Terahertz phenomena in the strong coupling regime

Kristinsson, Kristinn

2017

Kristinsson, K. (2017). Terahertz phenomena in the strong coupling regime. Doctoral thesis, Nanyang Technological University, Singapore.

<http://hdl.handle.net/10356/69475>

<https://doi.org/10.32657/10356/69475>

NANYANG
TECHNOLOGICAL
UNIVERSITY

TERAHERTZ PHENOMENA IN THE STRONG COUPLING REGIME

KRISTINN KRISTINSSON

School of Physical and Mathematical Sciences

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

2016

Acknowledgements

First and foremost I thank my supervisor, Prof. Ivan Shelykh, for guidance, suggestions, ideas, and all and any help he provided to me during my time as his student. I would not have started or finished this undertaking without his involvement.

Second I would like to thank the senior students and researchers I have worked with during the Ph.D., including Dr. Oleksandr Kyriienko, Prof. Timothy Liew, Prof. Oleg Kibis, Prof. Antonio Seridonio, and Dr. Ivan Savenko. They have provided an abundance of assistance to me, and helped me with both the smaller and larger scientific hurdles.

Third I want to thank the other members of our research group, Skender Morina, Helgi Sigurðsson, Kristín Arnardóttir, Anastasia Pervishko, Dr. Tania Espinosa Ortega, Kevin Dini and Vanik Shahnazaryan, for companionship and camaraderie during our time together.

Lastly I thank my wife, Margherita Zuppardo, for her love and support the last almost four years.

Table of content

Acknowledgements	3
Table of content	5
Abstract	7
1 Introduction	9
1.1 Terahertz radiation	10
1.2 Strong coupling and the exciton-polariton	12
1.2.1 The planar optical microcavity	13
1.2.2 Light-matter interaction and the exciton-polariton	17
1.2.3 Coulomb interaction and bistability	21
1.2.4 Polariton devices as terahertz emitters	29
1.3 Dipolaritons	32
1.3.1 Pulsed terahertz emission from dipolaritons	36
1.4 Strong coupling and photon dressing	38
1.4.1 Dressed states in atomic systems	38
1.4.2 Dressed states in condensed matter systems	41
2 Terahertz range emission from dipolariton system	47
2.1 Nonlinear interaction in dipolariton system	48
2.1.1 Model	48
2.1.2 Steady-state calculation and stability analysis	50
2.1.3 Steady-state solutions	53
2.1.4 Dynamic analysis	55

2.1.5	Emission power	58
2.2	Amplification with terahertz cavity	59
2.2.1	Model	60
2.2.2	Low-Q analytical solution	63
2.2.3	Numerical solutions	65
3	Stationary transport in graphene dressed by light	71
3.1	Graphene dispersion	71
3.2	Theory of dressed graphene	75
3.2.1	Model	76
3.2.2	Dispersion and wavefunctions for circular polarization	77
3.2.3	Dispersion and wavefunctions for linear polarization	80
3.2.4	Scattering rates and Boltzmann transport equation	85
3.2.5	Conductivity for circular polarization	88
3.2.6	Conductivity for linear polarization	90
3.3	Results and comments	94
4	Conclusions	97
	List of publications	99
	Appendix A Higher order mean field theory	101
	Appendix B DX-IX interaction constant	106
	Appendix C Calculation of overlap dipole matrix element	117
	Bibliography	119

Abstract

Emission and utilization of terahertz range electromagnetic radiation is an active area of research. This thesis is divided into two main parts, in the first of which a scheme of generation of THz radiation from the dipolariton system is presented. Dipolaritons, a form of exciton-polaritons, are hybrid light-matter particles, arising in a semiconductor microcavity with embedded double quantum wells from the strong coupling between the confined photon and spatially direct and indirect excitons. Oscillations between the direct and indirect excitons and associated oscillation in dipole moment is shown to couple to an external THz cavity mode, resulting in coherent THz emission.

In the second part of the thesis, the influence of THz radiation on transport properties of graphene is investigated. The Dirac-like nature of the graphene electron has long been of theoretical interest, but only recently observed experimentally. By strongly irradiating the graphene sheet, the electron wave-function is altered by the field, resulting in what is commonly called a “dressed electron”. The dressed graphene is shown to have strongly modified transport properties. Specifically it is shown that with linear polarization of the dressing field there arises a large anisotropy of conductivity between the directions parallel and perpendicular to the polarization direction.

1. Introduction

The interaction between light and matter is a fundamental topic, necessary to describe fundamental phenomena such as emission spectra as well as to predict and design fundamental modern devices such as lasers [1–3]. One of the most interesting phenomena at this boundary between condensed matter physics and quantum optics is the *strong coupling* regime – the situation where coupling strength between light and matter exceeds loss rates. In this case the individual photon and matter modes recede in lieu of new mixed light-matter eigenmodes of the combined system. Due to their hybrid nature they inherit critical properties of each parent particle.

In the electromagnetic spectrum one range of frequencies remains under-utilized. The elusive *terahertz* range, ranging from 0.3 to 3 THz, has been subject of much interest and active research in recent years [4,5]. In the second chapter of this thesis a scheme for the generation of continuous and relatively tunable THz range emission is presented. It is based on the system of *dipolaritons*, which is treated in section 1.3. The dipolariton system consists of an optical microcavity with double embedded quantum wells (QWs), where excitons possess non-zero dipole moment. Oscillations of the dipole moment induced by the external pump result in THz emission.

In the third chapter of this thesis a different form of strong coupling is investigated in *graphene*. This two-dimensional hexagonal lattice of carbon atoms has been of strong interest in the scientific community since its discovery, particularly due to its linear, Dirac-like dispersion [6,7]. In this chapter THz radiation serves as an ingredient rather than being the objective. By irradiating a graphene sheet with a non-absorptive THz field, a different form of strong coupling is realized where the effect of the field is to modify the dispersion of the graphene. The resulting particle

is referred to as a *dressed electron*. The polarization of the irradiation is shown to be crucial. While the circularly polarized field opens up a gap in the graphene dispersion, the linearly polarized field breaks the spatial symmetry of the graphene. The result is strong anisotropy of conductivity, with the conductivity in the direction perpendicular to the polarization of the conductivity being heavily suppressed for certain values of the irradiation intensity.

1.1 Terahertz radiation

Despite its relative obscurity, THz radiation has many important areas of application outside the laboratory, such as security imaging [8] and medical imaging [9, 10]. In the sciences it is important as well, in such domains as astronomy [11] and nondestructive spectroscopy [12]. However, there is still a lack of universal, flexible and efficient emission devices.

First and foremost among the existing technologies of THz frequency emission is the quantum cascade laser (QCL) [13]. The QCL was described theoretically in 1971 [14], and consists of a system of parallel QWs, with an external electric field that tilts the energy bands. This allows particles in one QW ground state to tunnel through the potential barrier to the next QW with energy matched excited state. Relaxing to the ground state again it emits a photon containing the energy difference, as illustrated in Fig. 1.1. A slight modification of the device was first realized experimentally in 1994 [15]. The QCL is not restricted to the THz range, but can rather be tuned by the QW width, altering the energy levels and thereby the emission frequency. The QCL has mainly been designed for infra-red emission, but has been realized for the upper ranges of the THz spectrum with output power reaching 145 mW [16].

A second class of devices that allow emission in the THz range act as optical to THz converters, where ultrashort laser pulses generate oscillations of charge density, thus acting as a classical oscillating dipole, emitting THz radiation [17]. This principle has been applied to oxide nanojunctions [18], graphite flakes [19] and semiconductor-metal interfaces [20]. A similar scheme of THz emission utilizes crys-

Figure 1.1: Energy diagram of a quantum cascade laser with the spatial growth direction marked as z . Each quantum well is shown as having a ground state and one excited states. An applied electric field tilts the energy structure in the z -direction, tuning the excited state and ground state of neighbouring QWs into resonance. Particles tunnel from one QW ground state to the next QW excited state, and relax into the ground state, releasing a photon.

tals with strong second-order nonlinear optical response [21]. The THz waves arise through difference frequency generation between close to equal frequency components that are naturally present in non-ideal lasers. This is referred to as optical rectification.

Other devices that can produce THz radiation include the Gunn diode and the free-electron lasers. Gunn diodes are principally designed for microwave emission with applications such as radar speed guns, but have been realized for emission in the lower limit of the THz range [22]. Free-electron lasers meanwhile are the most widely tunable of all lasers, their spectrum ranging from microwaves, through visible wavelengths, and even into X-ray frequencies [23]. Although powerful and efficient emission in the THz range was reported [24], the free-electron laser still lacks a lot of desirable properties. Its principal drawback is the bulky size of the apparatus, in which a free electron beam is made to oscillate in strong magnetic fields to produce the radiation, a scheme which does not allow miniaturization.

The exciton-polariton system, described in the next section, will be shown to allow further techniques of THz emission, exploiting an energy splitting between modes that naturally lies in the THz range. In particular the dipolariton system, with its large dipole moment, will be shown to be especially useful. In chapter 2 a

technique for production of strong, continuous emission of THz radiation from the dipolariton system is developed.

1.2 Strong coupling and the exciton-polariton

The most important concept behind the work presented in this thesis is the notion of strong coupling between a material system and the photonic mode. This principle builds on the interaction of spatially confined light modes with material modes. Any trapping of light will be characterized by a finite quality factor, commonly defined as

$$Q = \frac{\omega}{\Delta\omega}, \quad (1.2.1)$$

where ω is the frequency of the confined light mode, and $\Delta\omega$ is the linewidth of the mode coming from imperfect confinement. This quality factor describes how well the light is confined, and is proportional to the mean lifetime $\tau = Q/\omega$ of a photon. If the loss rate $1/\tau$ as well as the loss rate of the matter system are much smaller than the rate of coupling between the modes, strong coupling is achieved [25].

Strong coupling between individual quantum dots and photonic crystal modes allowed the investigation of several phenomena of interest to quantum information, such as entanglement and antibunching [26–29]. Strong coupling has also been observed with whispering gallery modes, where total internal reflection provides exceedingly high quality factor of light confinement [30–32].

The strong coupling system most relevant to this thesis however is that of the *microcavity exciton-polariton*. In this system light is contained in a planar optical microcavity, which traps light in one of the spatial dimensions [25]. The optically active media is represented by the embedded quantum well (QW) with excitonic resonance. If the energy of the exciton lies close to the energy of the cavity mode, multiple processes of exciton absorption and emission occur, and the system can be driven into the strong coupling regime.

1.2.1 The planar optical microcavity

In the optical microcavity the confinement of the light is provided by distributed Bragg reflectors (DBRs). A DBR consists of repeating alternating layers of dielectric media of thickness a and b , with refractive indices n_a and n_b respectively, as shown in Figure 1.2(a). To calculate the reflectivity of such a one-dimensional array, the standard approach is the use of the transfer-matrix method [33]. Lets consider an electromagnetic wave (EM wave) of wavelength λ travelling along one dimension, at normal incidence to the DBR layers. The wave in one medium is fully described by the electric field

$$E(z) = E_R e^{ikz} + E_L e^{-ikz},$$

where $k = 2\pi n/\lambda$, n is the refractive index, and the z -axis lies perpendicular to the DBR layers. We now define the vector $\Phi(z) = (E(z), dE(z)/dz)^T$. Since the electric field of freely propagating light is perpendicular to the travel direction, both the electric field and its z -derivative are tangential to the DBR layer boundaries. Maxwell equation boundary conditions furthermore dictate that the tangential components of both $E(z)$ and $dE(z)/dz$ are continuous over the boundary between two dielectric media. Thus the vector $\Phi(z)$ is continuous over the DBR layer boundaries.

It can be easily shown that the propagation of the EM wave through a layer of constant refractive index n_d of thickness d is described by the expression $\Phi(z+d) = \mathbf{T}_d \Phi(z)$, where $\mathbf{T}_d$, the transfer matrix over section d , is given by

$$\mathbf{T}_d = \begin{pmatrix} \cos k_d d & k_d^{-1} \sin k_d d \\ -k_d \sin k_d d & \cos k_d d \end{pmatrix}, \quad (1.2.2)$$

where $k_d = 2\pi n_d/\lambda$ is the wavenumber of the wave in the refractive medium. As such, the propagation of light trough one period in the DBR is given by $\Phi(z+a+b) = \mathbf{T}_b \Phi(z+a) = \mathbf{T}_b \mathbf{T}_a \Phi(z)$. Applying this combined transfer matrix once for every periodic layer, the effective transfer matrix for the propagation across the whole DBR is given by

$$\mathbf{T} = (\mathbf{T}_b \mathbf{T}_a)^N. \quad (1.2.3)$$

Figure 1.2: **(a)** Sketch of a distributed Bragg reflector (DBR). The DBR consists of alternating layers of refractive index n_a and n_b , with thicknesses a and b fulfilling the condition $n_a a = n_b b$. Incoming light of wavelength $\lambda = 4n_a a = 4n_b b$ is strongly reflected, with reflectivity which rapidly approaches unity as the number of layers grows. **(b)** Reflectivity characteristic of a DBR in the spectrum of light wavelength. The wavelength chosen by the interference condition (1.2.5) is 1000 nm. The refractive indices were chosen as $n_a = 1.5$ and $n_b = 2.1$, corresponding to alternating layers of SiO_2 and HfO_2 . The number of layers was $N = 12$.

From the total transfer matrix we can calculate the reflectivity of a DBR on a substrate of refractive index n' in air as $R = |r|^2$, where

$$r = \frac{T_{21} + k'k_0T_{12} + i(-k'T_{11} + k_0T_{22})}{-T_{21} + k'k_0T_{12} + i(k'T_{11} + k_0T_{22})}, \quad (1.2.4)$$

is the amplitude reflection coefficient, $k_0 = 2\pi/\lambda$ and $k' = 2\pi n'/\lambda$ are the wavenumbers in air and the substrate respectively, and T_{ij} are the matrix elements of $\mathbf{T}$.

Now if the thicknesses of the layers and the wavelength of the incident light are chosen such that

$$\frac{\lambda}{4} = n_a a = n_b b. \quad (1.2.5)$$

Then the single period transfer matrix is

$$\mathbf{T}_b \mathbf{T}_a = \begin{pmatrix} 0 & \frac{1}{k_b} \\ -k_b & 0 \end{pmatrix} \begin{pmatrix} 0 & \frac{1}{k_a} \\ -k_a & 0 \end{pmatrix} = \begin{pmatrix} -\frac{k_a}{k_b} & 0 \\ 0 & -\frac{k_b}{k_a} \end{pmatrix} = - \begin{pmatrix} \frac{n_a}{n_b} & 0 \\ 0 & \frac{n_b}{n_a} \end{pmatrix}. \quad (1.2.6)$$

As this matrix is diagonal, the N -th power is found by simply taking the N -th power of the individual components. From equation (1.2.4) we then find the amplitude reflection coefficient as

$$r = \frac{(n_a/n_b)^{2N} - n'}{(n_a/n_b)^{2N} + n'}. \quad (1.2.7)$$

This expression approaches ± 1 exponentially as number of layers N increases for non-equal refractive indices. The reflectivity also increases with the contrast between the refractive indices of the two materials. As such, with a reasonable number of layers, the DBR can be made very close to perfectly reflective.

For the dependance of reflectivity of a DBR on wavelength, away from the wavelength defined by the condition (1.2.5), see the Figure 1.2(b). For wavelengths close to the one defined by the interference condition the reflectivity remains close to unity, in what is referred to as the stopband. Outside the stopband the reflectivity is much smaller.

DBRs, being mesoscopic structures, are not trivial in construction. The traditional experimental process used is metalorganic chemical vapour deposition (MOCVP). In this process, pure gases of carefully chosen compounds are injected over the substrate, such that the desired substance forms on the substrate by chemical reaction [34]. This technique has reached considerable level of sophistication, with various groups having demonstrated samples with reflectivity as high as 97% in a 20 layer GaN/AlN structure [35], and above 99% in both a 60 layer $\text{Al}_{0.2}\text{Ga}_{0.8}\text{N}$ /GaN structure [36] and a 35 layer InP/InGaAlAs structure [37]. In another work nanomembranes of silicon were partially oxidized, and then tranferred on top of each other, forming a DBR of 3 layers of Si/SiO₂ pairs, resulting in 99.8% reflectivity [38].

