dagger \hat{\psi}_{k_2} \hat{\psi}_{k_1+k_2-k} \right\rangle + F_k e^{-i\omega t}. \quad (4.1.7)$$

The same equation in real space will take the form

$$i\hbar \frac{\partial \langle \psi_x \rangle}{\partial t} = \frac{1}{\sqrt{N}} \sum_{x'} E_{x'} \langle \psi_{x-x'} \rangle + \left(V_x - \frac{i\Gamma}{2} \right) \langle \psi_x \rangle + \alpha \langle \psi_x^\dagger \psi_x \psi_x \rangle + F_x e^{-i\omega t}. \quad (4.1.8)$$

where $F_x = \frac{1}{\sqrt{N}} \sum_k F_k e^{ikx}$ is the real space pump profile, gotten by taking the inverse Fourier transform of the k -space profile. When the density of the condensate is high, $\langle \psi^\dagger \psi \rangle \gg 1$ we can make the approximation that $\langle \psi_x^\dagger \psi_x \psi_x \rangle \approx \langle \psi_x^\dagger \psi_x \rangle \langle \psi_x \rangle$. We can then generalize this to a continuous function as

$$i\hbar \frac{\partial \Psi(x)}{\partial t} = \left(\hat{E} + V(x) + \alpha |\Psi(x)|^2 - \frac{i\Gamma}{2} \right) \Psi(x) + F(x) e^{-i\omega t}, \quad (4.1.9)$$

where $\hat{E}$ is the energy operator, and for the normal parabolic dispersion $\hat{E} = -\frac{\hbar^2 \nabla^2}{2m}$. Equation (4.1.9) is called the *Gross-Pitaevskii equation* after the physicists Evgenii P. Gross [98] and Lev P. Pitaevskii [99] who derived similar equations simultaneously in 1961.

4.1.2 Polariton BEC

The discussion above holds for bosons in general, but we are especially interested in studying exciton-polaritons and using the GP equation to calculate the dynamics of such a condensate. In the simplest cases of BEC, one studies bosons (e.g. cold atoms) which are cooled below the critical temperature and the bosons condense, reaching an equilibrium state with macroscopic occupation of the ground state. When the bosons in question are polaritons this becomes a bit trickier. Since polaritons have a short lifetime we rely on a constant external pump to keep a polariton population in our system. This means that we can not talk about the state being in equilibrium, since we will have constant flux of new particles in and out of the system. We can however talk about a steady-state, where the external pump and the dissipation are equal. In this case, it has been well established that it is possible to reach a coherent, macroscopic occupation of the ground state in exciton-polariton systems[3, 4, 100]. Inside the community of exciton-polariton physics, these states are consistently called polariton BEC or polariton condensate, even though some outside of the field consider that only states in equilibrium can be deemed such. Regardless of the specific definitions of a BEC however, we know that exciton-polaritons are bosons that can form a coherent state where we can use the mean-field approximation to define an order parameter like in the derivation of the GP equation above.

Apart from studying the spontaneous condensation of polaritons to the ground state, direct coherent excitation of polaritons with a resonant laser can also be considered. This will directly create a coherent state, which can also be described by a the driven-dissipative GP equation above.

Figure 4.1: The system considered consists of a periodic array of nanowires embedded into a planar microcavity consisting of two DBRs separated by a distance L_c . The wire separation is denoted by d .

4.2 Hyperbolic region in an array of quantum wires in a planar cavity

4.2.1 Model

In this chapter we will consider a system that consists of a periodic array of N semiconducting nanowires embedded into a planar microcavity, as shown in Fig. 4.1. The wires lie along the x direction and the separation along y axis is d . The cavity plane is perpendicular to the z -axis and the confinement length is L_c . The electrons are confined to one dimension (1D) while the photons can move in two dimensions (2D). We can assume that d is large enough so we can ignore the possibility of tunnelling between the wires. We also assume a parameter range where excitons are stable, i.e. low temperature and density.

The Hamiltonian of the system can be written as

$$H = H_{ph} + H_X + H_{int} \quad (4.2.1)$$

where the three terms correspond to the energy of the photons, excitons and the

interaction between the two, respectively. The photon Hamiltonian is

$$H_{ph} = \sum_{\mathbf{k}} \varepsilon_{ph}(\mathbf{k}) a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}}, \quad (4.2.2)$$

where ε_{ph} is the photon energy dispersion and $a_{\mathbf{k}}^{(\dagger)}$ is the photon annihilation (creation) operator. $\mathbf{k} = (k_x, k_y)$ is a 2D wave vector in the plane of the cavity. The photon energy, corresponding to the fundamental cavity mode, is

$$\varepsilon_{ph}(\mathbf{k}) = \frac{\hbar c}{\sqrt{\epsilon_c}} \sqrt{\left(\frac{\pi}{L_c}\right)^2 + \mathbf{k}^2}. \quad (4.2.3)$$

where ϵ_c is the relative permittivity of the cavity.

If exciton-exciton interactions are neglected, the excitonic term of the Hamiltonian will be

$$H_X = \sum_{n, k_x} \varepsilon_X(k_x) b_{k_x, n}^{\dagger} b_{k_x, n}, \quad (4.2.4)$$

where $b_{k_x, n}^{(\dagger)}$ is the annihilation (creation) operator for an exciton in wire n and ε_X is the exciton energy, given by

$$\varepsilon_X(k_x) = E_g - E_B + \frac{\hbar^2 k_x^2}{2M}. \quad (4.2.5)$$

E_g denoting the band gap energy of the semiconductor, E_B the exciton binding energy and M the exciton mass, which is simply the sum of the electron and hole effective masses.

We can include exciton-exciton interactions by including the term

$$H_X^{int} = \sum_{n, k_x, k'_x, q} U_q b_{k_x+q, n}^{\dagger} b_{k'_x-q, n}^{\dagger} b_{k_x, n} b_{k'_x, n}, \quad (4.2.6)$$

where U_q is the Fourier transform of the exciton-exciton pair repulsion potential.