The microcavity is formed by growing a defect layer with refractive index n on top of a DBR, and growing a second DBR on top, as shown in figure 1.3. Assuming the reflectance of the mirrors is perfect, the interference condition of a standing wave between the mirrors is [25]

$$k_z L = j\pi, \quad (1.2.8)$$

where k_z is the z -component of the wave vector of the light inside the cavity, L is the thickness of the cavity (defect layer) and j is a positive integer. The defect

Figure 1.3: Sketch of the optical microcavity. The microcavity is formed by two DBRs with alternating layers with indices of refraction n_a and n_b , and a defect layer of a third material with index of refraction n . The thickness L of the defect layer should be a whole multiple of the cavity mode half-wavelength $\lambda/2$.

layer should therefore have a thickness equal to a whole multiple of $\lambda/2$, where the wavelength λ of the resulting cavity mode lies in the stopband of the DBRs. Equivalently, the mode characterized by index J has wavelength $\lambda_j = 2L/j$. The free spectral range can be found as $\Delta\lambda_j = \lambda_j - \lambda_{j+1} = 2L/j(j+1)$. A microcavity distinguishes itself from other optical cavities by its thickness, which is typically one or several half wavelengths of the optical mode. The ratio

$$\frac{\Delta\lambda_j}{\lambda_j} = \frac{1}{j+1} \quad (1.2.9)$$

between the mode wavelength separation and the mode wavelength is therefore on the order of unity. This large free spectral range, along with the finite width of the stopband guarantees that in most cases light of only one wavelength will be contained in the microcavity.

If the light inside the microcavity is imagined to have a wave vector with a small non-zero component $k \ll k_z$ in the plane parallel to the mirrors, the energy of the mode becomes

$$E_c(k) = \frac{\hbar c}{n} \sqrt{k_z^2 + k^2} \approx \frac{\hbar c k_z}{n} + \frac{\hbar c k^2}{2n k_z}. \quad (1.2.10)$$

The first term is equal to the energy of the perpendicular incidence mode $\hbar\omega_c$, where $\omega_c = j\pi c/nL$. The second term can be written in the typical form $\hbar^2 k^2/2m_c$ of a quadratic dispersion, where the effective mass of the 2D motion of the cavity mode reads

$$m_c = \frac{j\pi n\hbar}{cL}. \quad (1.2.11)$$

This cavity mode mass is typically of the order of $10^{-5} - 10^{-4} m_0$, where m_0 is the free electron mass.

1.2.2 Light-matter interaction and the exciton-polariton

The optically active medium in the exciton-polariton system is the QW, a material with band energy lower than the defect layer material, grown inside the microcavity. In this layer electrons and holes can separate into two-dimensional hydrogen-like bound pairs, called excitons. The size of the Wannier-Mott (semi-conductor) exciton is characterized by its Bohr radius a_B , which is typically of the order of 10 nm. Its binding energy depends on the width of the QW, but is in most cases of the order of several millielectronvolt (meV). The dispersion of the 2D exciton is approximately parabolic, with the effective mass being the reduced mass $\mu = m_e m_h / (m_e + m_h)$ of the electron and hole.

It is important to consider the statistics of the modes of this system. Both electrons and holes are half-integer spin objects, and such are subject to the Fermionic commutation relations, which excludes identical particles from being in the same quantum state. The bound excitons, being composed from two half-spin objects have integer spin, and consequently their commutation relations are bosonic. This however is only valid as long as the wavefunctions of two electrons or holes of different excitons do not substantially overlap. As such we have an upper limit on the density of excitons n_{exc} for which we can treat them as bosons, which reads

$$n_{exc} \ll \frac{1}{a_B^2}. \quad (1.2.12)$$

If the density of excitons exceeds this limit, the Mott transition to electron-hole

plasma occurs.

We shall describe the system in the basis of second-quantized operators, where $\hat{a}_{\mathbf{k}}^\dagger$ ($\hat{a}_{\mathbf{k}}$) is the creation (annihilation) operator of a cavity photon with in-plane momentum $\mathbf{k}$, and $\hat{b}_{\mathbf{k}}^\dagger$ ($\hat{b}_{\mathbf{k}}$) the creation (annihilation) operator of a QW exciton with in-plane momentum $\mathbf{k}$. The Hamiltonian of the photon-exciton system can now be written as

$$\hat{\mathcal{H}}_0 = \sum_{\mathbf{k}} E_c(\mathbf{k}) \hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}} + \sum_{\mathbf{k}} E_x(\mathbf{k}) \hat{b}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}} + \sum_{\mathbf{k}} \frac{\hbar\Omega}{2} \left(\hat{a}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}} + \hat{b}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}} \right). \quad (1.2.13)$$

where $E_x(\mathbf{k}) = E_x^0 + \hbar^2 k^2 / 2\mu$ is the dispersion of the exciton. The interaction consists of two terms describing the opposite processes of recombination of an exciton accompanied by the emission of a cavity photon in the first term, and the absorption of a photon accompanied by the creation of an exciton in the second term. The interaction strength $\hbar\Omega$ is referred to as Rabi splitting. It is proportional to the dipole matrix element of the exciton, and is typically in the range of several meV, for example 4 meV in ref. [39].

The linear Hamiltonian (1.2.13) can be diagonalized straightforwardly. This results in new eigenfunctions representing new eigenmodes of the system. These eigenmodes are called *exciton-polaritons*, or polaritons as a shorthand. The annihilation operators of the *upper* and *lower polariton* are given respectively by

$$\begin{aligned} \hat{p}_{+\mathbf{k}} &= C_+ \hat{a}_{\mathbf{k}} + X_+ \hat{b}_{\mathbf{k}}, \\ \hat{p}_{-\mathbf{k}} &= C_- \hat{a}_{\mathbf{k}} + X_- \hat{b}_{\mathbf{k}}. \end{aligned} \quad (1.2.14)$$

Here $C_\pm$ and $X_\pm$ are called the Hopfield coefficients. The modulus squared of $C_\pm$ and $X_\pm$ gives the fraction of the photon and exciton contribution respectively to the

upper (+) and lower (-) polariton. The coefficients are given by

$$C_+ = X_- = \frac{E_c(\mathbf{k}) - E_-(\mathbf{k})}{\sqrt{(E_c(\mathbf{k}) - E_-(\mathbf{k}))^2 + (\hbar\Omega/2)^2}}, \quad (1.2.15)$$

$$X_+ = -C_- = \frac{\hbar\Omega/2}{\sqrt{(E_c(\mathbf{k}) - E_-(\mathbf{k}))^2 + (\hbar\Omega/2)^2}}. \quad (1.2.16)$$

The energy-wavevector dispersion is further given by [see Fig. 1.4(a)]

$$E_{\pm}(\mathbf{k}) = \frac{E_c(\mathbf{k}) + E_x(\mathbf{k})}{2} \pm \frac{1}{2} \sqrt{(E_c(\mathbf{k}) - E_x(\mathbf{k}))^2 + \hbar^2\Omega^2}. \quad (1.2.17)$$

Now we define $\hbar\Delta = E_c(0) - E_x(0)$ as the zero-wavevector detuning of the cavity mode relative to the exciton energy. We can then define the zero energy point as the exciton energy at zero wavevector to remove references to explicit mode energies. Then it follows that $E_x(\mathbf{k}) = \hbar^2 k^2 / 2\mu$.

For small wavevectors ($k \ll \sqrt{m_e\Omega/\hbar}, \sqrt{m_e\Delta/\hbar}$) the dispersion is approximately parabolic. Taking into account that the mass m_c of the cavity mode is smaller than that of the exciton by more than a factor of 10^{-3} the dispersion can be expressed as

$$E_{\pm}(\mathbf{k}) = \frac{\hbar^2 k^2}{2m_{\pm}} + \hbar \frac{\Delta \pm \sqrt{\Delta^2 + \Omega^2}}{2} \quad (1.2.18)$$

where

$$m_{\pm} = 2m_c \frac{\sqrt{\Delta^2 + \Omega^2}}{\sqrt{\Delta^2 + \Omega^2} \pm \Delta}. \quad (1.2.19)$$

For example, in the case of zero detuning $\Delta = 0$, the zero-wavevector effective mass of both polariton branches is $2m_c$. In the limit of large detuning, the reduction of the coupling efficiency is evident in that the effective mass of one polariton mode tends to the cavity mode mass m_c , and that of the other to infinity, reflecting the scale of the small wavevector approximation.

The existence of exciton-polaritons is as mentioned before dependent upon the strong coupling of the cavity mode and excitons, necessitating low losses. As the primary mechanism of loss of excitations in the polariton system is through the non-

Figure 1.4: **(a)** Dispersion of the upper (UP) and lower (LP) exciton-polariton modes is shown in accordance with eq. (1.2.17). Detuning at zero wavevector is taken as $\hbar\Delta = 0$, and the Rabi splitting as $\hbar\Omega = 6$ meV. The dispersion of the cavity mode (C) and exciton (X) is shown with red and blue dashed lines respectively. The color of the UP and LP lines indicates photon and exciton fractions, represented by the primary colors of red and blue respectively. The pink segments around zero wavevector indicate heavy mixing of the modes where they lie close in energy. Here the level repulsion is evident, with both polariton modes having quadratic dispersion with an effective mass on the order of that of the cavity mode. For large values of k the UP and LP modes become purely photonic and excitonic respectively. **(b)** Upper and lower polariton energy levels at zero wavevector are plotted as a function of the detuning of the exciton and cavity mode energy levels. Rabi splitting remains $\hbar\Omega = 6$ meV, and the coloring scheme is identical to that in (a). For large detunings the modes mix little. Around the anti-crossing point of zero detuning the mixing however is large, and the polaritons acquire properties of both constituent atoms.

zero transmission of the DBRs, considerable experimental sophistication is required to be able to observe what has been described. This was first achieved in 1992, when a reflectivity spectroscopy measurement of a microcavity with embedded QW was performed for various values of the cavity mode-exciton detuning [as in Fig. 1.4(b)], revealing an anti-crossing of the energy levels [39]. This level repulsion is evidence of coupling strength being sufficiently larger than losses, $\Omega \gg \tau_c^{-1}, \tau_X^{-1}$, where τ_c, τ_X are the lifetimes of the cavity mode and exciton. This shows strong coupling has been achieved.

Since then polaritons have been of strong interest both to experimental and

theoretical scientists, with review articles having been published in 2010 [40] and 2013 [41]. The phenomena that is perhaps of most fundamental interest of those studied in the polariton system is that of the Bose-Einstein condensate (BEC). BEC is a low-temperature state of bosonic matter in which occupation of the ground state becomes much larger than that of other states. As the polariton system is inherently lossy, with lifetimes on the order of picoseconds, a true BEC can not form. By balancing losses with gains, a quasi-equilibrium analog of BEC has been shown to be possible in the ground state of the lower polariton branch [42,43]. Because the critical temperature of condensation is inversely proportional to mass, and the effective mass of the polariton is on the order of the cavity mode mass, these first experimental observations of the polariton BEC were possible at standard cryogenic temperatures, a far cry from the nanokelvin temperatures at which a true BEC was first observed [44]. By more careful choice of materials, and advancement in experimental techniques, polariton BECs have since been observed at room-temperature [45,46].

The most well known application of the polariton BEC is that of the polariton laser, where the coherence of bosons in a BEC replaces population inversion as the mechanism of coherence [47]. In this scheme a condensate is produced at the highly mixed $k = 0$ point. Photons that leak through the DBRs will come in overwhelming part from the polaritons that form part of the BEC. As such the emitted radiation carries the necessary coherence of laser light. As there is no need for population inversion the threshold power is much lower than that of a conventional vertical-cavity surface-emitting laser (VCSEL) [48]. In an important step towards the wider adoption, an electrically injected polariton laser, which distinguishes itself from the earlier optically driven realizations, was independently realized experimentally by two groups in 2013 [49,50]. Since then the electrically injected polariton laser has also been shown to be possible at room temperature [51].

1.2.3 Coulomb interaction and bistability

An important consequence of the excitonic contribution to the polariton is the strong nonlinear interaction between excitations. Nonlinear interactions in photonic systems are for example widely discussed to allow for the generation of nonclassical

states of light, in the form of single photons or photon pairs [52, 53]. They necessary ingredients in devices that exploit various principles of quantum information, such as quantum cryptography [54]. In the domain of polaritonics it was shown that the nonlinear interactions between polaritons allow single photon emission [55].

An important property that arises from nonlinear interactions is that of multi-stability. It can be described in its most simple form in the polariton system. As was mentioned already, the origin of polariton nonlinear interaction is the Coulomb interaction between the charged electrons and holes of the exciton. This interaction modifies the Hamiltonian (1.2.13) adding a quadratic term

$$\hat{\mathcal{H}}_1 = \hat{\mathcal{H}}_0 + \frac{1}{2} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} M_{\mathbf{k}\mathbf{k}'\mathbf{q}} \hat{b}_{\mathbf{k}+\mathbf{q}}^\dagger \hat{b}_{\mathbf{k}'-\mathbf{q}}^\dagger \hat{b}_{\mathbf{k}} \hat{b}_{\mathbf{k}'}. \quad (1.2.20)$$

The interaction matrix element can be approximated as a constant for small wavevectors [56], as $M_{\mathbf{k}\mathbf{k}'\mathbf{q}} \approx 6E_b a_B^2 / S$, where E_b is the exciton binding energy and S the QW area.

We shall concern ourselves only with the lower energy LP mode. As such we will drop the “-” index on the lower polariton operator defined in eq. (1.2.14). Further we will neglect spatial dependence, as we wish only to describe in simplest terms a spatially homogeneous effect. As such we drop the “ $\mathbf{k}$ ” indices. In addition to the terms of the Hamiltonian (1.2.20) we shall also account for continuous wave (CW) coherent (single frequency) optical excitation of the cavity mode. In terms of the polariton operators we can then write the simplified Hamiltonian as [57]

$$\hat{\mathcal{H}} = \hbar\omega_p \hat{p}^\dagger \hat{p} + \frac{V}{2} \hat{p}^\dagger \hat{p}^\dagger \hat{p} \hat{p} + P_0 e^{-i\omega_d t} \hat{p}^\dagger + P_0^* e^{i\omega_d t} \hat{p}, \quad (1.2.21)$$

where $\hbar\omega_p$ is the polariton mode energy, $V = 6E_b a_B^2 |X_-|^4 / S$ is the polariton-polariton interaction constant, $|P_0|^2$ is the intensity of the driving, and ω_d its frequency.

To obtain equations of motion, the *mean-field approximation* shall be employed. It entails using the Heisenberg equation $d\hat{p}/dt = -i[\hat{p}, \hat{\mathcal{H}}]/\hbar$ to find the evolution of the operator, then taking the expectation value $\langle \hat{p} \rangle = \text{Tr}\{\hat{\rho}\hat{p}\}$, and applying the

approximation $\langle \hat{a}\hat{b} \rangle \approx \langle \hat{a} \rangle \langle \hat{b} \rangle$ for any two operators $\hat{a}$ and $\hat{b}$. By doing this approximation we discard information of the correlation between the modes, only retaining their magnitude and phase. Correlations play a large role in quantum phenomena, but as the number of excitations increase, the role of correlations diminishes. The effect that will be described is therefore often called semiclassical. In order to ascertain the validity of this approximation, higher order mean field equations were derived and compared to the equations used in this thesis, confirming the accuracy of the latter [see Appendix A].

The equation of motion obtained from the Hamiltonian (1.2.21) is

$$\frac{d\langle \hat{p} \rangle}{dt} = -i\omega_p \langle \hat{p} \rangle - i\alpha |\langle \hat{p} \rangle|^2 \langle \hat{p} \rangle - \frac{1}{2\tau} \langle \hat{p} \rangle - i\tilde{P}_0 e^{-i\omega_d t}, \quad (1.2.22)$$

where $\alpha = V/\hbar$ and $\tilde{P}_0 = P_0/\hbar$. Here we added as the third term the decay process phenomenologically, with lifetime of excitation τ . For a well constructed cavity this lifetime is on the order of picoseconds. The stationary solution to this equation can be found with a function of the form

$$\langle \hat{p} \rangle = \psi e^{-i\omega_d t}, \quad (1.2.23)$$

where $|\psi|^2 = |\langle \hat{p} \rangle|^2 = N$ is expectation value of the number of polaritons. This yields an algebraic equation for ψ as

$$(\omega_d - \omega_p)\psi - \alpha |\psi|^2 \psi + \frac{i}{2\tau} \psi = \tilde{P}_0. \quad (1.2.24)$$

This is a third order equation in the complex quantity ψ . If we multiply the equation by its complex conjugate we can get a simpler equation for the real number N as

$$\left[(\delta - \alpha N)^2 + \frac{1}{4\tau^2} \right] N = |\tilde{P}_0|^2, \quad (1.2.25)$$

where $\delta = \omega_d - \omega_p$ is the detuning of the pump from the polariton mode energy. The equation (1.2.25) describes the steady state number of polaritonic excitations under a certain strength of coherent CW excitation. The central property of this

equation is that being a third order in N it admits up to three different solutions of population sizes for each value of the pump power, two of which are stable. Thus the system demonstrates the phenomenon of bistability.