Finally, the last term of equation (4.2.1) is

$$H_{int} = \sum_{n,\mathbf{k}} \Omega_n(\mathbf{k}) a_{\mathbf{k}}^\dagger b_{k_x,n} + h.c., \quad (4.2.7)$$

where Ω_n is the coupling energy between the light and the excitons and $h.c.$ is the Hermitian conjugate of the previous term. For now we need not worry about the full form of the coupling constant and will only consider the n dependence. As discussed in §1.2, the coupling constant is the product of the electric field in the cavity and the dipole moment matrix element of the transition:

$$\Omega_n = \mathcal{E} d_X. \quad (4.2.8)$$

Since the excitons are confined to 1D wires along the x axis, the dipole moment matrix element d_X is oriented along that axis. This means that the coupling only takes place between the excitons and x -polarized light and coupling to the y -polarization can be ignored.

The spatial dependence of d_X is simply that it is confined to the semiconductor nanowires, so $d_x \propto \sum_n \delta_{y,nd}$. The electric field of the cavity photons can be considered as a plane wave in the xy plane, so $\mathcal{E} \propto e^{iyk_y}$. Combining these two observations we see that we can write

$$\Omega_n = e^{indk_y} \Omega_0(\mathbf{k}). \quad (4.2.9)$$

In order to get rid of the implicit dependence on the variable n , we can introduce a Fourier transform of the exciton operators as

$$b_{\mathbf{k}} = \frac{1}{\sqrt{N}} \sum_n e^{indk_y} b_{k_x,n}, \quad b_{\mathbf{k}}^\dagger = \frac{1}{\sqrt{N}} \sum_n e^{-indk_y} b_{k_x,n}^\dagger, \quad (4.2.10)$$

where k_y is a quasi-momentum arising from the periodicity of the system. since equation (4.2.10) is invariant under the transformation $k_y \rightarrow k_y + \frac{2\pi}{d}$ it is natural to constrict our consideration to the first Brillouin zone (BZ), $-\frac{\pi}{d} < k_y < \frac{\pi}{d}$. For consistency we can fold the cavity photon dispersion ε_{ph} into this first BZ, in an

analogous way to the approach normally taken when considering electrons in crystal lattices. We then define the new photon dispersion as

$$\varepsilon_{ph}^{(\lambda)}(\mathbf{k}) = \begin{cases} \frac{\hbar c}{\sqrt{\epsilon_c}} \sqrt{\left(\frac{\pi}{Lc}\right)^2 + k_x^2 + (|k_y| - \lambda \frac{\pi}{d})^2} & \text{if } \lambda \text{ is even} \\ \frac{\hbar c}{\sqrt{\epsilon_c}} \sqrt{\left(\frac{\pi}{Lc}\right)^2 + k_x^2 + (|k_y| + (\lambda - 1) \frac{\pi}{d})^2} & \text{if } \lambda \text{ is odd,} \end{cases} \quad (4.2.11)$$

with $\lambda = 1, 2, \dots$ denoting the band number. The photon Hamiltonian described in equation (4.2.2) is then equivalent to

$$H_{ph} = \sum_{\mathbf{k}, \lambda} \varepsilon_{ph}^{(\lambda)}(\mathbf{k}) a_{\mathbf{k}}^{\lambda\dagger} a_{\mathbf{k}}^{\lambda}, \quad (4.2.12)$$

where the sum over k_y now only goes over $-\frac{\pi}{d} < k_y < \frac{\pi}{d}$, and the creation (annihilation) operators now create (annihilate) a photon in a specific band λ .

Combining equations (4.2.4) and (4.2.10) we find that the exciton Hamiltonian is simply

$$H_X = \sum_{\mathbf{k}} \varepsilon_X(k_x) b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}}. \quad (4.2.13)$$

The interaction term can also be rewritten to be confined to the first BZ. Combining equations (4.2.7) and (4.2.10) we arrive at

$$H_{int} = \sum_{\mathbf{k}} \Omega_R(\mathbf{k}) a_{\mathbf{k}}^{\lambda\dagger} b_{\mathbf{k}} + h.c. \quad (4.2.14)$$

where the Rabi frequency has been normalized to $\Omega_R = \sqrt{N} \Omega_0$, since Ω_0 corresponded to the coupling of a single wire to a photon spread over the whole cavity.

Now we can combine equations (4.2.12), (4.2.13) and (4.2.14) to arrive to the complete Hamiltonian

$$H = \sum_{\mathbf{k}} \left[\sum_{\lambda} \varepsilon_{ph}^{(\lambda)}(\mathbf{k}) a_{\mathbf{k}}^{\lambda\dagger} a_{\mathbf{k}}^{\lambda} + \varepsilon_X(k_x) b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} + \Omega_R \sum_{\lambda} \left(a_{\mathbf{k}}^{\lambda\dagger} b_{\mathbf{k}} + a_{\mathbf{k}}^{\lambda} b_{\mathbf{k}}^{\dagger} \right) \right], \quad (4.2.15)$$

where, as before, the sum over k_y is confined to the first BZ, $-\frac{\pi}{d} < k_y < \frac{\pi}{d}$ while the

sum over k_x is not restricted.

To better visualize the meaning of the results we can define the Hamilton density $\mathcal{H}_{\mathbf{k}}$, where $H = \sum_{\mathbf{k}} \mathcal{H}_{\mathbf{k}}$, and consider its matrix form in the basis {Exciton, Photon in band 1, Photon in band 2, ...}:

$$\mathcal{H}_{\mathbf{k}} = \begin{bmatrix} \varepsilon_X & \Omega_R & \Omega_R & \cdots \\ \Omega_R & \varepsilon_{ph}^{(1)} & 0 & \\ \Omega_R & 0 & \varepsilon_{ph}^{(2)} & \\ \vdots & & & \ddots \end{bmatrix}, \quad (4.2.16)$$

where the $\mathbf{k}$ dependence of each term has been suppressed for clarity. It is clear that this is simply a system of infinite number of modes where the first mode is coupled to all the others. We can tune the cavity parameters in a way to choose what the energies of the cavity photon modes will be relative to the exciton energy. The photon modes with resonant energy to the exciton mode will experience anti-crossing behaviour with a band gap of $\sim \Omega_R$, while photon modes that lie far away from the exciton mode will hardly be affected.