To analyze eq. (1.2.25) further, it is useful to consider the excitation number N as a free variable, and the pumping power as the dependent variable [see Fig. 1.5(a)]. If the pumping intensity increases monotonically with N there is no multistability. If there is multistability there will be a point where the derivative of pumping power with regards to N is zero. The derivative reads

$$\frac{d|\tilde{P}_0|^2}{dN} = 3\alpha^2 N^2 - 4\delta\alpha N + \delta^2 + \frac{1}{4\tau^2}. \quad (1.2.26)$$

Setting the derivative equal to zero yields an equation second order in N , with solutions

$$N_{\pm} = \frac{2\delta \pm \sqrt{\delta^2 - 3/4\tau^2}}{3\alpha}. \quad (1.2.27)$$

The solution for excitation numbers (1.2.27) should be real and positive. The condition of realness implies that $\delta^2 > 3/4\tau^2$. The condition of positivity implies that $2\delta > \sqrt{\delta^2 - 3/4\tau^2}$, which is fulfilled if and only if the detuning δ is positive, as long as the realness condition is fulfilled. We can therefore express both conditions as a single necessary and sufficient condition for multistability on the driving laser frequency, which reads

$$\omega_d > \omega_p + \frac{\sqrt{3}}{2\tau}. \quad (1.2.28)$$

It is important to consider the nature of these solutions to the eq. (1.2.25), as we know from nature many different systems that have solutions that are not stable to small perturbations, such as the proverbial pen balanced on its tip. The correct way to do perform this stability analysis is to go back to the dynamic equation (1.2.22), and replace the solution (1.2.23) with a solution with a small added time-dependent deviation:

$$\langle \hat{p} \rangle = (\psi + \delta\psi(t))e^{-i\omega_d t}, \quad (1.2.29)$$

where ψ is the solution to eq. (1.2.24). Now this ansatz is inserted into the dynamic equation, and all terms of second order or higher in the small deviation $\delta\psi(t)$ are

Figure 1.5: **(a)** CW pumping intensity $|P_0|^2$ as a function of polariton numbers $N = |\psi|^2$, in accordance with eq. (1.2.25), for three different values of the lifetime τ of the polariton, producing qualitatively different behaviors. The topmost line is for $\tau < \sqrt{3}/2\delta$, for which condition (1.2.28) is not fulfilled, and the pumping power increases monotonically with polariton number N . As such, there is no multistability. The lowest line represents $\tau > \sqrt{3}/2\delta$, implying there is a range of pumping intensity where each value corresponds to multiple values of the CW polariton population. The middle line illustrates the intermediate equality case, in which there is no multistability, but a saddle point. **(b)** Imaginary part of the eigenvalue Ω given by eq. (1.2.34) as a function of polariton numbers N . For $N < \delta/3\alpha$ and $N > \delta/\alpha$ the imaginary part is constant $-1/2\tau$. Between these values the imaginary parts of the two eigenvalues of Ω split, with Ω_- carrying an additional negative imaginary part and Ω_+ carrying an additional positive imaginary part of up to $\delta/\sqrt{3}$. If inequality (1.2.28) is fulfilled, the imaginary part becomes positive in the range $N_- < N < N_+$. **(c)** Qualitative plot of polariton numbers as a function of CW pump intensity for the case where there is multistability. The dashed line indicated the solution in unstable. If the pumping intensity is increased adiabatically from zero the polariton number stays on the lower branch, until it reaches N_- . Increasing the pumping power further makes the solution jump to the higher branch, represented by the arrow between the branches. Lowering the pumping power slowly again has the polariton number staying on the upper branch, until it reaches N_+ . At that point lowering the pumping power further results in the polariton numbers jumping down to the lower branch, as indicated by the other arrow, completing the hysteresis curve.

neglected. The equation (1.2.24) is then used to eliminate terms not containing $\delta\psi(t)$. This yields a dynamic equation for the deviation reading

$$i\frac{d}{dt}\delta\psi(t) = \left(-\delta - \frac{i}{2\tau} + 2\alpha|\psi|^2\right)\delta\psi(t) + \alpha\psi^2\delta\psi(t)^*. \quad (1.2.30)$$

We now consider the deviation to have the explicit form

$$\delta\psi(t) = ue^{-i\Omega t} + ve^{i\Omega^* t}. \quad (1.2.31)$$

This will allow us to say something about the behavior of the deviation by the possible values of the frequency Ω . If the imaginary part of the frequency is positive and non-zero, the deviation decays to zero in both terms, and the solution is stable. If however it is negative, even the smallest of deviations will increase exponentially from the solution to eq. (1.2.25), and it is therefore as unstable as a pen resting on its tip. Substituting ansatz (1.2.31) into eq. (1.2.30) yields a matrix eigenvalue problem $\mathcal{M}\delta\Psi = \Omega\delta\Psi$, where $\delta\Psi = (u, v)^T$, and

$$\mathcal{M} = \begin{pmatrix} -\delta - \frac{i}{2\tau} + 2\alpha|\psi|^2 & \alpha\psi^2 \\ -\alpha\psi^{*2} & \delta - \frac{i}{2\tau} - 2\alpha|\psi|^2 \end{pmatrix} \quad (1.2.32)$$

This eigenvalue problem has a solution if and only if

$$\det(\mathcal{M} - \Omega\mathcal{I}_2) = 0, \quad (1.2.33)$$

where $\mathcal{I}_2$ is the two-dimensional identity matrix, and $\det$ denotes the matrix determinant. The eq. (1.2.33) is second order in Ω , and upon substituting $N = |\psi|^2$ has the two solutions

$$\Omega_{\pm} = -\frac{i}{2\tau} \pm \sqrt{(\delta - 2\alpha N)^2 - \alpha^2 N^2}. \quad (1.2.34)$$

The first point that should be made is that the imaginary part of Ω_- is always negative. Therefore it is the sign of the imaginary part of Ω_+ that decides whether the solution is stable or not, as both modes need to decay. The argument of the

square root and its sign play an important part, so it is useful to define the function

$$f(N) = (\delta - 2\alpha N)^2 - \alpha^2 N^2. \quad (1.2.35)$$

This function is upwards opening parabolic, and takes its minima $f(N^*) = -\delta^2/3$ at $N^* = 2\delta/3\alpha$. Both zeros of the function lie in the positive N domain, at $\delta/3\alpha$ and δ/α . As such for $N < \delta/3\alpha$ and for $N > \delta/\alpha$ the function $f(N)$ is positive, and $\sqrt{f(N)}$ is real. [see Fig. 1.5(b)]. In these ranges the imaginary part reads as constant $\text{Im } \Omega_+ = -1/2\tau$.

For $\delta/3\alpha < N < \delta/\alpha$ the function $f(N)$ is negative, and can be written as $\sqrt{f(N)} = \sqrt{-|f(N)|} = i\sqrt{|f(N)|}$. The imaginary part of the frequency Ω_+ then reads

$$\text{Im } \Omega_+ = -\frac{1}{2\tau} + \sqrt{|f(N)|}. \quad (1.2.36)$$

This imaginary part then takes its maxima $\text{Im } \Omega_+ = -1/2\tau + \delta/\sqrt{3}$ at $N = N^*$. Therefore, if the inequality condition (1.2.28) for multistability does not hold, $\text{Im } \Omega_+$ is always negative, and the solution is stable everywhere. However, if the inequality holds, and there is multistability, it follows that the maxima of the imaginary part is positive at N^* , and that in some interval the solution is unstable.

The imaginary part (1.2.36) is an increasing function in the range $N < N^*$, and decreasing for $N > N^*$. From eq. (1.2.27) it follows that $N_- < N^*$ and $N_+ > N^*$, and that $f(N_\pm) = 1/4\tau^2$. This implies that in the range $N_- < N < N_+$ we have that

$$\text{Im } \Omega_+ > -1/2\tau + \sqrt{f(N_\pm)} = 0. \quad (1.2.37)$$

Consequently the solution is unstable between N_- and N_+ .

As this instability occurs between the turning points in the $|P_0|^2$ - N dependency, we can conclude that out of the three possible population number solutions, the middle solution is always unstable, and decays to either the upper or lower population number under CW excitation [see Fig. 1.5(c)]. We are left with two stable solutions, which is commonly referred to as *optical bistability*.

It is also worthwhile to comment on the behavior of the system when the CW

pump strength is varied adiabatically. If the pump strength is initially zero, and then increased slowly, the number of polaritons in the system will be given by the lower branch, even as the pump enters the bistable region. However if it reaches, and is increased above, the end of the bistable region, the only CW solution is the continuation of the upper branch, and as such the polariton number sharply increases. When lowering the pumping power is then decreased slowly, when entering the bistable region from above, the population number now stays on the upper branch. As a principal characteristic of the bistable system we effectively have our system in two different states, as a non-Markovian property. When the pumping power is then decreased further to below the lower bound of the bistable region, the population numbers decrease to again lie on the lower branch. This hysteresis curve is what is used to identify bistable behavior in experiments.

This behavior can be understood intuitively by observing the form of equation (1.2.22). The effective frequency of the polariton mode in the prescence of a population $N = |\psi|^2$ changes as

$$\omega_p \rightarrow \omega_p + \alpha N. \quad (1.2.38)$$

This is referred to as the *blueshift* of the mode, as its energy shifts higher with increased population. The mode is also broadened by its finite lifetime. If inequality (1.2.28) is fulfilled, the pump energy lies above the broadened mode $N = 0$ energy. As N increases the energy level blueshifts, and at some point comes into resonance with the pump energy. The increased efficiency of the resonant pump increases sharply the polariton number, corresponding to the jump from the lower to the upper branch at the turning point.

The properties of the optical bistability of the polariton system recieved considerable interest following its experimental observation [57–60]. Taking into account the spin structure of the polariton the bistability phenomena can be enriched further [61, 62]. Spontaneous polarization ring formation were predicted by real-space calculations in Ref. [63], and later observed experimentally [64]. Another work investigates numerically and experimentally ballistic propagation of polariton “bullets” [65], an idea that was developed further into a ballistic polariton transistor [66, 67]. Other experiments have also demonstrated the viability of switching

Figure 1.6: Figure is from Ref. [70]. The propagation of a polariton signal along the patterned potential. A strong pulse at $t = 0$ creates a small region of high polariton density at the bottom left end of the channel, corresponding to the upper branch of polariton bistability. This upper branch density propagates along the circuit without decay of signal.

between stable polariton population states of different spin [68, 69]. A theoretical scheme of non-ballistic signal propagation instead subjects the whole system to a background pump with intensity inside the area of bistability, taking the binary polariton number, either on the upper or lower branch, as the signal value [70]. The 'branch value' is shown to propagate as a domain wall along potential channels, as shown in Fig. 1.6. As the signal is continually amplified by the background pump there is no decay. This scheme was further complemented with polaritonic designs for logic gates [71].

1.2.4 Polariton devices as terahertz emitters

The polariton system described above has been proposed as a source of THz emission. In this section these proposals are reviewed.

The upper and lower polariton branches are split by the Rabi energy $\hbar\Omega$, which is of the order of several meV. As the terahertz range lies in the energy domain in 1 to 12 meV, transitions between the upper and lower polariton branch could in principle be used to emit terahertz photons. However no radiative process can couple these states since the exciton state of the upper and lower polariton is the same. As presented in Ref. [72], and further developed in Refs. [74, 75], it is possible to envisage a different exciton state to hybridize with a polariton mode, that facilitates a radiative transition. It is proposed to design the QW such that the exciton $2X$,

consisting of a QW ground state electron and the first QW excited state of the hole, has an energy E_{2X} that lies close to the energy of the upper branch polariton $E_U = E_0 + \hbar\Omega/2$. Here the energy of the cavity mode and lowest energy exciton are taken as equal $E_C = E_{1X} = E_0$. Linear coupling between the two exciton modes can be achieved by applying an electric field across the microcavity structure. The coupling strength δ can exceed losses (condition for strong coupling) with an electric field of a few kV/cm, while being smaller than the Rabi energy $\hbar\Omega$.

The configuration of energy levels then becomes as shown in Figure 1.7(a), where $U_{\pm}$ are the energy levels that arise with the mixing of the upper polariton and the $2X$ exciton. Due to the much larger distance in energy to the lower polariton mode L , mixing is negligible between them. Now, as the higher energy states contain fractions of excitons of parity different from that of the exciton fraction in the lower polariton, radiative transitions are allowed, with THz photon energy

$$\hbar\omega_{T\pm} = E_{U\pm} - E_L = \hbar\Omega + \frac{E_{2X} - E_0 - \frac{\hbar\Omega}{2}}{2} \pm \frac{1}{2} \sqrt{\left(E_{2X} - E_0 - \frac{\hbar\Omega}{2}\right)^2 + \delta^2}. \quad (1.2.39)$$

The authors report that the calculated quantum efficiency of the THz emission reaches 1.5% for realistic parameters [72].

Another work proposes using semiconductors with a nonvanishing second-order susceptibility in order to allow the upper-to-lower polariton transition [73].

In another work it is proposed to harness transitions between $2p$ excitons and the lower $1s$ exciton-polariton branch for THz emission [76]. The $2p$ Rydberg state of the QW ground state exciton cannot be directly excited with a single photon due to selection rules, but must rather be created through a two-photon process. Radiative decay to the lower polariton branch is however allowed, and the transition lies in the THz range. The energy levels and transitions involved are shown in figure 1.7(b). This THz transition between the lower polariton branch and the $2p$ exciton has been observed experimentally [77].

In yet another work a scheme for a “bosonic cascade laser” is presented [78]. Here the energy levels down which excitations relax is not the tilted real space QW ladder of the conventional QCL, but rather eigenstates of a parabolic confinement

Figure 1.7: **(a)** Energy level configuration in the scheme described in Ref. [72]. The original energy levels are that of the cavity mode (C), the lowest energy $1S$ exciton and the excited-hole-exciton level $2X$. The cavity mode and the $1X$ exciton form upper (UP) and lower polariton (LP) energy levels, split by the Rabi energy $\hbar\Omega$. The $2X$ exciton level lies close to the upper polariton, and they are split by a smaller electric field-induced interaction δ into $U+$ and $U-$ levels. Due to the different parities of the $1X$ and $2X$ excitons radiative transitions are allowed between the $U\pm$ and LP levels, with frequency $\omega_{T\pm}$ defined in eq. (1.2.39). **(b)** Energy levels in the scheme described in Ref. [76]. The ground state is denoted as $|0\rangle$. The $2p$ exciton state must be excited by simultaneous absorption of two photons due to optical selection rules. The transition from the $2p$ state to the lower polariton (LP) with its $1s$ exciton admixture is optically allowed, with energy $\hbar\omega_T$ in the THz range.

potential of the exciton-polaritons. Eigenstates of a particle in a parabolic potential are equidistant in energy, and therefore apt for a cascade effect. By exciting the m -th eigenstate of the potential, and coupling the THz transition to a THz frequency cavity, a quantum efficiency of up to $m/2$ was calculated.

Last among the schemes of THz generation from the cavity-QW system is that of THz transitions between asymmetric intersubband polariton branches [79]. Intersubband polaritons differ from exciton-polaritons in that the cavity mode is not brought into resonance with an excitonic transition, but rather with a transition between conduction subbands [80]. By breaking wavefunction centrosymmetry by the design of the QWs, radiative transitions between intersubband polariton branches become allowed. The authors report wide tunability of the emission as a result of the ability to control intersubband transitions externally.

1.3 Dipolaritons

A different type of exciton that will play a pivotal role in the second chapter of this thesis is the *indirect exciton* (IX), where the hole and the electron are spatially separated. It was first considered theoretically in 1976, as a possible mechanism to realize Cooper pairing [81]. Most early theoretical studies continued to focus on quantum condensation phenomena, such as superfluidity and superconductivity, which is facilitated by the longer recombination time of indirect excitons as compared to that of the Wannier-Mott direct exciton [82–85]. Other theoretical works investigated such aspects as bound states in high magnetic fields [86], bound states of asymmetric number of holes and electrons [87], and spin polarization phenomena [88]. Experimental works on the subject are mainly focused on investigation of quantum collective phenomena [89–95]. Other aspects treated include experimental techniques for constructing trapping potentials [96, 97], and spectral fluctuations observed in IX photoluminescence (PL) experiments [98, 99]. Additionally, diffusive transport phenomena were investigated both theoretically [100] and experimentally [101].

The indirect exciton system is construed as two parallel quantum wells separated by a tunneling barrier. Binding energy of indirect excitons is lower than that of direct excitons, and electrons and holes are spatially separated. Their energy levels can however be tuned into resonance by applying an electric field across the structure in the direction perpendicular to the QW planes. As indirect excitons possess a dipole moment due to the spatial separation of the charges, its energy depends strongly on the electric field. The states of direct and indirect excitons are coupled together by tunneling. Holes, having much higher effective mass than electrons, tunnel across the QWs much less efficiently, and this process is generally ignored, and only the tunneling of electrons is accounted for. The real space energy diagram of the system then becomes that of the tilted ladder shown in Fig. 1.8(a), where the energy levels of electrons in the two QWs is matched.

Fig. 1.8(b) shows the exciton energy levels and their shift with the applied field as reported in Ref. [102]. The electric field is taken as $\mathbf{F} = -F\mathbf{e}_z$, where z is the growth direction of the QWs. The IX energy shift reads $\Delta E_{IX} = -Fd_i$ where

Figure 1.8: **(a)** Real space energy diagram of the double quantum wells. Direct excitons (DX) form in a single QW. The indirect exciton (IX) consists of a hole in QW1 and an electron in QW2, with an effective separation L . The energy of the DX and IX is tuned into resonance with the electric field $\vec{F} = -F\vec{e}_z$ across the growth direction of the QWs. **(b)** Figure is from Ref. [102]. Energy levels of the system shown as a function of electric field strength F . DX and IX energy levels are indicated with full and dashed black lines respectively, and the diagonalized eigenmodes of the double QW system with red lines. The upper lines represent excited excitonic modes, and are beyond the scope of this thesis. The lower DX and IX levels are the exciton modes described in this section, and are in resonance close to $F = 10^4$ V/cm. The levels anti-cross, which is evidence of level repulsion due to interaction through tunneling.

$$d_i = eL \quad (1.3.1)$$

is the IX dipole moment, with L as the effective electron-hole separation. In Ref. [102] resonance between the indirect exciton and direct exciton was measured at $F_0 = 12.5$ kV/cm, for 10 nm InGaAs QWs, separated by a 4 nm GaAs barrier. At such large fields direct excitons become polarized in the QW in the direction of the applied field, as described by the quantum-confined Stark effect. The consequent energy shift $\Delta E_{DX} = -Fd_d$ is given by the induced dipole moment

$$d_d = 24 \left(\frac{2}{3\pi} \right)^6 \frac{e^2 F m^* d^4}{\hbar^2}, \quad (1.3.2)$$

where e is the elementary charge, m^* is the effective mass, and d is the QW width. For the QW discribed above this dipole moment remains smaller than the IX dipole moment by several orders of magnitude, $d_d \approx 10^{-3}d_i$.

In the simplest case the Hamiltonian of the system can be represented in form of a 2-by-2 matrix, with terms for the free energy of the DX and IX modes, as well as the process of tunneling. This can be written in the base $\{|DX\rangle, |IX\rangle\}$ as

$$\hat{\mathcal{H}} = \hbar \begin{pmatrix} \omega_D & -J/2 \\ -J/2 & \omega_I \end{pmatrix}, \quad (1.3.3)$$

where $\hbar\omega_i$ is the energy of the direct (D) and indirect (I) excitons, and J is the rate of tunneling through the barrier. This tunneling rate depends exponentially on the barrier thickness; for barriers with thickness of several nanometers the tunneling energy $\hbar J$ is a few meV [103].

This double QW system can be placed inside a microcavity [see Fig. 1.9(a)], with the photon mode resonant with the direct excitonic transition in one the QWs. By constructing the two QWs with materials with slightly different bandgaps, or with different geometry, resonant excitation of a DX mode in the other QW can be evaded. Then the system will host three modes near resonance, with the DX linearly coupled to the cavity mode with Rabi frequency Ω , and to the IX mode with tunneling frequency J . If both frequencies are larger than losses there will be strong coupling, and the three modes give way to new linearized eigenmodes, called upper, middle and lower *dipolariton* [102–105].

The Hamiltonian describing these three modes and the couplings described above will then read

$$\hat{\mathcal{H}} = \hbar \begin{pmatrix} \omega_c & \Omega/2 & 0 \\ \Omega/2 & \omega_D & -J/2 \\ 0 & -J/2 & \omega_I \end{pmatrix}. \quad (1.3.4)$$

Using operators of second quantization this Hamiltonian can be rewritten as

$$\hat{\mathcal{H}}_0 = \hbar\omega_c \hat{a}^\dagger \hat{a} + \hbar\omega_D \hat{b}^\dagger \hat{b} + \hbar\omega_I \hat{c}^\dagger \hat{c} + \frac{\hbar\Omega}{2} (\hat{a}^\dagger \hat{b} + \hat{b}^\dagger \hat{a}) - \frac{\hbar J}{2} (\hat{b}^\dagger \hat{c} + \hat{c}^\dagger \hat{b}). \quad (1.3.5)$$

Figure 1.9: **(a)** Sketch of the dipolariton system. The optical cavity formed by two DBRs hosts the cavity mode (C), while the two QWs host the direct (DX) and indirect excitons (IX). The cavity mode and direct excitons interact through radiative absorption and emission with frequency Ω , and the direct and indirect excitons are coupled with tunneling frequency J . The energy of the indirect exciton is tuned with the applied field $\mathbf{F}$. **(b)** Figure is from Ref. [103]. Energy levels of the system as measured by photoluminescence (PL), as a function of the bias V applied across the structure, proportional to the electric field strength. Dashed lines indicate the non-interacting C, DX and IX energy levels in blue, green and red respectively. Full lines indicate the strong coupling linearized upper (UP), middle (MP) and lower (LP) dipolaritons, with dots marking peaks in the PL spectra. The colors of the full lines indicate fractions of the original modes. The inset shows the values of the tunneling interaction constant $\hbar J$ as a function of barrier width.

Here $\hat{a}^\dagger$ ($\hat{a}$), $\hat{b}^\dagger$ ($\hat{b}$) and $\hat{c}^\dagger$ ($\hat{c}$) denotes the creation (annihilation) operator of the cavity mode, direct excitons and indirect excitons. Their energies are $\hbar\omega_c$, $\hbar\omega_D$ and $\hbar\omega_I$ respectively, as is expressed by the first three terms of the Hamiltonian (1.3.5). The fourth term describe the cavity photon absorption accompanied with the formation of a direct exciton, as well as the antithetic process of exciton radiative recombination. The final term describes the electron tunneling that transmutes the exciton between the direct and indirect configuration. We again neglect spatial dynamics and thus omit wave vector indices.

1.3.1 Pulsed terahertz emission from dipolaritons

In this section a scheme for generation of pulsed THz range emission from Ref. [106] is presented. The central idea of the authors of this work is that oscillations of dipole moment coming from transitions between direct and indirect excitons induce electromagnetic waves by the laws of classical electrodynamics. The system is excited by a short laser pulse, and then allowed to decay. Excitations are found to oscillate between the DX and IX mode, with a frequency in the THz range. As in classical electromagnetics emission frequency matches the frequency of charge oscillation the emitted radiation will lie in the THz range. By using Hertz dipole equation for emitted radiation intensity the authors estimate the radiated power with the equation

$$I = N_{IX}^2 \frac{d_i^2 \omega^4}{3\pi \varepsilon_0 c^3}, \quad (1.3.6)$$

where N_{IX} is the amplitude of oscillations of indirect exciton numbers, ω is the frequency of oscillation, ε_0 is vacuum permittivity, and c is the speed of light. Using experimentally achievable values of indirect exciton density the authors claim emission powers of up to $I \approx 1.4 \mu\text{W}$. Furthermore, by varying the applied electric field, the scheme is claimed to be viable in a range of frequencies of up to 3.5 THz.

The Hamiltonian is given as that of eq. (1.3.5) with terms added to describe the excitation of the system:

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + P(t)\hat{a}^\dagger + P(t)^*\hat{a}. \quad (1.3.7)$$

The pumping expression was chosen as $P(t) = P_0 f(t) e^{i\omega_p t}$, where $|P_0|^2$ denotes the intensity of the pump, ω_p its frequency, and $f(t)$ is an envelope function $0 < f(t) < 1$. The authors use Heisenberg's equations of motion for the expectation values of the mode operators, adding decay terms to account for the finite lifetimes τ_i of the modes:

Figure 1.10: Figures are from Ref. [106]. **(a)** Energy diagram for regime I, with energies given in terms of tunneling frequency J . The cavity mode (C) is far detuned from anti-crossing of the direct (DX) and indirect exciton (IX) modes. The pump detuning $\hbar\Delta_p = \hbar\tilde{\Delta}J = 11.5$ meV puts it close to the excitonic modes. **(b)** Exciton number dynamics for one pump pulse in regime I, with electric field taken as $F = 0.95F_0$. The time profile envelope $f(t)$ of the pump is shown in shaded area, with a duration of 1 ps. The relaxation of the system is accompanied by anti-phase oscillations between the DX and IX modes. The inset shows large t oscillations, with a frequency of 1.5 THz. **(c)** Energy diagram for regime II. Here are modes of the system are close in energy. **(d)** Exciton number dynamics in regime II. Exciton density immediately post-pulse is much higher than in regime I, but decay much faster due to the more efficient coupling with the lossy cavity mode. Inset shows large t oscillations, with frequency 1.3 THz.

$$\frac{\partial\langle\hat{a}\rangle}{\partial t} = -i\omega_c\langle\hat{a}\rangle - i\frac{\Omega}{2}\langle\hat{b}\rangle - \frac{1}{2\tau_c}\langle\hat{a}\rangle - iP(t)/\hbar, \quad (1.3.8)$$

$$\frac{\partial\langle\hat{b}\rangle}{\partial t} = -i\omega_D\langle\hat{b}\rangle - i\frac{\Omega}{2}\langle\hat{a}\rangle + i\frac{J}{2}\langle\hat{c}\rangle - \frac{1}{2\tau_D}\langle\hat{b}\rangle, \quad (1.3.9)$$

$$\frac{\partial\langle\hat{c}\rangle}{\partial t} = -i\omega_I\langle\hat{c}\rangle + i\frac{J}{2}\langle\hat{b}\rangle - \frac{1}{2\tau_I}\langle\hat{c}\rangle. \quad (1.3.10)$$

The authors examine two physically different regimes separately. In regime I the cavity mode energy is chosen as lying far below the resonant excitonic modes [see Fig. 1.10(a)]. In this regime, the coupling of the cavity mode to the excitonic modes is weak, while the excitonic modes are heavily mixed. The excitations created during the pump pulse are shown to decay while oscillating between the two excitonic modes [see Fig. 1.10(b)]. Coupling to the lossy cavity mode is inefficient, so the decay time is comparatively long. In the regime II the cavity mode is brought close to the exciton resonance. Due to the more efficient coupling to the cavity mode the resultant oscillations are less regular, decay faster, but have a higher initial amplitude [see Fig. 1.10(d)].

1.4 Strong coupling and photon dressing

In section 1.2 strong interaction in the QW system with excitonic resonance was described in detail. Another widely treated strong coupling mechanism was first treated in atomic physics, arising in the quantum mechanical treatment of the interaction of atoms with a monochromatic laser field. By treating both the atom and field quantum mechanically the true eigenstates of the system can be found, an improvement on the earlier semi-classical approach [1, 3]. This “atom+photon system” is termed atom *dressed by photon*.

1.4.1 Dressed states in atomic systems

The inaugural effect described in the terms of dressed states is the Autler-Townes effect, named after its discoverers [107]. This effect describes the appearance of sidebands in atomic spectra in the presence of a laser field resonant with an atomic transition. The authors described it as a “Stark effect for rapidly varying fields” in analogy with the splitting of atomic spectra in the presence of a static electric field. In general, shifts of spectra due to non-static electric fields are today referred to as dynamic, or AC, Stark effects. The Autler-Townes effect was first observed in radio frequencies in the discoverers’ original 1955 work [107], and at optical frequencies

two decades later [108–110].

Another regime of dynamic Stark effects that has been studied is illumination with far-ultraviolet radiation, inducing transitions into the continuum. This results in modification of ionization processes, producing a strongly altered multi-peak photoelectron spectrum [111–114]. This is a result of the Gaussian time profile of the pulse of radiation that decreases the binding energy of the atom. There are therefore two times at which the energy of the emitted electron waves have the same energy, when the pulse is growing and when the pulse is decreasing. The peaks of the spectrum result where the same-energy waves interfere constructively.

Yet another incarnation of dynamic Stark effects is when the frequency of the laser is tuned away from all atomic transitions. In this case atomic spectra are shifted as by the quadratic (DC) Stark effect, with the cycle root-mean-square of the laser electric field entering as the effective field strength [115, 116]. This result can be shown straightforwardly, which serves to illustrate concepts that are important in the third chapter of this thesis. For an atom with zero inherent dipole moment and isotropic polarizability α , the Stark shift due to a static electric field of magnitude F reads

$$\Delta E = -\frac{1}{2}\alpha F^2. \quad (1.4.1)$$

To describe the interaction between the atom and an electromagnetic wave the static field should be replaced by the time-dependent field strength $\mathcal{E} \cos \omega t$. As in the case being treated the frequency ω is in resonance with no transition of the atom, only one energy level needs to be accounted for, and the Hamiltonian can be written $\mathcal{H} = \frac{1}{2}\alpha \mathcal{E}^2 \cos^2 \omega t$. The Schrödinger equation therefore reads

$$i\hbar \frac{\partial \psi(t)}{\partial t} = \frac{1}{2} \left[\alpha \mathcal{E}^2 \cos^2 \omega t \right] \psi(t), \quad (1.4.2)$$

where $\psi(t)$ is the time-dependent amplitude of the atom state. As the Hamiltonian is time-dependent, there is no single energy (frequency) solution. However, according to the Floquet theorem [117] a homogeneous and linear differential equation with

periodic coefficients admits a solution of the form

$$\psi(t) = e^{-iEt/\hbar}\phi(t), \quad (1.4.3)$$

where $\phi(t)$ is periodic with period $2\pi/\omega$. This is analogous to the Bloch theorem, where periodicity in time has replaced the spatial periodicity of the solid state lattice. As such E , the analog of the Bloch quasimomentum, is termed the quasienergy of the dressed state.

In the case of the simple one level atom the solution can be calculated directly from the formal solution $\psi(t) = \exp\left\{-i\int_0^t \mathcal{H}(s)ds\right\}$ of the Schrödinger equation. Integrating the cosine squared yields the solution

$$\psi(t) = \exp\left[-i\frac{\alpha\mathcal{E}^2}{4\hbar}t\right] \exp\left[-i\frac{\alpha\mathcal{E}^2}{8\hbar\omega}\sin 2\omega t\right]. \quad (1.4.4)$$

By observing that the second factor is $2\pi/\omega$ periodic, and comparing with the form of the Floquet solution (1.4.3), the quasienergy of the atom state dressed by the non-resonant field can be identified as

$$E = \frac{1}{4}\alpha\mathcal{E}^2. \quad (1.4.5)$$

As the root-mean-square of the sinusoidal field is $\mathcal{E}/\sqrt{2}$, this result can indeed be found from the DC Stark shift (1.4.1) simply by substituting the static field strength F with the electromagnetic wave RMS.

Another topic of illumination of atomic systems with strong coherent radiation is that of the laser-assisted electron scattering (LAES) [118, 119]. When the interaction between the incident field and the atom can be assumed small, and the photon energy is small compared to the electron energy, the scattering of the electron with the atom is adequately described by the perturbative theory developed by Kroll and Watson [120]. The main prediction of this theory is that for large field strengths, electron-atom scattering processes where multiple photons are absorbed or emitted become prominent. The fingerprint of these processes were observed in experimental scattering spectra soon after their prediction [121]. Going beyond

this theory by increasing the interaction strength between the laser light and the matter system necessitates crossing to the dressed-atom picture. The first works advancing this theory treated the interaction between the field and the scattered electron nonperturbatively, while treating the field-atom interaction with first order time-dependent perturbation theory [122–125]. The semi-perturbative treatment restricts the electric field strength as $\mathcal{E} \ll e/4\pi\epsilon_0 a_0^2 \cong 5 \times 10^9$ V/cm, the atomic unit of electric field strength. Eventually Floquet-class techniques were utilized to obtain non-perturbative solutions [126, 127].