To estimate the Rabi splitting we start by remembering that $\Omega_R = \sqrt{N}\Omega_0 = \sqrt{N}|d_X||\mathcal{E}|$. The electric field is given by equation 1.2.2, so

$$|\mathcal{E}| = \sqrt{\frac{\varepsilon_{ph}}{2\varepsilon_c\varepsilon_0 V}}. \quad (4.2.17)$$

the cavity volume is here and $V = NdLL_c$. Next we need to calculate the dipole matrix element. Making use of equation (1.2.3) and writing the position vector as $\hat{\mathbf{r}} = (\hat{x}_1, \hat{x}_2, \hat{x}_3)$, we can write [101]

$$\begin{aligned} |d_X|^2 &= \sum_{i=1}^3 |\langle f | e\hat{x}_i | i \rangle|^2 \\ &= 2L |\psi_0(x=0)|^2 |\langle u_v | e\hat{x} | u_c \rangle|^2 \end{aligned} \quad (4.2.18)$$

where $|f\rangle$ and $|i\rangle$ are the final and initial states of the crystal, referring to the exciton and ground state of the wire. ψ_0 is the exciton envelope function for the ground state, e is the electron charge and $|u_{v(c)}\rangle$ is the Bloch wave function for the valence (conduction) band.

To calculate ψ_0 we need to solve a problem analogous to the 1D hydrogen atom, where the hole takes the role of the proton. To solve this problem we will use the Loudon model, which consists of shifting the Coulomb potential by a distance δ to get rid of some unphysical peculiarities of the problem [102].

Looking for solutions approximating the form

$$\psi_0(x) = \sqrt{\alpha} e^{-\alpha|x|} \quad (4.2.19)$$

we find that the parameter α needs to obey the relation [103]

$$\frac{\alpha}{2} e^{\alpha/2} = \frac{1}{4\delta}. \quad (4.2.20)$$

We can estimate the parameter δ as the diameter of the 1D wires considered.

In our numerical calculations we use parameters corresponding to ZnO wires. Solving equation (4.2.20) for diameters of about 100 nm [104] we find that $\alpha \approx 2/a_B^{3D}$, where $a_B^{3D} = \frac{4\pi\hbar^2\epsilon_w\epsilon_0}{\mu e^2}$ is the 3D exciton Bohr radius, μ the reduced mass of the exciton and ϵ_w the relative permittivity of the wire. From equation (4.2.19) we see that the exciton radius is simply $a_B^{1D} = 1/\alpha = a_B^{3D}/2$.

Second, we see that $|\langle u_v|\hat{x}|u_c\rangle|$ is simply the distance between the electron and hole: a_B^{1D} which we have already calculated.

After substituting these terms, equation (4.2.18) becomes

$$|d_X|^2 = e^2 a_B^{3D} L. \quad (4.2.21)$$

Lastly, we combine equations (4.2.17) and (4.2.21) and end up with

$$\Omega_R = \sqrt{\frac{e^2 \mathcal{E}_{ph} a_B^{3D}}{2\epsilon_c \epsilon_0 d L_c}}. \quad (4.2.22)$$

4.2.2 Calculations and results

To diagonalize the Hamiltonian we need to restrict our calculations to a finite number of photon bands. It is easy to check if the cut-off chosen affects the results by checking if the resulting dispersion has converged by comparing results with varying number of bands. We find that limiting the consideration to a few photon bands close in energy to the exciton mode is sufficient. In our calculations we only consider 4 photon bands, along side the exciton band.

The resulting dispersion found by diagonalizing the Hamiltonian in equation (4.2.15) for $k_x = 0$ and standard parameters for ZnO can be seen in blue in figure 4.2. The cavity parameters have been tuned so that the energy of the exciton mode falls (a) in between the first and second photon mode, (b) inside the second photon mode and (c) between the second and third photon modes. The uncoupled modes have been plotted in gray for comparison. The flat band we can see in figure 4.2(a) is typical in polariton systems with engineered periodic potential. It can be used for realizing incompressible polariton states [105] as well as Landau-Zener Bloch Oscillations [106]. Figure 4.3 shows a zoom in on this band. The shaded region depicts the linewidth of an polariton with lifetime 20 ps. Since the variations of the band are smaller than the linewidth, the band is effectively flat.

We find that regardless of the choice of where the exciton mode coincides with the photon modes, the dispersion will exhibit a hyperbolic region, where the effective masses along the two directions x and y have opposite signs.

An example of how this dispersion feature can be utilized is demonstrated in figure 4.5. There it is shown what happens when the system is optically pumped with a positive wave vector in the y direction k_y and frequency ω_0 . We find that if the frequency is chosen to resonate with the second branch of the dispersion (marked by a darker, thicker line in (a)) the wave packet travels in the negative y direction. If the pump however resonates with the fourth band (seen in (b)) the wave packet travels in the positive direction. We can therefore route a polariton wave packet by controlling the pump energy.

As mentioned above, the excitons in the system can interact with each other and

Figure 4.2: Dispersion relation of the system for $k_x = 0$ and three different values of the confinement length of the cavity L_c . The confinement length has been chosen so that the exciton energy falls (a) in between the first and second photon modes, (b) inside the second band, and (c) between the second and third bands. The dispersion, found from equation (4.2.15), is shown in blue while the thinner, gray lines are the uncoupled modes. The k_x dependence of the second branch in (c) can be seen in figure 4.4. All the graphs are calculated for standard parameters corresponding to ZnO wires separated by $d = 1\mu\text{m}$.