One result noted in most of the works in the semi-perturbative approach [122–124], and confirmed by the Floquet calculations [127], is the appearance of a zero-angle peak in the differential scattering cross section. This effect is associated with the process of absorption or emission of one photon by the scattered electron. Very recent works of the group of Yamanouchi [128, 129] have been the first to directly observe the effect of the dressed atom in experiments, including measuring the zero-scattering angle peak in the cross section. Until then most experiment had been adequately described by the Kroll-Watson theory (see e.g. the recent works [130, 131]).

1.4.2 Dressed states in condensed matter systems

The modification of the physical properties of atoms under intense illumination has analogies also in the physics of condensed matter. In fact, exciton-polaritons, as described in section 1.2, are the result of the resonant interaction between an electromagnetic wave and CW excitons. A more direct analogy with the Autler-Townes effect is found when the exciton density exceeds the limit (1.2.12), implying $n_{exc} > 1/a_B^2$, in which case the individual electrons and holes behave as fermions [132].

Three early experimental works reported shifts of exciton energy levels in GaAs quantum wells in the presence of strong non-resonant laser light [133–135], in full analogy with the case of the non-resonantly illuminated atom. The authors of both works emphasize that as the illumination is not resonant with the exciton transition there is no absorption. As such the level shifts are not explained by exciton

populations, but should be interpreted in terms of a dynamic Stark effect.

When it comes to dynamic properties of the dressed condensed matter system, most attention has been put in describing non-stationary effects due to alternating current (AC) potentials [136], such as tunneling through potential barriers [137] and scattering of quantum pumps [138]. Another pair of works described persistent dissipationless currents arising in dressed states in nanostructures with broken time-reversal symmetry [139,140]. Stationary direct current (DC) conductivity properties of the dressed condensed matter system however remained largely unexplored until Refs. [141,142]. In these works a two-dimensional electron gas (2DEG) with dispersion relation $E(k) = \hbar^2 k^2 / 2m$ is conceived as illuminated by a strong monochromatic electromagnetic wave propagating perpendicularly to the 2DEG plane. The spatial profile of the field is assumed homogenous, and its frequency ω far from any band transitions. The Schrödinger equation of such a system reads

$$i\hbar \frac{\partial \psi(t)}{\partial t} = \frac{1}{2m} \left[\hat{\mathbf{p}} - \frac{e}{c} \mathbf{A}(t) \right]^2 \psi(t), \quad (1.4.6)$$

where

$$\mathbf{A}(t) = \frac{c\mathcal{E}_0}{\omega} \cos(\omega t), \quad (1.4.7)$$

is the vector potential of the linearly polarized laser field, and $\mathcal{E}_0$ the corresponding electric field strength.

This Schrödinger equation is very similar to the equation (1.4.2) for an atom illuminated by a non-resonant field. However, as the Hamiltonian also contains the momentum operator the solution cannot be calculated straight from the formal solution to the Schrödinger equation. The corresponding equation was first solved by Volkov for the relativistic case of Dirac electrons [143]. The solution even to the non-relativistic equation (1.4.6) is therefore commonly referred to as a Volkov wavefunction, and arises also for example in the study of laser-assisted electron scattering through the interaction between the scattered electron and the dressing

field (see e.g. [119, 122]). The solution reads

$$\psi_{\mathbf{k}}(t) = \exp \left\{ -\frac{i}{\hbar} \left(\left[E(k) - \frac{e^2 \mathcal{E}_0^2}{4m\omega^2} \right] t - \frac{e\hbar \mathcal{E}_0 \cdot \mathbf{k}}{m\omega^2} \sin(\omega t) + \frac{e^2 \mathcal{E}_0^2}{8m\omega^3} \sin(2\omega t) \right) \right\} \quad (1.4.8)$$

As the Hamiltonian is time-dependent and periodic, this solution should be interpreted in the context of Floquet theory. Indeed the last two terms in the complex exponential are periodic, with period $2\pi/\omega$. This allows the expression inside the square brackets to be interpreted as the quasienergy dispersion of the dressed 2DEG. The dynamics Stark shift is independent of the wavevector, and is the same as for dressing by non-resonant field of an atom, with the polarizability taken as $\alpha = e^2/m\omega^2$.

In Ref. [141] the Volkov wavefunction was used to derive scattering matrix elements $w_{\mathbf{k}'\mathbf{k}}$, defined as the probability per unit time of an electron in state $\psi_{\mathbf{k}}$ to scatter to state $\psi_{\mathbf{k}'}$, in the presence of a weak disorder potential $U(\mathbf{r})$. The scattering rates were found in first order perturbation theory as

$$w_{\mathbf{k}'\mathbf{k}} = \frac{2\pi}{\hbar} J_0 \left(\frac{e\mathcal{E}_0 \cdot (\mathbf{k} - \mathbf{k}')}{m\omega^2} \right)^2 |U_{\mathbf{k}'\mathbf{k}}|^2 \delta(E(k') - E(k)), \quad (1.4.9)$$

where J_0 is the zeroth-order Bessel function of the first kind, and $U_{\mathbf{k}'\mathbf{k}}$ is the Fourier space representation of the disorder potential. The author emphasizes as a central result of the work [141] that backscattering ($\mathbf{k}' = -\mathbf{k}$) can be suppressed by choosing the electric field strength $\mathcal{E}_0$ such that the Bessel function equals zero [see Fig. 1.11(a)]. As such the laser intensity can be used to control the transport properties of the 2DEG.

In Ref. [142] the above result is investigated further by calculating stationary DC conductivity of the dressed 2DEG using the Boltzmann equation. Calculations are performed separately for linear and circular polarization of the laser light, and, for linear polarization, for the cases where bias is applied in the direction parallel or perpendicular to the polarization direction. The authors find that the decreased backscattering leads to an increase in the conductivity, with an oscillatory behavior that is especially apparent for circular polarization [see Fig. 1.11(b)].

Figure 1.11: **(a)** Figure in from Ref. [141]. Backscattering parameter $W = w_{-\mathbf{k}\mathbf{k}}/w_{-\mathbf{k}\mathbf{k}}^{(0)}$ is calculated as a function of electric field strength $\mathcal{E}_0$ of the dressing field. Here $w_{\mathbf{k}'\mathbf{k}}^{(0)} = 2\pi|U_{\mathbf{k}'\mathbf{k}}|^2\delta(E(k') - E(k))/\hbar$ is the scattering matrix element in the absence of the dressing field. **(b)** Figure is from Ref. [142]. Conductivity as calculated with the Boltzmann equation as a function of irradiation intensity $I = c\varepsilon_0|\mathcal{E}_0|^2/2$. Red, full (blue, dashed) line indicates conductivity in the direction parallel (perpendicular) to the polarization of a linearly polarized dressing field. Green, dashed-dotted line indicates the (isotropic) conductivity in the case of circularly polarized dressing field.

A specific case of condensed matter that is of special importance is graphene, a 2D hexagonal lattice of carbon atoms. It possesses a Dirac-like linear low-energy dispersion relation, and consequently its density of states is linear in energy (and wavevector), and disappears at zero chemical potential [144, 145]. This characterizes graphene as a semimetal. Investigations into the effect of dressing by a laser field have largely concentrated on the band structure of the laser-dressed graphene. Several works have found a gap opening in the dispersion at zero wavevector, inducing an insulator transition [146–149]. In Ref. [149] this is accomplished by making an ansatz for the graphene wavefunction in the form of an expansion of the periodic part of the Floquet wavefunction. Faster oscillating terms that arise are then discarded to obtain an approximal solution.

Other works have found gaps appearing in the dispersion where the energy difference between the valence and conduction band equals the photon energy, in analogy

with the atomic Autler-Townes effect [150–152]. Other effects investigated in the dressed graphene system includes the DC Hall effect [147], optical response of the dressed electron [153], Klein tunneling through potential barriers [154], and the appearance of chiral edge states characteristic of topological insulators [155].

2. Terahertz range emission from dipolariton system

This chapter describes research and results published in the works [A1] and [A2]. In section 1.3.1 a scheme from Ref. [106] describing generation of pulsed emission of radiation in the terahertz range based on the dipolariton system was presented. It is however known that there can arise in systems with nonlinearity in two coupled modes the regime of *parametric instability*, with continuous oscillations between modes under continuous wave (CW) excitation [156]. In the first part of this chapter nonlinear effects are taken into account in analogy with section 1.2.3, where nonlinear effects in the simpler exciton-polariton system were explored. Such elegant analytical results as in polariton system do not arise in the dipolariton system, but conditions for parametric oscillations are explored numerically, and it is shown that continuous THz range oscillations between the direct and indirect exciton modes can be induced. In accordance with the classical argument in Ref. [106] this can be argued to produce THz emission, which is shown to be tunable in the mid- to upper-THz range.

In the second part of this chapter, the dipolariton is imagined to be enclosed in a THz cavity. Interaction between the quantized THz mode and the dipolariton system is investigated, and shown to enhance the emission.

2.1 Nonlinear interaction in dipolariton system

As in this section we describe the same physical system as in section 1.3, only additionally accounting for nonlinearities, the sketch in Fig. 1.9(a) continues to visualize our system.

2.1.1 Model

The Hamiltonian of the dipolariton system is that given in eq. (1.3.7), with the addition of nonlinear terms representing the Coulomb interaction between excitons. As such we can write the Hamiltonian as

$$\begin{aligned} \hat{\mathcal{H}}_0 = & \hbar\omega_c\hat{a}^\dagger\hat{a} + \hbar\omega_D\hat{b}^\dagger\hat{b} + \hbar\omega_I\hat{c}^\dagger\hat{c} + \frac{\hbar\Omega}{2}(\hat{a}^\dagger\hat{b} + \hat{b}^\dagger\hat{a}) - \frac{\hbar J}{2}(\hat{b}^\dagger\hat{c} + \hat{c}^\dagger\hat{b}) \\ & + \frac{V_{DD}}{2}\hat{b}^\dagger\hat{b}^\dagger\hat{b}\hat{b} + \frac{V_{II}}{2}\hat{c}^\dagger\hat{c}^\dagger\hat{c}\hat{c} + V_{DI}\hat{b}^\dagger\hat{c}^\dagger\hat{b}\hat{c} + P(t)\hat{a}^\dagger + P(t)^*\hat{a}, \end{aligned} \quad (2.1.1)$$

where $\hat{a}^\dagger(\hat{a})$, $\hat{b}^\dagger(\hat{b})$ and $\hat{c}^\dagger(\hat{c})$ are the creation (annihilation) operators of the cavity mode (C), direct exciton (DX) and indirect exciton (IX) respectively. Furthermore $\hbar\omega_c$, $\hbar\omega_D$ and $\hbar\omega_I$ are the energies of the C, DX and IX modes, $\hbar\Omega$ is the C-DX Rabi splitting, $\hbar J$ is the the DX-IX tunneling strength, and V_{DD} , V_{II} and V_{DI} are the matrix elements of the interaction between two direct excitons, two indirect excitons, and between a direct and an indirect exciton respectively. CW pumping is represented by $P(t) = P_0 e^{-i\omega_p t}$, where $|P_0|^2$ is the pumping intensity and ω_p the frequency of the pump.

The terms that are new in (2.1.1) are the first three terms of the second line. The first term of the second line represents the interaction between two direct excitons, and is therefore the same as the excitonic nonlinear term in section 1.2.3. We can therefore identify the interaction constant as $V_{DD} = 6E_b a_B^2 / S$ [56], taking the exciton binding energy as $E_b = 8$ meV and Bohr radius as $a_B = 10$ nm, and the system area as $S = 100 \mu\text{m}^2$. The second term describes the interaction between two indirect excitons. The interaction constant V_{II} was taken as found in Ref. [157], with the electron-hole separation taken as $L = 12$ nm. The third term describes

the interaction between one direct and one indirect exciton. The calculation of the interaction constant is presented in Appendix B. Additionally the Rabi coupling and tunneling strength were chosen as $\hbar\Omega = \hbar J = 6$ meV [103].

To investigate the properties of the system dynamic equations for the particle operators are found from Heisenberg's equation $d\hat{o}/dt = -i[\hat{o}, \hat{\mathcal{H}}_0]/\hbar$, $\hat{o} = \hat{a}, \hat{b}, \hat{c}$. As in the context of this work population numbers are large and correlation effects negligible, expectation values are taken of the equations of motion, and the mean field approximation $\langle \hat{o}_i \hat{o}_j \rangle \approx \langle \hat{o}_i \rangle \langle \hat{o}_j \rangle$ used to decompose expectation values of operator products. As an illustration, the equation of motion of the expectation value of operator $\hat{b}$ of the direct exciton is found as follow.

The operator $\hat{b}$ commutes with the operators of the other modes,

$$[\hat{b}, \hat{a}] = [\hat{b}, \hat{a}^\dagger] = [\hat{b}, \hat{c}] = [\hat{b}, \hat{c}^\dagger] = 0. \quad (2.1.2)$$

Furthermore, as a bosonic operator, the intramode commutation relations reads

$$[\hat{b}, \hat{b}^\dagger] = 1, \quad (2.1.3)$$

and lastly by definition $[\hat{b}, \hat{b}] = 0$. The DX-DX and DX-IX nonlinear terms yield the commutators

$$[\hat{b}, \hat{b}^\dagger \hat{b}^\dagger \hat{b} \hat{b}] = [\hat{b}, \hat{b}^\dagger \hat{b}^\dagger] \hat{b} \hat{b} = \hat{b}^\dagger [\hat{b}, \hat{b}^\dagger] \hat{b} \hat{b} + [\hat{b}, \hat{b}^\dagger] \hat{b}^\dagger \hat{b} \hat{b} = 2\hat{b}^\dagger \hat{b} \hat{b} \quad (2.1.4)$$

and

$$[\hat{b}, \hat{b}^\dagger \hat{c}^\dagger \hat{b} \hat{c}] = [\hat{b}, \hat{b}^\dagger] \hat{c}^\dagger \hat{b} \hat{c} = \hat{c}^\dagger \hat{b} \hat{c}. \quad (2.1.5)$$

The Heisenberg equation for the operator is

$$\frac{d\hat{b}}{dt} = -i\omega_D \hat{b} - i\frac{\Omega}{2} \hat{a} + i\frac{J}{2} \hat{c} - i\frac{V_{DD}}{\hbar} \hat{b}^\dagger \hat{b} \hat{b} - i\frac{V_{DI}}{\hbar} \hat{c}^\dagger \hat{b} \hat{c} \quad (2.1.6)$$

It is convenient to perform the change of variables $\hat{b} \rightarrow e^{-i\omega_c t} \hat{b}$, as this removes the reference to absolute mode energies, and allows speaking only about the detunings $\delta_\Omega = \omega_c - \omega_D$ and $\delta_J = \omega_I - \omega_D$ between modes.

Next the expectation value is taken of both sides of eq. (2.1.6), and the mean field approximation used to decompose the nonlinear terms. Lastly the finite lifetime of the mode is accounted for by addition of term $\partial\langle\hat{b}\rangle/\partial t|_{decay} = -\langle\hat{b}\rangle/2\tau_D$, where τ_D is the lifetime of a direct exciton. This yields eq. (2.1.8).

Performing this procedure for all three modes yields the system of coupled equations

$$\frac{\partial\langle\hat{a}\rangle}{\partial t} = -i\frac{\Omega}{2}\langle\hat{b}\rangle - \frac{1}{2\tau_c}\langle\hat{a}\rangle - i\tilde{P}(t), \quad (2.1.7)$$

$$\frac{\partial\langle\hat{b}\rangle}{\partial t} = -i\left[-\delta_\Omega + \frac{V_{DD}}{\hbar}|\langle\hat{b}\rangle|^2 + \frac{V_{DI}}{\hbar}|\langle\hat{c}\rangle|^2\right]\langle\hat{b}\rangle - i\frac{\Omega}{2}\langle\hat{a}\rangle + i\frac{J}{2}\langle\hat{c}\rangle - \frac{1}{2\tau_D}\langle\hat{b}\rangle, \quad (2.1.8)$$

$$\frac{\partial\langle\hat{c}\rangle}{\partial t} = -i\left[\delta_J - \delta_\Omega + \frac{V_{DI}}{\hbar}|\langle\hat{b}\rangle|^2 + \frac{V_{II}}{\hbar}|\langle\hat{c}\rangle|^2\right]\langle\hat{c}\rangle + i\frac{J}{2}\langle\hat{b}\rangle - \frac{1}{2\tau_I}\langle\hat{c}\rangle. \quad (2.1.9)$$

Here the pumping term is modified by the change of variables as $\tilde{P}(t) = e^{i\omega_c t}P(t)/\hbar$. For CW excitation it reads $\tilde{P} = P_0 e^{-i\Delta_p t}$ where $\tilde{P}_0 = P_0/\hbar$ and $\Delta_p = \omega_p - \omega_c$ is the pump detuning. Decay terms were introduced for the cavity and indirect exciton modes in the same way as for the direct exciton. Lifetimes of the modes were chosen as $\tau_c = 5$ ps, $\tau_D = 1$ ns, and $\tau_I = 100$ ns.

2.1.2 Steady-state calculation and stability analysis

The coupled equations (2.1.7-2.1.9) cannot be solved analytically. Nevertheless, to analyze the CW excitation steady-state solution to the equations we proceed as for the exciton-polariton system in section 1.2.3. We make the ansatz of rotating wave solutions with the frequency of the driving field for all three modes:

$$\langle\hat{o}_i\rangle = e^{-i\Delta_p t}\psi_i, \quad i = c, D, I \quad (2.1.10)$$

As time-dependence of the modes and the pumping term is the same it can then be eliminated from the equations, and we have only coupled algebraic equations in the order parameter ψ_i^0 , that gives the population numbers $N_i = |\psi_i|^2$ of the modes

$i = c, D, I$. The resulting equations read

$$\Delta_p \psi_c = \frac{\Omega}{2} \psi_D - \frac{i}{2\tau_c} \psi_c + \tilde{P}_0, \quad (2.1.11)$$

$$\Delta_p \psi_D = \left[-\delta_\Omega + \frac{V_{DD}}{\hbar} |\psi_D|^2 + \frac{V_{DI}}{\hbar} |\psi_I|^2 \right] \psi_D + \frac{\Omega}{2} \psi_c - \frac{J}{2} \psi_I - \frac{i}{2\tau_D} \psi_D, \quad (2.1.12)$$

$$\Delta_p \psi_I = \left[\delta_J - \delta_\Omega + \frac{V_{DI}}{\hbar} |\psi_D|^2 + \frac{V_{II}}{\hbar} |\psi_I|^2 \right] \psi_I - \frac{J}{2} \psi_D - \frac{i}{2\tau_I} \psi_I. \quad (2.1.13)$$

Now, as the equations are linear in ψ_c , we can eliminate the variable between the first and the second equation. From eq. (2.1.11):

$$\psi_c = \left(\Delta_p + \frac{i}{2\tau_c} \right)^{-1} \left(\frac{\Omega}{2} \psi_D + \tilde{P}_0 \right) = \frac{\Delta_p - \frac{i}{2\tau_c}}{\Delta_p^2 + \frac{1}{4\tau_c^2}} \left(\frac{\Omega}{2} \psi_D + \tilde{P}_0 \right). \quad (2.1.14)$$

We denote the modified direct exciton energy and decay rates, and the effective DX pumping term as

$$E'_D = -\hbar\delta_\Omega + \frac{\Omega^2/4}{\Delta_p^2 + \frac{1}{4\tau_c^2}} \Delta_p \quad (2.1.15)$$

$$\gamma'_D = \frac{\hbar}{2\tau_D} + \frac{\Omega^2/4}{\Delta_p^2 + \frac{1}{4\tau_c^2}} \frac{\hbar}{2\tau_c}, \quad (2.1.16)$$

$$P'_0 = \frac{\Omega/2}{\sqrt{\Delta_p^2 + \frac{1}{4\tau_c^2}}} e^{-i\phi} P_0, \quad (2.1.17)$$

where $\phi = \arctan(1/2\tau_c\Delta_p)$. The pumping term acquires a phase factor, but as this can be interpreted as simply shifting time by ϕ/ω_d it does not change the steady state solutions. As such this phase factor can be ignored.

We proceed by writing $E_I = \hbar\delta_J - \hbar\delta_\Omega$ and $\gamma_I = \hbar/2\tau_I$. This allows writing the resulting equations in the more elegant form

$$0 = \left[E'_D - \hbar\Delta_p - i\gamma'_D + V_{DD}|\psi_D|^2 + V_{DI}|\psi_I|^2 \right] \psi_D - \frac{\hbar J}{2} \psi_I + P'_0, \quad (2.1.18)$$

$$0 = \left[E_I - \hbar\Delta_p - i\gamma_I + V_{DI}|\psi_D|^2 + V_{II}|\psi_I|^2 \right] \psi_I - \frac{\hbar J}{2} \psi_D. \quad (2.1.19)$$

Equations (2.1.18), (2.1.19) are coupled third-order algebraic equation, that admit the complex solutions ψ_D , ψ_I . They cannot be solved in closed form, and were solved numerically. When the solutions have been found for each value of the pumping intensity P_0 , the stability of the solution should be investigated. As in the case of the single exciton-polariton mode, we add to our earlier ansatz a small time-dependent deviation:

$$\langle \hat{o}_i \rangle = (\psi_i + \delta\psi_i(t))e^{-i\Delta_p t}, \quad i = D, I, \quad (2.1.20)$$

where $\delta\psi_i(t) \ll 1$. To investigate the dynamics of the small deviation we recast equations (2.1.18), (2.1.19) as the dynamic equations

$$i\hbar \frac{\partial \psi_D}{\partial t} = \left[E'_D - i\gamma'_D + V_{DD}|\psi_D|^2 + V_{DI}|\psi_I|^2 \right] \psi_D - \frac{\hbar J}{2} \psi_I + P'_0, \quad (2.1.21)$$

$$i\hbar \frac{\partial \psi_I}{\partial t} = \left[E_I - i\gamma_I + V_{DI}|\psi_D|^2 + V_{II}|\psi_I|^2 \right] \psi_I - \frac{\hbar J}{2} \psi_D. \quad (2.1.22)$$

Upon inserting ansatz (2.1.20) into the above equations, and neglecting all terms of second or higher order in $\delta\psi_i$, and cancelling the rotating-wave phase factor, we get the dynamic equations for the deviation

$$\begin{aligned} i\hbar \partial_t \delta\psi_D = & \left[E'_D - \hbar\Delta_p - i\gamma'_D + 2V_{DD}|\psi_D|^2 + V_{DI}|\psi_I|^2 \right] \delta\psi_D + V_{DD}\psi_D^2 \delta\psi_D^* \\ & + V_{DI}\psi_D\psi_I \delta\psi_I^* + \left[V_{DI}\psi_I^*\psi_D - \frac{\hbar J}{2} \right] \delta\psi_I, \end{aligned} \quad (2.1.23)$$

$$\begin{aligned} i\hbar \partial_t \delta\psi_I = & \left[E_I - \hbar\Delta_p - i\gamma_2 + V_{DI}|\psi_D|^2 + 2V_{II}|\psi_I|^2 \right] \delta\psi_I + V_{II}\psi_I^2 \delta\psi_I^* \\ & + V_{DI}\psi_I\psi_D \delta\psi_D^* + \left[V_{DI}\psi_D^*\psi_I - \frac{\hbar J}{2} \right] \delta\psi_D. \end{aligned} \quad (2.1.24)$$

We now give the time-dependence of the deviation an explicit form $\delta\psi_i = u_i e^{-iEt/\hbar} + v_i e^{iE^*t/\hbar}$. Inserting into the above equations yields an eigenvalue problem

for $\delta\Psi := (u_D, v_D, u_I, v_I)$, $\mathcal{M}\delta\Psi = E\delta\Psi$, where

$$\mathcal{M} = \begin{pmatrix} E'_D - \hbar\Delta_p - i\gamma'_D & +2V_{DD}|\psi_D|^2 & V_{DD}\psi_D^2 & V_{DI}\psi_D\psi_I^* - \frac{\hbar J}{2} & V_{DI}\psi_D\psi_I \\ & +V_{DI}|\psi_I|^2 & & & \\ & & -E'_D + \hbar\Delta_p - i\gamma'_D & & \\ -V_{DD}(\psi_D^*)^2 & -2V_{DD}|\psi_D|^2 & -V_{DI}\psi_D^*\psi_I^* & -V_{DI}\psi_D^*\psi_I + \frac{\hbar J}{2} & \\ & -V_{DI}|\psi_I|^2 & & & \\ & & E_I - \hbar\Delta_p - i\gamma_I & & \\ V_{DI}\psi_D^*\psi_I - \frac{\hbar J}{2} & V_{DI}\psi_D\psi_I & +V_{DI}|\psi_D|^2 & V_{II}\psi_I^2 & \\ & & +2V_{II}|\psi_I|^2 & & \\ & & & -E_I + \hbar\Delta_p - i\gamma_I & \\ -V_{DI}\psi_D^*\psi_I^* & -V_{DI}\psi_D\psi_I^* + \frac{\hbar J}{2} & -V_{II}(\psi_I^*)^2 & -V_{DI}|\psi_D|^2 & \\ & & & -2V_{II}|\psi_I|^2 & \end{pmatrix} \quad (2.1.25)$$

For each stationary state ψ_D, ψ_I , as found from eqs. (2.1.18),(2.1.19), the matrix above can be solved numerically for four eigenvalues E_α . Due to the form of the deviation, we see that it decays in time if all imaginary parts are negative, $\text{Im}(E_\alpha) < 0 \forall \alpha$. This implies fluctuations around the CW stationary value of the order parameters die out, and the solution is stable. If however the imaginary part of one eigenvalue is non-negative, $\text{Im}(E_\alpha) \geq 0$, the solution is not stable. If additionally $\text{Re}(E_\alpha) > 0$ then the solution is parametrically unstable [156].

2.1.3 Steady-state solutions

In calculating the dependence of the population numbers on the pumping strength, and the stability of the solutions, mode detunings were chosen as $\hbar\delta_\Omega = -3$ meV and $\hbar\delta_J = 1$ meV, the latter corresponding to the value $F = 0.945F_0$ of the applied electric field, where $F_0 = 12.5$ kV/cm is the field strength corresponding to the resonance of the direct and indirect exciton modes [102]. In Fig. 2.1(a) a phase diagram of the pumping intensity is constructed as a function of the pumping detuning $\hbar\Delta_p$

Figure 2.1: **a** Phase diagram of pump intensity as a function of pump detuning $\hbar\Delta_p$ on the horizontal axis and indirect exciton number $N_I = |\psi_I|^2$ on the vertical axis. Lighter colors indicate higher pumping strength needed to reach the indicated IX population. White area in top left corner cannot be reached for any pumping strength. Areas enclosed in black lines indicate parametric instability (PI) of the solution. The two dark bands, indicating low necessary pumping power, are due to resonance with the middle and upper polariton modes [see energy diagram in (b)]. These resonances start at 2 meV (MP) and 7 meV (UP), and blue-shift with increased exciton population due to the nonlinear terms in the Hamiltonian. The white line at $\hbar\Delta_p = 4.5$ meV indicates the pumping detuning used in the results in next section. The red crosses indicate two different solutions for N_I for the same pumping power. The lower solution is stable, and the higher solution is parametrically unstable. **b** Energy diagram of the dipolariton system as a function of the applied field. Dashed lines indicate the cavity (C, green), direct exciton (DX, blue), and indirect exciton (IX, red) modes, and full lines the lower (LP), middle (MP) and upper polariton (UP). The vertical line indicates the chosen value of the applied field $F = 0.945 F_0$. The short horizontal lines and the roman numerals indicate the pump detunings used to construct the multistability diagrams in Fig. 2.2.

and indirect exciton population $N_I = |\psi_I|^2$. It should be interpreted in the sense of how strong pumping is required to reach the desired IX number for a given detuning of the pump. As such it highlights the resonances of our system with the diagonal-

ized dipolariton modes with lower pumping power. Additionally it shows that for a given pump detuning the indirect exciton number has an upper bound.

Next pump detuning were chosen in accordance with the energy diagram in Fig. 2.1(b) such that the pump energy lies slightly above the lower (i), middle (ii) and upper (iii,iv) polariton. Each pump detuning was used to construct a multistability curve in the pump power–occupation number plane, shown in Fig. 2.2. The multistability curve of the direct exciton mode is superficially similar to the exciton-polariton bistability curve. However, where the exciton-polariton is unstable, and decays to either the upper or lower branch, the DX middle branch is parametrically unstable, indicating the system oscillates. The indirect exciton multistability curve however decreases when pumping power is increased past the range of multistability, confirming the upper bound behavior seen in the phase diagram.

2.1.4 Dynamic analysis

As the indirect exciton fraction of the lower polariton is low, and IX numbers consequently low in the LP multistability, it is unsuitable for producing large oscillations of dipole moment. Further the upper polariton multistability is complicated by the fact that the middle polariton multistability extends down to zero pump power. As such the pump detuning is chosen as $\hbar\Delta_p = 4.5$ meV, slightly above the upper polariton energy. Pump intensity is chosen as $|\tilde{P}_0|^2 = 1.1 \times 10^{29} \text{ s}^{-2}$, as illustrated by a vertical arrow in the MP multistability curve in Fig. 2.2(ii). The equations of motion (2.1.7)-(2.1.9) are then solved numerically, with the CW pump turning on linearly [see Fig. 2.3]. Provided the switching is slow enough (>5 ps), the occupation numbers remain on the lower stable branch, with $N_I \approx 2500$ and $N_D \ll N_I$, as indicated by the lower cross in Fig. 2.1(a) and the lower branch in Fig. 2.2(ii). By then applying an additional pulse of pumping incoherent oscillations appear in the system. After approximately 0.5 ns these oscillations have regularized, and become harmonic, corresponding to the parametric instability of the middle branch of multistability. For the chosen parameters the indirect exciton occupation oscillates between 0.1×10^4 and 2.6×10^4 , and have a frequency of 1.75 THz.

If the pumping power however is turned on quickly (<2 ps), the system does

Figure 2.2: Multistability curves for occupation numbers as a function of CW pumping power. Blue curves correspond to direct exciton steady-state numbers, and red curves to indirect exciton numbers. Stable solutions are indicated by thinner darker lines, while parametrically unstable solutions by thicker, lighter lines. **(i)** Pump detuning $\hbar\Delta_p = -1$ meV is slightly above the LP mode [as indicated in Fig. 2.1(b)]. The multistability is due to the blueshift of the LP mode. IX numbers are rather low, reflecting the low IX fraction in the LP mode. **(ii)** Pump detuning $\hbar\Delta_p = 4.5$ meV is slightly above the MP mode, and the resulting multistability is due to blueshift of this mode. Due to larger IX populations this regime is apt to induce large oscillations of dipole moment in the parametrically unstable range, and is used in the following section. The pumping power chosen for dynamic analysis is indicated by the vertical arrow. **(iii,iv)** Pump detuning $\hbar\Delta_p = 8.5$ meV is slightly above the UP mode energy. The multistability shown in the insets is due to the blueshift of the UP mode. The main plots however, going to much higher pumping strengths, show multistability due to the blueshift of the MP mode. Due to the complex behaviour at small pumping powers this regime is not appropriate to facilitate the scheme of this work.

not remain stable on the lower branch of multistability, but becomes parametrically unstable. This is in contrast to the bistability behavior of exciton-polaritons, where

Figure 2.3: Dynamic analysis of the driven nonlinear dipolariton system. Pump detuning 4.5 meV is slightly above the middle polariton energy, and CW pump strength $|\vec{P}_0|^2 = 1.1 \times 10^{29} \text{ s}^{-2}$ is inside the multistability range. The pump is turned on linearly from $t = 0$ with a long turning on time. Consequently at $t = 1000$ ps the system remains on the lower stable branch, with $N_I \approx 2500$ and $N_D \ll N_I$. A short (1 ps) additional pump pulse of the same intensity as the CW pump is applied, inducing oscillations in the occupancies. At $t = 1500$ ps the system has settled into harmonic oscillations between the direct and indirect exciton occupancies.

it passes between branches only when the pump power passes beyond the range of bistability.

The energy level configuration can be controlled by varying the applied field strength F . This shifts the detuning δ_J between the direct and indirect exciton. This alters the frequency of oscillations displayed in the system, giving a measure of control over the emission. The dependence of the frequency of oscillation on the applied field is shown in Fig. 2.4(a). We find a range of possible frequencies of emission from 1.5 to 2.2 THz. If the field strength is decreased the limit $0.7F_0$ of the figure the time for oscillations to settle into harmonicity increases rapidly, limiting the usefulness of this limit. When the the field however is increased past the upper limit $1.1F_0$, oscillations become anharmonic, as shown in Fig. 2.4(b).

Figure 2.4: **(a)** Dependence of frequency of oscillations on the applied electric field strength in units of the resonance field $F_0 = 12.5$ kV/cm. Cavity mode detuning is $\hbar\delta_\Omega = -3$ meV and pump detuning $\hbar\Delta_p = 4.5$ meV. Changing the applied field shifts the exciton energy levels. We find the oscillations being controllable from 1.5 to 2.2 THz. **(b)** Dynamic analysis of the IX occupation numbers for electric field $F = 1.2F_0$. In this range of the applied field strength the long-term oscillations are not harmonic, and therefore not suitable for generation of emission.