Figure 4.3: A zoom in of the flat band in figure 4.2(a). Naturally, the band is not exactly flat at this scale. However, when compared to the linewidth of a realistic polariton (shaded region, comparable to a polariton lifetime of 20 ps) we see that the variations are small enough to be neglected.

Figure 4.4: Detailed view of the second branch of the dispersion relation shown in figure 4.2(c). It is clear that the effective mass and group velocity along the y axis is negative around the point $(0,0)$, while both are positive along the x direction.

Figure 4.5: When the parameters of the system are chosen so that the exciton energy lies between the second and third photon bands, the resulting dispersion exhibits not just a negative mass along y direction, but also negative group velocity. (a) shows the dispersion for $k_x = 0$ with the second branch shown thicker and darker than the others. (b) shows the time evolution of the system when the second polariton branch is pumped with a pulse of a positive $k_y = \frac{\pi}{30d}$. We see that the pulse propagates along the negative y axis. In the case where the pump is resonant with the fourth branch, shown in (c), but with the same k_y value as before, the propagation will be along the positive y axis. Both cases are calculated with standard parameters of ZnO wires and $d = 1\mu\text{m}$.

for higher concentrations this becomes significant. Since polaritons are composites of light and excitons we can expect them to exhibit non-linear effects when the exciton concentration is high enough. Equation (4.2.15) neglected the exciton-exciton interaction term but we can calculate the effect it will have from our current results. Since the interaction term will make the Hamiltonian non-linear, the Hamiltonian can no longer be represented with a matrix to be diagonalized like before. We can however always diagonalize the non-interacting part (like we did before) and then make sure to express the interaction term in the relevant basis (the polariton operators, instead of the exciton operators) [107]. We can focus on studying the effect that the exciton-exciton interaction has on a specific polariton branch (SPB) by writing the Hamiltonian as

$$H_{SPB} = \sum_{\mathbf{k}} \varepsilon_{SPB} p_{\mathbf{k}}^{\dagger} p_{\mathbf{k}} + H_X^{int}, \quad (4.2.23)$$

where ε_{SPB} is the eigenvalue of equation (4.2.15) corresponding to the branch in question and $p^{(\dagger)}$ is the corresponding polariton annihilation (creation) operator. The term H_X^{int} was defined in equation (4.2.6) but we need to rewrite it in terms of the polariton operators p instead of the exciton operators b . To do that we define the effective coupling constant

$$g_{\mathbf{k}, \mathbf{k}', q_x} = U_{q_x} X_{\mathbf{k}+q_x} X_{\mathbf{k}'-q_x} X_{\mathbf{k}} X_{\mathbf{k}'}, \quad (4.2.24)$$

where X_k is the exciton fraction in the SPB at a given $\mathbf{k}$. Now we can write

$$H_{Xint} = \sum_{\mathbf{k}, \mathbf{k}', q_x} g_{\mathbf{k}, \mathbf{k}', q_x} p_{\mathbf{k}+q_x}^{\dagger} p_{\mathbf{k}'-q_x}^{\dagger} p_{\mathbf{k}} p_{\mathbf{k}'}. \quad (4.2.25)$$

The resulting dispersion when H_{SPB} is diagonalized is not qualitatively different than the previous result, it is merely blueshifted by approximately $g\langle p^{\dagger} p \rangle$. The non-linearity affects the behaviour in another way, however. We can study the dynamics of the system using mean field theory and the Gross-Pitaevskii equation discussed in section §4.1.1.

Figure 4.6: When an initial condition ψ_0 of homogeneous density and a phase kink at $x = 0$ as shown in the bottom plot (phase along y is constant) is made to evolve we see an emergence of a soliton-like behaviour. The calculation is done using Gross-Pitaevskii equation, with ZnO wires separated by $d = 1\mu\text{m}$, and the effective polariton-polariton coupling set to be a constant $g = 0.006 \text{ meV } \mu\text{m}^2$.

The non-linearity of the Hamiltonian along with the peculiar nature of the dispersion lead to interesting stable solutions. For example we have found that the system sustains dark soliton-like solutions similar to those seen by Walker et al in the lower polariton branch of slab polariton waveguides [108]. Figure 4.6 shows the time evolution of $|\psi|^2$ in a cross-section along the x -axis, when the initial condition ψ_0 is a uniform field with a discontinuity in the phase, as shown at the bottom of the figure. While the distribution we observe has clear characteristics of a dark soliton pattern (along with a phase jump), it is not a sufficient criteria for identifying solitons [109]. Nevertheless, systems which can sustain the propagation of self-preserving states is highly relevant for signal propagation applications, where other nonsolitonic solutions have been proposed such as the X-wave solution [110].

4.2.3 Conclusion

In this chapter we have shown many interesting properties a system of 1D excitons in an array of quantum wires, coupled with a 2D photon mode, exhibits. We show how the discrete, periodic nature in one direction leads to the formation of a band structure along that direction. The asymmetry between the directions leads to a hyperbolic region, where the masses along the two axes, along and parallel to the wires, have different signs. To illustrate the implications of the peculiar dispersion we first showed results based on simulation with the Gross-Pitaevskii equation where a polariton pulse will propagate in opposite directions depending on the exact energy of the external pump creating it. We also showed that the system can sustain dark-soliton like patterns.

Strictly speaking, it has been shown that the simultaneous localization of photon and exciton components should not be possible in real space due to different photon and exciton dispersions [111]. Polariton solitons propagate over a finite distance, before dephasing of the Rabi oscillations between exciton and photon components take place. We have not considered the effects of spatially varying dissipation; this was recently predicted to have rise to spontaneous currents in periodic polariton lattices [112]. Another system we could compare to consists of a 1D BEC where the dispersion has been modified with Raman dressing to give rise to a region with negative mass [113]. It was shown that the system exhibits shock waves, soliton trains and self-trapping. We would expect that by pumping our system along the y -direction we could get similar effects, though these effects have all been achieved in polariton systems before [114–116].