2.1.5 Emission power

As discussed in section 1.3, each indirect exciton possesses a dipole moment $d_i = eL$, where L is the electron-hole separation. The applied field-induced dipole moment of the direct exciton is much lower $d_d \ll d_i$, and can be neglected in the following. The total z-direction dipole moment of the system is then simply the $D_z(t) = N_I(t)d_i$. In the case of parametric instability the population numbers N_I oscillate indefinitely under CW pumping, and can be written

$$D_z(t) = N_I^0 d_i \frac{\cos(\omega_T t) + 1}{2}, \quad (2.1.26)$$

where ω_T is the angular frequency of oscillation, and N_I^0 is the amplitude of oscillation. This illustrates how oscillations in the indirect exciton occupancy directly corresponds to oscillations in the z-direction dipole moment. Each IX-DX pair acts as a classical dipole, with the electron going back and forth between the quantum wells.

To estimate the emitted power, it is straightforward to use the classical equation for emission from an oscillating dipole antenna. The far-field radiation intensity then should be [158]

$$I_c = \frac{\langle \ddot{D}_z^2 \rangle}{6\pi\epsilon_0 c^3} = \frac{(N_I^0 d_i \omega^2)^2}{48\pi\epsilon_0 c^3}, \quad (2.1.27)$$

where ϵ_0 is the vacuum permittivity, c is the speed of light, and $\langle \cdot \rangle$ denotes the cycle average. As an example, in the case of the oscillations in Fig. 2.3 the frequency is $\omega/2\pi = 1.75$ THz, and the amplitude of oscillation 2.5×10^4 . This yields a total emitted power of $I_c \approx 0.9$ nW.

It is important to note in eq. (2.1.27) that the emitted power is proportional to the square of the number of oscillators. This phenomena is termed superradiance [159], in which coherence between oscillators induces emission superlinear in oscillator numbers. It is highly desirable, as it allows reaching high emission power at the engineering stage by up-scaling the device, as long as coherence is maintained across the whole device area. Coherence is typically maintained by a coherent pumping laser. If for example the pumping spot diameter is $100 \mu\text{m}$, the emitted power becomes $I_c \approx 5.3 \mu\text{W}$.

2.2 Amplification with terahertz cavity

In the preceeding section the dipolariton system, and the emission due to oscillations of the dipole moment, was described classically. This has the drawback that possible quantum effects are not described. Neither does the model account for the energy that should be lost due to radiation. To calculate the effect quantum mechanically, it is easiest to put the system into an external cavity, tuned to resonance with the THz oscillations in the system (see Fig. 2.5). This will also further the desired effect by enhancing emission through bosonic stimulation. The orientation of the THz cavity should be perpendicular to the growth direction of the dipolariton structure for effective coupling of the THz cavity and the indirect exciton z -oriented dipole moment to take place.

Figure 2.5: Sketch of the dipolariton system as enclosed in the THz cavity. The DBRs form an optical cavity, housing photons (C). The photons couple with Rabi frequency Ω to a resonant exciton transition in QW1 to form direct excitons (DX). Electrons can tunnel with frequency J to QW2 to form indirect excitons (IX). Oscillations between DX and IX correspond to an oscillating z-direction dipole moment, that couples to the THz mode, housed by the external cavity.

2.2.1 Model

The Hamiltonian of the system dipolaritons + THz cavity is simply the Hamiltonian $\mathcal{H}_0$, eq. (2.1.1), of the dipolariton system, with the addition of a free energy term of the THz cavity photons, and an interaction term:

$$\mathcal{H} = \mathcal{H}_0 + \hbar\omega_T \hat{a}_T^\dagger \hat{a}_T + \hat{V}. \quad (2.2.1)$$

Here ω_T is the angular frequency of the terahertz mode, $\hat{a}_T^\dagger$ and $\hat{a}_T$ are the creation and annihilation operators of a THz cavity photon, and $\hat{V}$ is the interaction between the THz cavity photons and the dipolariton system. This interaction can be described in the dipole approximation, as

$$\hat{V} = -\hat{\mathbf{d}} \cdot \hat{\mathcal{E}}_T, \quad (2.2.2)$$

where $\hat{\mathbf{d}}$ is the dipole moment operator of the dipolariton system, and $\hat{\mathcal{E}}_T$ is the operator of the THz cavity photon electric field. The single mode THz field can be expressed as

$$\hat{\mathcal{E}}_T = \sqrt{\frac{\hbar\omega_T}{2\varepsilon V}}(\hat{a}_T\mathbf{e} + \hat{a}_T^\dagger\mathbf{e}^*). \quad (2.2.3)$$

Here ε is the dielectric constant and V is the quantization volume of the THz cavity, and $\mathbf{e}$ is the polarization vector. Using the operators for the excitonic modes, the dipole moment operator can be expanded in the basis of these states:

$$\hat{\mathbf{d}} = \mathbf{d}_{dd}\hat{b}^\dagger\hat{b} + \mathbf{d}_{ii}\hat{c}^\dagger\hat{c} + \mathbf{d}_{di}\hat{b}^\dagger\hat{c} + \mathbf{d}_{id}\hat{c}^\dagger\hat{b}, \quad (2.2.4)$$

where $\mathbf{d}_{jk} = \langle j|\hat{\mathbf{d}}|k\rangle$ are the dipole matrix elements of the excitonic states. As we have translational invariance in the x - y plane, matrix elements have only a z -component $\mathbf{d}_{jk} = d_{jk}\mathbf{e}_z$, where $d_{jk} = -e\langle j|\hat{z}|k\rangle$.

As discussed previously, the indirect exciton dipole moment is simply the elementary charge e multiplied by the effective electron-hole separation L ,

$$d_{ii} = -e\langle IX|\hat{z}|IX\rangle = -eL. \quad (2.2.5)$$

The direct exciton acquires a dipole moment from the applied field F as described by the quantum-confined Stark effect:

$$d_{dd} = -e\langle DX|\hat{z}|DX\rangle = -24\left(\frac{2}{3\pi}\right)^6 \frac{e^2 F m^* d^4}{\hbar^2}. \quad (2.2.6)$$

Here m^* is the electron effective mass in the QW material, and d is the QW thickness. We take parameters from Ref. [102], where measurements were made on $d = 10$ nm thick $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ QW. The effective mass is then $m^* = 0.06m_e$, where m_e is the free electron mass. The applied field we assume is close to the resonance field, $F \approx F_0$. We find that the direct exciton dipole moment is much smaller than that of the indirect exciton, $d_{dd} \approx 10^{-3}d_{ii}$, and we proceed by neglecting it in what follows.

Finally the overlap dipole moment $d_{id} = d_{di}$ is calculated in Appendix C. For the chosen parameters it is estimated as $d_{id} \approx 0.17d_{ii}$.

As the interaction is proportional to the dot product $\mathbf{e} \cdot \mathbf{e}_z$, the interaction is maximal when the THz mode is linearly polarized in the z -direction, $\mathbf{e} = \mathbf{e}_z$. Assuming so, we can write the interaction as

$$\hat{V} = [g\hat{c}^\dagger\hat{c} + \tilde{g}(\hat{b}^\dagger\hat{c} + \hat{c}^\dagger\hat{b})](\hat{a}_T + \hat{a}_T^\dagger), \quad (2.2.7)$$

where the interaction constants read $g = eL\sqrt{\hbar\omega_T/2\varepsilon V}$ and $\tilde{g} = -d_{id}\sqrt{\hbar\omega_T/2\varepsilon V}$.

It is pedagogic to consider the special case where the excitonic modes are resonant, $\omega_D = \omega_I$, and the cavity mode far detuned from the excitonic modes. In this case level repulsion generates two new eigenmodes of the excitons. We can term the two new modes as the symmetric exciton, with creation operator $\hat{a}_s^\dagger = (\hat{b}^\dagger + \hat{c}^\dagger)/\sqrt{2}$ and energy $\hbar\omega_s = \hbar\omega_D - \hbar J/2$, and the antisymmetric exciton, with creation operator $\hat{a}_a^\dagger = (\hat{b}^\dagger - \hat{c}^\dagger)/\sqrt{2}$ and energy $\hbar\omega_s = \hbar\omega_D + \hbar J/2$. The interaction (2.2.7) can be rewritten in these modes as

$$\hat{V} = -\left[\left(\frac{g}{2} + \tilde{g}\right)\hat{a}_s^\dagger\hat{a}_s + \left(\frac{g}{2} - \tilde{g}\right)\hat{a}_a^\dagger\hat{a}_a\right](\hat{a} + \hat{a}^\dagger) + \frac{g}{2}(\hat{a}\hat{a}_s\hat{a}_a^\dagger + \hat{a}^\dagger\hat{a}_s^\dagger\hat{a}_a) + \frac{g}{2}(\hat{a}\hat{a}_a\hat{a}_s^\dagger + \hat{a}^\dagger\hat{a}_a^\dagger\hat{a}_s), \quad (2.2.8)$$

The first term describes the dipole interaction between the electric field of the THz mode and the static dipole moment of the symmetric and antisymmetric excitons. The third term however describes the dynamic processes where first a THz photon is annihilated and a lower energy symmetric exciton is excited to the antisymmetric state, as well as the opposite process where an antisymmetric exciton relaxes to the symmetric state, accompanied by the creation of a THz photon. As the exciton level splitting is on the THz scale, this process can be made resonant by tuning the THz cavity to the energy gap. This processes that are present also in the more complicated three-mode polaritons, and should straightforwardly add to the occupancy of the THz cavity mode. The third and final term represents the anti-resonant processes where first a THz photon is annihilated and an antisymmetric exciton simultaneously relaxes to the symmetric state, and where a symmetric exciton is excited to the antisymmetric state and a THz photon is simultaneously created. These processes are energy non-conservant, and are typically neglected.

We derive equations of motion in the same way as in section 2.1.1, applying the

Heisenberg equation to the operators, taking expectation values, and decoupling operators with the mean field approximation. As no terms of the Hamiltonian couple the THz mode and the optical cavity mode, the equation for $\langle \hat{a} \rangle$ remains unchanged from eq. (2.1.7). The equations for $\langle \hat{b} \rangle$ and $\langle \hat{c} \rangle$ acquire terms coupling them to the THz mode. Finally the THz order parameter $\langle \hat{a}_T \rangle$ is also governed by a dynamic equation, yielding a system of four coupled dynamic equations. The change of variables $\hat{o}_i \rightarrow e^{-i\omega_c t} \hat{o}_i$ is performed on all operators except those of the THz mode.

$$\frac{\partial \langle \hat{a}_T \rangle}{\partial t} = -i\omega_T \langle \hat{a}_T \rangle + i\frac{g}{\hbar} |\langle \hat{c} \rangle|^2 + i\frac{2\tilde{g}}{\hbar} \text{Re}[\langle \hat{b} \rangle^* \langle \hat{c} \rangle] - \frac{1}{2\tau_T} \langle \hat{a}_T \rangle, \quad (2.2.9)$$

$$\frac{\partial \langle \hat{a} \rangle}{\partial t} = -i\frac{\Omega}{2} \langle \hat{b} \rangle - \frac{1}{2\tau_c} \langle \hat{a} \rangle - i\tilde{P}(t), \quad (2.2.10)$$

$$\begin{aligned} \frac{\partial \langle \hat{b} \rangle}{\partial t} = & -i \left[-\delta_\Omega + \frac{V_{DD}}{\hbar} |\langle \hat{b} \rangle|^2 + \frac{V_{DI}}{\hbar} |\langle \hat{c} \rangle|^2 \right] \langle \hat{b} \rangle - i\frac{\Omega}{2} \langle \hat{a} \rangle + i\frac{J}{2} \langle \hat{c} \rangle \\ & + i\frac{2\tilde{g}}{\hbar} \text{Re}[\langle \hat{a}_T \rangle] \langle \hat{c} \rangle - \frac{1}{2\tau_D} \langle \hat{b} \rangle, \end{aligned} \quad (2.2.11)$$

$$\begin{aligned} \frac{\partial \langle \hat{c} \rangle}{\partial t} = & -i \left[\delta_J - \delta_\Omega + \frac{V_{DI}}{\hbar} |\langle \hat{b} \rangle|^2 + \frac{V_{II}}{\hbar} |\langle \hat{c} \rangle|^2 - \frac{2g}{\hbar} \text{Re}[\langle \hat{a}_T \rangle] \right] \langle \hat{c} \rangle + i\frac{J}{2} \langle \hat{b} \rangle \\ & + i\frac{2\tilde{g}}{\hbar} \text{Re}[\langle \hat{a}_T \rangle] \langle \hat{b} \rangle - \frac{1}{2\tau_I} \langle \hat{c} \rangle, \end{aligned} \quad (2.2.12)$$

Here the lifetime of the THz photon was introduced as $\tau_T = Q/\omega_T$, where Q is the quality factor of the THz cavity. For a given occupation $N_T = |\langle \hat{a}_T \rangle|^2$ of the THz mode, the emission intensity is estimated straightforwardly as the product of the total THz cavity leakage rate N_T/τ_T and the energy of each individual THz photon $\hbar\omega_T$,

$$I = \frac{N_T \hbar\omega_T}{\tau_T}. \quad (2.2.13)$$

2.2.2 Low-Q analytical solution

Before solving numerically the eqns. (2.2.9)-(2.2.12) to find the CW pumping THz photons numbers, it is instructional to first consider a analytical approximation that can be made for small values of the quality factor Q . Under this condition also the

THz occupancy N_T will be small. As such all the terms in eqns. (2.2.10)-(2.2.12) proportional to $Re[\langle\hat{a}_T\rangle] \sim \sqrt{N_T}$ can be neglected, and the dipolariton equations revert to ones without the THz cavity, (2.1.7)-(2.1.9). We know that these equations can be solved in certain regimes to give nondecaying oscillations of indirect exciton numbers under CW pumping, $|\langle\hat{c}\rangle|^2 = N_I(t) = N_0[\cos(\omega t) + 1]/2$. In the equation for the THz mode (2.2.9) we can further neglect the smaller $\tilde{g}$ term, and the equation reads

$$\frac{\partial\langle\hat{a}_T\rangle}{\partial t} = -i\omega_T\langle\hat{a}_T\rangle + i\frac{gN_0}{2\hbar} \left[\frac{e^{i\omega t} + e^{-i\omega t}}{2} + 1 \right] - \frac{1}{2\tau_T}\langle\hat{a}_T\rangle. \quad (2.2.14)$$

Now the THz mode equation is decoupled from the dipolariton equations, with the indirect exciton number oscillations entering as a driving term in eq. (2.2.14). We now assume that the THz cavity is in resonance with the IX oscilltions. Out of the three terms in the driving term, only the one proportional to $e^{-i\omega_T t}$ is resonant, and we neglect the others. We make a rotating wave ansatz $\langle\hat{a}_T\rangle = e^{-i\omega_T t}\psi_T$, and insert into the resulting equation, and get

$$\psi_T = i\frac{gN_0\tau_T}{2\hbar}. \quad (2.2.15)$$

As a result, the THz cavity occupancy is $N_T = |\psi_T|^2 = g^2N_0^2\tau_T^2/4\hbar^2$, and the resulting estimate for the power of emission is

$$I_q = \frac{g^2N_0^2\tau_T\omega_T}{4\hbar} = \frac{g^2N_0^2Q}{4\hbar} \quad (2.2.16)$$

It should be immediately noticed from eq. (2.2.16) that the superradiant property found in the classical estimate of the emission is confirmed by the quantum mechanical treatment of the THz dynamics. Secondly we see a linear enhancement of the emission power in the quality factor Q . To estimate the power amplification of the THz cavity we can divide this quantum estimate with the classical estimate (2.1.27):

$$\frac{I_q}{I_c} = \frac{12\pi\epsilon c^3 g^2 Q}{d_i^2 \hbar \omega_T^4}. \quad (2.2.17)$$

Recalling that the interaction constant reads $g = d_i \sqrt{\hbar \omega_T / 2 \epsilon V}$ allows us to write cancel the indirect exciton dipole moment, the dielectric constant, and one factor of the THz mode energy:

$$\frac{I_q}{I_c} = 6\pi \frac{c^3 Q}{\omega_T^3 V}. \quad (2.2.18)$$

Finally the angular frequency of an electromagnetic field and its wavelength are related as $\omega \lambda = 2\pi c$. As such

$$\frac{I_q}{I_c} = 6\pi \left(\frac{\lambda_T}{2\pi} \right)^3 \frac{Q}{V} = \frac{3}{4\pi} \frac{\lambda_T^3}{V} Q, \quad (2.2.19)$$

where λ_T is the wavelength of the THz field inside the cavity. This fraction exactly coincides with the Purcell factor, $I_q/I_c = F_P$, that was originally found when calculating the enhancement of spontaneous emission from fluorescent molecules [160].