5. Terahertz cascade from a nanoparticle

5.1 Terahertz radiation

Terahertz (THz) radiation is loosely defined as electromagnetic radiation of frequencies falling into the range of 0.3 THz to 10 THz [117], which corresponds to wavelengths of 30 μm to 1 mm and falls into the far infrared bordering on the microwave range. Historically there has not been much interest in this range, partly due to the high absorption of THz radiation in the atmosphere and partly because the commonly employed techniques used for the study of neighbouring bands do not work in the THz range. In the last decades the interest has however been rapidly growing. Scientists have found many uses for THz radiation in varying fields, such as in astrophysics [118] and biology [119]. There have also been an increase in more practical uses, such as cancer diagnosis [120] and security imaging [121]. Despite the advances that have been made in the development of THz devices, there is still a lack of flexible, universal and efficient THz emitters.

The current reigning champion of THz emission is the *quantum cascade laser* (QCL). The concept of the QCL was first theoretically proposed in 1971 [122] but was only experimentally achieved in 1994 [123]. The first QCL emitted light in the mid-infrared range, but the nature of the device meant that its structure could be tailored to emit larger wavelengths, even in the THz range. Initially QCLs could only operate at low temperatures and were difficult to construct, but over the last

two decades there has been great progress in regards to both of those aspects [124].

One of the greatest strengths of the QCL is its tunability, which stems from the fact that the optical transitions at play are intersubband transitions (that is, transition between quantized states in the same energy band) instead of relying on a band gap of a material. QCLs consists of multiple semiconductor quantum well (QW) heterostructures subjected to an external electric field that tilts the energy bands. In simplified terms, the electric field is tuned so that the ground state of one QW aligns with the first excited state of the next one. Figure 5.1 shows a schematic of the conduction band of such a structure. When the excited state of the first quantum well is pumped, it leads to an optical transition to the ground state, emitting a photon of a frequency set by the dimension of the QW. The electron can then tunnel from the ground state of the first QW to the excited state of the second, repeating the same process again. This cascade of electrons is where the QCL gets its name from, and means that we get multiple photons for each excitation. The frequency of the emitted photon is highly tuneable since it depends on the dimensions of the system. The QCL has mainly been used in the infrared range, but has shown great promise for use in the upper THz range [124].

Another way of generating THz radiation is to excite photoconductive emitters using a femtosecond laser [125]. This method converts optical light to low energy THz radiation by firing an ultra short pulse at a photoconductive medium which leads to the generation of oscillations of charge density. This principle can be used in a variety of mediums to achieve radiation in the THz range, such as semiconductor-metal interfaces [126], nanojunctions [127] and graphite flakes [128]. Yet other ways exist. Gunn diodes are commonly used for emission in the microwave range, but have been shown to work in the lower THz range as well [129]. Intense THz radiation has been reported from the Novosibirsk free electron laser [130], but since it principally depends on the oscillation of strong magnetic fields it will always remain bulky. Out of all these devices, none fully fulfil all the requirements set by the possible applications. None manage to span the whole THz spectrum while being efficient and operate at room temperature in a compact form. Due to that, there is still an ongoing search for new THz sources and detectors.

Figure 5.1: A simplified diagram showing the energy levels of the quantum cascade laser. The QCL is made up from a row of quantum wells along the z -axis. The structure is then subjected to an electric field so that the first excited state is resonant with the ground state of the next well. Particles can then tunnel from one the ground state of one QW to the excited state of the next QW, and there relax into the ground state, releasing a photon.

In this work we aim to merge the concept of THz cascades based on fermionic transitions in semiconductor heterostructures with the concept of using artificial atoms. Instead of using excitons as artificial atoms, as was proposed by Liew *et al.* [131, 132], we consider semiconductor nanoparticles, which lie among the most compact of man-made systems. It has been suggested that the alloy composition of such nanoparticles can be varied spatially [133, 134]. We show that in principle this leads to the possibility of engineering multiple transitions with the same THz range frequency in a single nanoparticle. To obtain a suitable alloy composition profile, we introduce a numerical optimization algorithm, inspired by learning algorithms for neural networks. We calculate the optimized THz transition rates for a typical GaAlAs based system. Considering a collection of nanoparticles in a compact cavity, quantum efficiencies greater than unity are readily obtained.

5.2 Nanoparticles with hydrogen impurity

5.2.1 Model

Let us start by considering a spherical nanoparticle with a hydrogenic impurity (ion with charge $+e$ in the center) with radius R , shown in figure 5.2(a). We take the particle to be spherically symmetric but let the material composition vary along the radial direction r . This can physically be achieved by varying the alloy composition in materials such as $\text{Al}_x\text{Ga}_{1-x}\text{As}$, where the aluminium concentration x can take any value from 0 to 1. For the most part, our approach will not depend strongly on how this inhomogeneity is manufactured, but for any numerical calculations we will be using the parameters consistent with an $\text{Al}_x\text{Ga}_{1-x}\text{As}$ nanoparticle.

If the material composition is changing that means that both the dielectric constant ϵ and the effective mass μ vary, so we will take both ϵ and μ to be a function of a parameter x , which will in turn be a function of r . In a $\text{Ga}_{1-x}\text{Al}_x\text{As}$ this dependence is linear in x [134]:

$$\epsilon(x) = 12.9(1 - 0.22x)\epsilon_0 \quad \text{and} \quad \mu(x) = 0.063(1 + 1.32x)m_0, \quad (5.2.1)$$

Figure 5.2: (a) In this chapter we consider a spherical nanoparticle of radius R with a hydrogenic impurity and inhomogeneous material composition. The impurity can be described as an ion of charge $+e$ in the center of the sphere. We assume spherical symmetry in the material composition, for both easier calculations and experimental realization. (b) Multiple nanoparticles can then be embedded into a photonic cavity, where the photon mode resonates with the THz emission ladder.

where ϵ_0 is the vacuum permittivity and m_0 is the free electron mass.

The Hamiltonian of the system will be a functional of the function $x(r)$ [135]:

$$H[x(r)] = -\frac{\hbar^2}{2}\nabla\frac{1}{\mu(x(r))}\nabla + \frac{e^2}{4\pi}\int_r^R\frac{dr}{\epsilon(x(r))r^2}, \quad (5.2.2)$$

where the wave function, ψ needs to fulfil the boundary condition $\psi(r) = 0$ for $r \geq R$.

Now, our goal is to find a function $x(r)$ which leads to a ladder of transitions of a specified frequency in the THz range. To do this we need to fix other parameters of the system: the nanoparticle radius R , the coveted transition frequency ω_{THz} and the set of levels that are to take part in the ladder $\{L_i\}$. Note that the transitions need to take place between levels that obey the selection rule $\Delta\ell = \pm 1$, where ℓ is the orbital angular momentum quantum number. We also want to make sure that the highest state of the ladder can be easily excited, meaning that it should be excitable by a transition of higher energy than THz frequency.

To find a function $x(r)$ that has the intended result we make use of variational calculus. The energy of a level denoted by i can be written as

$$E_i = \int_0^R \underbrace{\psi_i^*(r)H[x(r)]\psi_i(r)}_{f(r,x(r))} 4\pi r^2 dr, \quad (5.2.3)$$

where ψ_i is the eigenvalue of the Hamiltonian corresponding to level i . If x is varied by δx the energy level will change by

$$\delta E_i = \int \frac{\delta f}{\delta x} \delta x d^3r \quad (5.2.4)$$

From this we can calculate how to vary the function x to achieve a desired shift in each energy level. We take $x \rightarrow x + \delta x$ where the variation is on the form

$$\delta x = \beta C \Delta E \frac{\delta f}{\delta x}, \quad (5.2.5)$$

with $C^{-1} = \int \left(\frac{\delta f}{\delta x}\right)^2 d^3r$ as a normalization constant and $\Delta E = E_{ideal} - E_i$ where

E_{ideal} is the energy we want to shift level i to coincide with. The factor $\beta \leq 1$ can be tuned to keep the variation small for each step of the iteration so that the system gradually relaxes to an optimum condition. This is then repeated for each level in the chosen level set $\{L_i\}$. This algorithm can then be iterated until each energy level E_i is sufficiently close to the ideal energy.

It is important to note that this is not the only choice that we can make for δx . We could have chosen $\delta x \propto \left(\frac{\delta f}{\delta x}\right)^{-1}$, where the normalization constant would then simply be the volume of the particle. This however would not serve as a good choice since the function $\frac{\delta f}{\delta x}$ can generally take the value 0, leading to a divergence in δx . This would lead to two problems: firstly it would mean that x would need to take large values to fit the calculations, but by definition it can only take values between 0 and 1. Secondly, even if capping x at 1 would be a good approximation, it would mean that the function x would change rapidly around the divergent points which is not desirable as this would be difficult to reproduce experimentally. Also, the parameter β is chosen phenomenologically. Variational calculus assumes that the variation δx is small compared to the function x , but the definition in equation (5.2.5) with $\beta = 1$ will not necessarily fulfil that. We therefore include the parameter $\beta \leq 1$ which can be tuned so that the function δx is never large. Since we iterate the algorithm multiple times we can take β less than unity when needed, so that the energy levels will edge closer to the coveted energies.

After we have found parameters that give agreeable results we can then use the final wave functions to calculate the dipole moment corresponding to available transitions in the system. We then set up rate equations coupling the occupations of the levels as well as the THz photon mode. By introducing a finite lifetime for the photon mode we can simulate putting the nanoparticle inside a cavity resonant to the THz frequency of the photons. Furthermore, we can modify the rate equations to have multiple particles inside the cavity.

We choose the radius R so that the transition energies between the level set $\{L_i\}$ are as close to the coveted energy $\hbar\omega_{THz}$ as needed. When choosing the sets of levels to consider we limit ourselves to the case where the highest level is a p state (angular momentum $\ell = 1$) so that it can be excited by a laser from the ground state. Once

the values of R , ω_{THz} and the level set have been chosen, we modify the function $x(r)$ as described in equation (5.2.5). After enough iterations the energies of the levels corresponding to the chosen level set $\{L_i\}$ should make up an equidistant ladder, with the distance $\hbar\omega_{THz}$.

The function $x(r)$ calculated from $R = 125$ nm, $\omega_{THz} = 1.16$ THz and $\{L_i\} = \{3s, 4p, 5d, 6f, 6d, 6p\}$ after 20 iterations can be seen in Fig. 5.4a, along with the initial condition. Note that the level set does not include the ground state $1s$. This is intentional, since we want to make sure that the system can be pumped by an infrared source. The energy difference between the ground state and the $6p$ state under these parameters happens to lie in that range. Our simple iterative procedure does not guarantee finding a global optimum to the problem at hand, but it can efficiently find a local optimum for a well-chosen initial condition. The graph also shows an approximation of the function which should be easier to fabricate experimentally, plotted in solid light red. This approximation is a step function with 25 steps. Using the approximate functions will lead to a less exact ladder of transitions. This means that we need to take a more lossy cavity, where the cavity photon mode has a larger linewidth and a shorter lifetime.

We find the wave functions of the relevant states by finding the Eigenfunction of the Hamiltonian that corresponds to the energy level. The radial part of the wave functions of the states forming the cascade can be seen plotted on the right hand side of figure 5.4. Since the nanoparticle is spherically symmetric, the angular dependence will be described by the spherical harmonics like in the case of the hydrogen atom. For clarity the intensity of the radial wave functions scaled by r^2 is plotted ($|\psi|^2 r^2$ is proportional to the intensity of a given state at particular r integrated over the angular coordinates). Fig. 5.3 shows the modified levels for the same parameters. The thick, red lines are the chosen set of levels which are to form the equidistant ladder. The black, solid arrows correspond to transitions resonant with the coveted frequency ω_{THz} while the gray, dashed arrows are possible transitions which result in radiation of a different frequency and therefore lead to loss in our system. We assume an external pump that excites electrons in the ground state to the top of our ladder, the $6p$ state. After the electron has been relaxed to

the $3s$ state it will decay to the ground state through lower energy states such as $1p, 2p$ and $3p$. The transition rates between these states and the $3s$ state on one hand and the ground state on the other hand will determine the lifetime of the $3s$ state.

For the current parameters, the ground state of the nanoparticle has the energy -0.11 meV, which means that to excite the particle from the ground state to the top of the ladder we need to pump with a continuous wave of energy of about 5 meV, which is in the infrared range. This puts a limit on our working temperature since the energy of the transition must be much smaller than the thermal energy. This means that the current set up only work in a cryogenic setting with temperature below 58 K. It should be noted that other materials could lead to a higher excitation energy leading to higher possible working temperatures.

Once we have the function $x(r)$, we can find the eigenfunctions of the Hamiltonian in Eq. (5.2.2). Using the eigenfunctions we can calculate the transition rates between the possible transitions. The transition rate between two states labelled with i and j respectively, can be written [136]

$$W_{ij} = \frac{\alpha^4 c}{6a_B^*} \left(\frac{\Delta E_{ij}}{Ry^*} \right)^3 \left(\frac{|\langle i | \mathbf{r} | j \rangle|}{a_B^*} \right)^2, \quad (5.2.6)$$

where α is the fine structure constant, a_B^* and Ry^* are the effective Bohr radius and Rydberg energy for GaAs and ΔE_{ij} is the energy difference between the levels i and j .

With the transition rates known we can write out rate equations for the modes in our system. With n_i denoting the occupation of level i and n_γ the occupation of the photon mode, we have

Figure 5.3: The energy levels in the nanoparticle after using the recursive algorithm discussed in text. The chosen set of levels that make the equidistant ladder are denoted by thick red lines while other levels in the system are thinner, blue lines. The transitions shown by black, solid arrows are all of a single transition energy corresponding to $\omega_{THz} = 1.16$ THz (or 0.76 meV), while the gray, dashed arrows correspond to transitions of different energies and lead to a loss in the system. Other transitions are forbidden due to selection rules.

Figure 5.4: The left hand graph shows the function $x(r)$, which describes the Al concentration in a GaAlAs nanoparticle from of the algorithm described in this paper. The lighter, dashed line corresponds to the initial guess for x . The final result, shown by the dark, solid line, was gotten after only 20 iterations, at which point the convergence was deemed high enough. This $x(r)$ gives rise to an equidistant ladder of transitions for previously chosen energy levels. The step function plotted in solid light red is an approximation of the former curve which should be easier to fabricate. The number of steps is 25. The graph on the right depicts the radial part of the wave functions of said levels. The parameters used were for a $\text{Ga}_{1-x}\text{Al}_x\text{As}$ nanoparticle of radius $R=125$ nm, where the energy difference between the selected levels corresponds to radiation frequency of $\omega_{THz} = 1.16$ THz.

$$\begin{aligned} \frac{dn_i}{dt} = & \sum_{j>i} W_{ij} [n_j(1-n_i)(s_{ij}n_\gamma + 1) - n_i(1-n_j)s_{ij}n_\gamma] \\ & + \sum_{j<i} W_{ij} [n_j(1-n_i)s_{ij}n_\gamma - n_i(1-n_j)(s_{ij}n_\gamma + 1)] \\ & - \frac{n_i}{\tau_i}, \end{aligned} \quad (5.2.7)$$

$$\frac{dn_\gamma}{dt} = N_{QD} \sum_{i,j>i} s_{ij} W_{ij} [(n_j - n_i)n_\gamma + n_j(1 - n_i)] - \frac{n_\gamma}{\tau_\gamma}, \quad (5.2.8)$$

where the levels are ordered in terms of increasing energy, E_i , and $s_{ij} = 1$ if $|E_i - E_j| = E_\gamma$ and $s_{ij} = 0$ otherwise. N_{NP} is the number of particles in the THz cavity. τ_γ is the photon lifetime and τ_i the lifetime of state i in the ladder. The former depends on the quality of the THz cavity while latter will depend on the transition dipole moment of state i to the surrounding states that are not a part of the ladder. In our calculation we looked at the transition rate for each state to lower states outside of the ladder and used their sum to determine the lifetime. This method gives us an overestimate of the loss since we disregard Pauli blocking (by assuming each outside state is always empty) and we disregard the process where the electron can return to a THz emitting state.

We can now use these rate equations to calculate the quantum yield (QY), or how many THz photons we produce each time we excite the nanoparticle to the highest level of the ladder, here the 6s state. When we consider multiple particles in the same cavity we increase the amount of THz photons and therefore increase the probability of stimulated scattering between the transitions marked with solid black arrows in Fig. 5.3. This calculation was done by adding to the coupled rate equations the mode of escaped particles, $n_{esc} = \frac{n_\gamma}{\tau_\gamma}$. The QY is then simply $QY = \frac{n_{esc}(T)}{N_{NP}}$, where the time T is taken to be $T \gg \tau_\gamma$. Fig. 5.5 shows how the QY depends on N_{NP} . The thicker blue line shows the results while using the continuous $x(r)$ we got from running our algorithm for 20 iterations (solid dark curve in the top of Fig. 5.4) and a cavity quality factor of 10^6 . We see that the QY starts low but when the number of nanoparticles in the cavity increases it gets close to the theoretical limit of 5. The

Figure 5.5: The relationship between the quantum yield (QY) and the number of nanoparticles in the THz cavity (N_{NP}). The QY describes how many THz photons we get from the system for each excitation to the highest state. The darker blue line corresponds to the QY when material profile of the nanoparticles follows the smooth $x(r)$ curve shown in figure 2 and the cavity quality factor is taken to be on the order of 10^6 . The red curve is the result when the material profile is described by the step function in the same figure. In that case the quality factor needs to be lower to accommodate for the increased linewidth. Here we take it to be 10^3 . Both curves were calculated using the same parameters as before, nanoparticles of radius $R = 125$ nm and the THz transition is $\omega_{THz} = 1.16$ THz.

thinner red line in Fig. 5.5 corresponds to the results where the material profile is taken to be the step function approximating the continuous function from before. This will be a lot easier to achieve experimentally, but it requires a cavity photon mode with a larger linewidth. Here the quality factor is 10^3 . The shorter lifetime of the photons in the cavity means that we need more particles to achieve the same quality factor as before. We see that we are however still able to achieve a QY of almost 5 for a realistic number of particles, or about $N_{NP} = 10^6$. If we assume a cavity of dimensions $1 \text{ mm} \times 1 \text{ mm} \times 1 \text{ mm}$ that leads to a packing factor of only 10^{-4} , which means that the distance between nanoparticles is large.

As our system essentially amounts to a gain medium, with population inversion, located in a cavity it could also be considered a THz laser. A coherent statistics would be expected of the emission, although we do not calculate it here, restricting ourselves to a semiclassical theory.

5.2.2 Conclusion

We have proposed a new source for THz radiation, a nanoparticle whose material composition changes along the radial direction. The composition is chosen in a way that gives rise to an equidistant ladder of transitions in the THz range. Using this approach we can achieve quantum efficiency far exceeding unity. Here we show numerical calculations for $\text{Ga}_{1-x}\text{Al}_x\text{As}$ nanoparticles to verify our claims, where we find that the quantum efficiency approaches 5, which is the theoretical limit of the current parameters. With increasing the computational power, it would be possible to increase the number of levels considered which could lead to even higher quantum efficiency.

6. Conclusion

This thesis theoretically examined four different systems where light becomes strongly coupled with different material modes leading to different effects stemming from the different dimensionality and natures of the systems.

The first system, discussed in chapter 2, consisted of a semiconductor slab embedded into a zero-dimensional microcavity, leading to vacuum band gap opening of the electron dispersion caused by the interactions with the cavity photon mode. The renormalized dispersion of the three dimensional electron, dressed by coupling to the vacuum photon mode of the cavity, was calculated using the Green's functions approach. The resulting dispersion shows multiple gaps, and their number and structure depend on the direction that the 3D electron is travelling in. The total band gap is on the order of hundreds of μeV , which is larger than other previously predicted band gaps of a similar origin in higher dimensions, and is large enough to be measurable.

In chapter 3 the Green's functions approach is used again, this time to consider a particle in a quantum ring coupling to external circularly polarized light. Since the polarization breaks time reversal symmetry, the particles in the ring exhibit an energy difference of particles with the same magnitude of angular momentum but different directions. For small rings the energy shift of the first few pairs of excited states were considered (the ground state being stationary, the first two excited states being the ones travelling in the opposite directions, with angular momentum quantum number ± 1 , the next pair with ± 2 and so on). The difference of the energies of the first two excited states will always be less than the energy of the levels themselves, but will change sign discontinuously when the frequency of the

light passes resonances of the system. The viability of Aharonov-Bohm oscillations was considered for large rings, where the asymmetry of the dispersion would lead to an oscillation of the conductance through the ring. The results showed that the phase difference will always be a lot less than unity, meaning that no oscillations take place. This contradicts previous results based on perturbative calculations.

Chapter 4 looks at an array of quantum wires embedded into a planar micro-cavity. The coupling between one dimensional excitons in the wires and the two dimensional cavity photon mode leads to the formation of exciton-polaritons. The dispersion, calculated using a Hamiltonian approach based on second quantization, shows a hyperbolic region, where the effective masses along different directions have opposite signs. This is caused by the inhomogeneity of the two directions along and perpendicular to the wires. The system shows some peculiar effects, such as: an incoming pulse can experience either positive or negative refraction depending on the exact energy of the external pump. The system can also sustain dark-soliton like patterns.

The last chapter proposes a new source for terahertz radiation, and outlines a new way of searching for new such sources. The system under consideration here is a spherically symmetric nanoparticle with a hydrogenic impurity, where the material composition varies along the radius in a way that leads to a ladder of equidistant energy levels, where the transition is in the THz range. The material considered is $\text{Ga}_{1-x}\text{Al}_x\text{As}$, where the aluminium concentration x is varying. The function $x(r)$ is found using an iterative algorithm, designed for the energy levels to reach the aforementioned equidistant ladder. The efficiency of particle as a THz source is identified as the number of THz photons achieved by one excitation to the highest state, called the quantum efficiency. For a single particle the quantum efficiency might not be impressive, but when multiple particles are put in a cavity resonant to the THz photons being emitted the efficiency can reach close to the theoretical limit, bound by the number of states taken into consideration during the calculation.

List of Publications

Relevant to this thesis

- [A1] O.V. Kibis, K.B. Arnardottir & I.A. Shelykh, “Band gaps induced by vacuum photons in closed semiconductor cavities,” *Phys. Rev. A* **90**, 055802 (2014).
doi: 10.1103/PhysRevA.90.055802
- [A2] K.B. Arnardottir, I.V. Iorsh, T.C.H. Liew & I.A. Shelykh, “Hyperbolic Region in an Array of Quantum Wires in a Planar Cavity,” *ACS Photonics* **4** (5), 1165 (2017).
10.1021/acsp Photonics.7b00040
- [A3] K.B. Arnardottir & T.C.H. Liew, “Terahertz cascades from a nanoparticle,”
arXiv:1703.07119.

Other publications

- [A4] V.E. Sacksteder, K.B. Arnardottir, S. Kettemann & I.A. Shelykh, “Topological Effects on the Magnetoconductivity in Topological Insulators,” *Phys. Rev. B* **90**, 235148 (2014).
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