2.2.3 Numerical solutions

The eqns. (2.2.9)-(2.2.12) were now solved numerically under CW pumping. Parameters were kept the same as in 2.1.4 and figure 2.3, with mode detuning as $\hbar \delta_\Omega = -3$ meV and $\hbar \delta_J = 1$ meV, and the pump detuning $\hbar \Delta_p = 4.5$ meV, slightly above the middle polariton energy. For these parameters a region of parametric instability exists in the pumping strength, and by turning the pump strength on quickly, the system is driven into the parametrically unstable state. The resulting oscillations in IX occupancy should couple to the THz mode, and result in occupancy of the THz cavity. Emission power is then calculated as a function of quality factor Q via eq. (2.2.13), using the steady state THz occupation number $N_T = |\langle \hat{a}_T \rangle|^2$ (see Fig. 2.6).

The behavior of the output power on the quality factor is nonmonotonous. At small values of the quality factor (up to $Q = 17$ and $Q = 30$ for two different values of the pumping strength) the power increases rapidly, following the low- Q approximation. Past this point however the emission power decreases, before increasing again. The emission power however does not reach again the value of the low- Q maximum. This is very desirable, as quality factors very accessible in practice yield optimal emission capacity. Pumping strength $|\tilde{P}_0|^2 = 1.1 \times 10^{29} \text{ s}^{-2}$ yielded

Figure 2.6: Plot of power emitted from the THz cavity, as calculated from eq. (2.2.13) using the steady state THz occupation under CW pumping, as a function of the THz cavity quality factor. The full line marked *A* indicates the pumping strength used in section 2.1.4 and figure 2.3, $|\tilde{P}_0|^2 = 1.1 \times 10^{29} \text{ s}^{-2}$. The dashed line indicates the analytical power estimate (2.2.16) for pump *A*, valid at low values of the quality factor. The numerical result coincides with this approximation up to approximately $Q = 17$, at which the emission power peaks. When the quality factor is increased further the emission power drops as the increased feedback of the THz mode on the excitons suppresses the IX oscillations. The calculation was repeated for slightly different pumping strength $|\tilde{P}_0|^2 = 0.9 \times 10^{29} \text{ s}^{-2}$, indicated by dotted line *B*. A similar behavior is observed, with the peak shifted to approximately $Q = 30$.

without the THz cavity $I_c = 0.9 \text{ nW}$. With a $Q = 17$ cavity emission is increased to $I_q = 51 \text{ nW}$, an enhancement of factor $F_P = I_q/I_c \approx 56$. To better understand the nonmonotonous behaviour of the emission power, it is instructional to look at the detailed dynamics of the indirect exciton and THz photon occupancy.

As discussed in section 2.1.4, after the switching to the parametrically unstable regime, oscillations of indirect exciton numbers settle into harmonicity in a few hundred picoseconds. In Fig. 2.7(a) the dynamics of the THz occupancy is plotted on a logarithmic scale, for different values of the THz cavity quality factor. Parameters are the same as before, with pumping strength chosen as $|\tilde{P}_0|^2 = 1.1 \times 10^{29} \text{ s}^{-2}$. We see that at first when IX oscillations are irregular oscillations of small THz numbers appear. After approximately 200 ps, when IX oscillations are becoming harmonic, a large increase of the THz occupancy occurs. For small values of the

Figure 2.7: **(a)** Plot of dynamics of the natural logarithm of the THz mode occupancy for different values of the THz cavity quality factor Q . Pumping intensity is $|\hat{P}_0|^2 = 1.1 \times 10^{29} \text{ s}^{-2}$. For up to $Q = 17$, corresponding to the maximal occupancy it grows to its steady state value with little feedback on the dipolariton system. At $Q = 23$ we can observe a new behavior where THz occupancy grows to a large value, and then decays as the high THz occupancy influences the IX dynamics. The steady state value is lower than for $Q = 17$. At $Q = 40$ this effect is so strong that the steady state occupancy is as low as for $Q = 10$. At $Q = 100$ and $Q = 200$ the suppression effect is diminished, and steady state occupancy is larger than for $Q = 17$. The inset shows the THz occupancy at the end of the period on the ps scale. There are small undulations in the occupancy of the same frequency as the IX oscillations. **(b)** Plot of IX dynamics for the same conditions as in (a). The dashed line repeats the dynamics shown in Fig. 2.3 in the absence of the THz cavity. For the lower quality factor $Q = 17$, at the limit of the no-feedback approximation, the IX oscillation amplitude is similar to the no-cavity case, but shifted to lower numbers. For $Q = 40$, where emission power takes a local minima, the oscillation amplitude is strongly suppressed by the THz mode. For $Q = 200$ the oscillation amplitude has increased, but is still smaller than for $Q = 17$.

quality factor, $Q \leq 17$, the THz occupancy stays at or near the peak value in the large- t steady state. From Fig. 2.7(b) we see that at this value of the quality factor the amplitude of oscillation of IX numbers is very close to the amplitude in the absence of the THz cavity, although the oscillations are shifted to slightly lower occupancies. This illustrates the essence of the approximation used to find the low- Q analytical estimate (2.2.16), shown in Fig. 2.6 to be valid at up to $Q = 17$, that the THz occupancy feedback on the excitonic mode is negligible.

Comparing with Fig. 2.6, the quality factor $Q = 23$ corresponds to a point where the output power is dropping as the quality factor is increased. In Fig. 2.7(a) it is shown that for this quality factor, after peaking at ~ 200 ps, the THz occupancy decays to below the steady state occupancy for $Q = 17$. At $Q = 40$, where the output power is lowest, the same dynamic is present, with THz occupancy decreasing by three orders of magnitude from its peak, to a steady state occupancy close to that of the $Q = 10$ cavity. Looking at the corresponding IX steady state dynamics in Fig. 2.7(b), this can be explained by the fact that the amplitude of IX oscillations is much lower than for $Q = 17$. At $Q = 100$ and through $Q = 200$ the steady state occupancy increases again, and the amplitude of IX-oscillations recovers, although remaining smaller than for $Q = 17$.

For the highest values of the quality factor, especially for $Q = 200$, oscillations at the 100 ps scale can be observed in the THz occupancy, that decay as it reaches the steady state. As the quality factor is increased further these oscillations persist further, until at around $Q \geq 10^3$ the oscillations persist in the steady state, as shown in Fig. 2.8. The oscillations have a period of approximately 70 ps, 100 times as long as the IX oscillations. The oscillations can be intuitively explained from the preceeding observations. The large number THz photons excited as the IX oscillations become harmonic suppress the IX oscillation amplitude. As such the THz occupancy decays, and the feedback on the excitonic modes is lifted, and the IX oscillations recover. This cycle continuously repeats, and the THz occupancy oscillates. The period of the oscillation is 73 ps. At the peak of the oscillations the occupancy is $N_T = 8500$. The corresponding emission power is $I = 86$ nW, slightly higher than the $Q = 17$ maximum. As such this functionality mimics the Q -switched laser,

Figure 2.8: Dynamics of the THz mode occupancy for quality factor $Q = 1200$. For this high a quality factor the THz occupancy becomes very large. The feedback on the excitonic modes suppresses the oscillations and the occupancy falls. When the feedback consequently disappears the oscillations recover, and a large number of THz photons are again excited. The cycle continues, with no steady state developing. These standing oscillations of THz occupancy mimic the output of the Q -switched laser, with a period of approximately 70 ps.

although to less a dramatic of an effect.

As for the case where there is no THz cavity, it is important to address the tunability of emission frequency. By simply changing the strength of the applied electric field F the detuning of the indirect exciton mode can be changed. The change in the energy level structure of the system changes the frequency of the IX oscillations. Changing the applied field strength alone will however bring the oscillations out of resonance with the THz cavity, and would therefore be useless. If we however imagine that the resonance frequency of the THz cavity can be changed, for example by applying stress to deform the cavity, the THz cavity can be imaged to be kept in resonance as the applied field is varied.

A surface plot of the emission power as a function of the electric field strength and the THz cavity eigenfrequency is shown in Fig. 2.9. It is immediately clear that the light areas corresponding to the highest emission power follow closely the red line, indicating the dependence of the frequency of oscillations shown in Fig. 2.4(a) in the absence of the THz cavity. In the lower electric field limit of the graph the

Figure 2.9: Density plot of the emission power as a function of field F and THz cavity eigenfrequency. The optical detuning is $\hbar\delta_\Omega = -1$ meV, pump energy is $\hbar\Delta_p = 4.5$ meV and pump intensity is $|\tilde{P}_0|^2 = 1.1 \times 10^{29} \text{ s}^{-2}$. The quality factor of the THz cavity is $Q = 17$. Lighter colors indicate higher emission power. The red line indicates the IX oscillation frequency as given in Fig. 2.4(a).

emission power diminishes as the region of multistability shifts to lower pumping strengths. By additionally altering the pumping strength to suit the energy level configuration, the range of emission frequency could be extended to higher frequencies. As the electric field is increased past the limit of this plot the IX oscillations become anharmonic. Although not depicted, at this point it was found that emission power dropped to close to zero.

3. Stationary transport in graphene dressed by light

This chapter describes research and results published in the work [A3]. The effect of dressing by light on the stationary transport properties of graphene are investigated. The first section reviews how the most important property of graphene, its linear dispersion, arises as a consequence of its hexagonal lattice structure.

3.1 Graphene dispersion

Graphene consists of a two-dimensional hexagonal lattice of carbon atoms. Carbon atoms have the atomic number 6, and consequently have 6 electrons. They are organized into a filled $1s$ orbital, a filled $2s$ orbital, and two electrons in the $2p$ orbital. The $2s$, $2p_x$ and the $2p_y$ orbitals can hybridize, producing three sp^2 orbitals. They are oriented equiangularly in a plane. One sp^2 orbital from each of neighbouring carbon atoms can form a strong σ bond, contributing one electron each. The carbon-carbon separation of the σ bond is $a = 0.142$ nm. Each carbon atom can in this way bond with with three other carbon atoms, naturally forming the hexagonal lattice [see Fig. 3.1(a)]. The remaining p_z orbital can form π bonds with adjacent carbon atoms. But as the orbital only has one electron, the π bonds are only half full.

The primitive cell of graphene can be chosen as indicated by the red lines in

Figure 3.1: **(a)** Diagram of the graphene hexagonal lattice. The carbon atoms are shown as black orbs, labelled *A* and *B* identifying their sublattice. The trigonal sp^3 hybridized orbitals are shown as light blue spheroids. Overlapping orbitals illustrate the σ bonds between graphene atoms. The hexagonal lattice emerges as a consequence of the planar three-partition bonds. The elementary cell of the graphene lattice structure is shown in red lines, with lattice vectors $\mathbf{a}_1$ and $\mathbf{a}_2$. The next-neighbour vectors are $\boldsymbol{\delta}_i$, $i = 1, 2, 3$, shown in blue. **(b)** The dispersion of graphene $\epsilon_{\mathbf{k}} = -t |\phi(\mathbf{k})|$, plotted in the Brillouin zone, as found from the tight-binding approximation. The lattice vectors of the reciprocal lattice $\mathbf{b}_1$, $\mathbf{b}_2$ are shown in red. The dispersion is linear around the two points K_+ and K_- .

Fig. 3.1(a). Each cell contains two atoms. The lattice vectors read

$$\mathbf{a}_1 = \frac{a}{2}(3, \sqrt{3}), \quad \mathbf{a}_2 = \frac{a}{2}(3, -\sqrt{3}). \quad (3.1.1)$$

The two atoms of the primitive cell *A* and *B* form separate sublattices. To find the eigenmodes of the lattice, we should pass to the Fourier space, or momentum space representation. The lattice vectors of the reciprocal lattice are given by $\mathbf{b}_1 = 2\pi \mathbf{a}_2 \times \mathbf{e}_z / \mathbf{a}_1 \cdot (\mathbf{a}_2 \times \mathbf{e}_z)$ and $\mathbf{b}_2 = 2\pi \mathbf{e}_z \times \mathbf{a}_1 / \mathbf{a}_2 \cdot (\mathbf{e}_z \times \mathbf{a}_1)$. This gives us

$$\mathbf{b}_1 = \frac{2\pi}{3a}(1, \sqrt{3}), \quad \mathbf{b}_2 = \frac{2\pi}{3a}(1, -\sqrt{3}). \quad (3.1.2)$$

To describe this system, creation (annihilation) operators can be defined for a Wan-

nier state electron at site A of cell i , $\hat{a}_i^\dagger$ ($\hat{a}_i$), and at site B of cell i , $\hat{b}_i^\dagger$ ($\hat{b}_i$). Here we neglect the spin degree of freedom. If we make the approximation of electrons hopping only between adjacent carbon atoms, we can describe the system with the following tight-binding Hamiltonian:

$$\mathcal{H} = -t \sum_{\langle i,j \rangle} \left(\hat{a}_i^\dagger \hat{b}_j + \hat{b}_i^\dagger \hat{a}_j \right). \quad (3.1.3)$$

Here $\langle i,j \rangle$ indicates summation over the indices i, j such that cells i and j are adjacent. The Hamiltonian should now be expressed in terms of the operators for Bloch states of the sublattice A ($\hat{a}_{\mathbf{k}}^\dagger$, $\hat{a}_{\mathbf{k}}$) and the sublattice B ($\hat{b}_{\mathbf{k}}^\dagger$, $\hat{b}_{\mathbf{k}}$), with wavevector $\mathbf{k}$. The Wannier states can be written in terms of the Bloch states as

$$\hat{a}_i^\dagger = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}_{Ai}} \hat{a}_{\mathbf{k}}^\dagger, \quad (3.1.4)$$

$$\hat{b}_i^\dagger = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}_{Bi}} \hat{b}_{\mathbf{k}}^\dagger. \quad (3.1.5)$$

Here $\mathbf{R}_{Ai}$ and $\mathbf{R}_{Bi}$ are the positions of the carbon atom sites in cell i of the sublattice A and B respectively. The Hamiltonian can then be written as

$$\mathcal{H} = -\frac{t}{N} \sum_{\mathbf{k}, \mathbf{k}'} \left(\hat{a}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}'} \sum_{\langle i,j \rangle} e^{-i\mathbf{k} \cdot \mathbf{R}_{Ai} + i\mathbf{k}' \cdot \mathbf{R}_{Bj}} + \text{h.c.} \right). \quad (3.1.6)$$

Here h.c. denotes the hermition conjugate. To perform the summation in the cell indices i and j , we note that for a given site $\mathbf{R}_{Ai}$ on sublattice A , the position of the adjacent atoms on sublattice B are found by the adding the nearest neighbour vectors [see Fig. 3.1(a)]

$$\boldsymbol{\delta}_1 = \frac{a}{2}(1, \sqrt{3}), \quad \boldsymbol{\delta}_2 = \frac{a}{2}(1, -\sqrt{3}), \quad \boldsymbol{\delta}_3 = -a(1, 0). \quad (3.1.7)$$

The summation over the cell indices then yields

$$\sum_{\langle i,j \rangle} e^{-i\mathbf{k} \cdot \mathbf{R}_{Ai} + i\mathbf{k}' \cdot \mathbf{R}_{Bj}} = \sum_i \sum_{l=1}^3 e^{-i\mathbf{k} \cdot \mathbf{R}_{Ai} + i\mathbf{k}' \cdot (\mathbf{R}_{Ai} + \boldsymbol{\delta}_l)} = \sum_i e^{-i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{R}_{Ai}} \phi(\mathbf{k}'), \quad (3.1.8)$$

where $\phi(\mathbf{k}) = \sum_{l=1}^3 e^{i\mathbf{k} \cdot \boldsymbol{\delta}_l}$. Finally the summation over all the Wannier sites yields a Kronecker delta, $\sum_i e^{-i(\mathbf{k}-\mathbf{k}') \cdot \mathbf{R}_{Ai}} = N\delta_{\mathbf{k},\mathbf{k}'}$. The summation over $\mathbf{k}'$ in eq. (3.1.6) is then resolved by the Kronecker delta, and the Hamiltonian becomes

$$\mathcal{H} = -t \sum_{\mathbf{k}} \left(\phi(\mathbf{k}) \hat{a}_{\mathbf{k}}^\dagger \hat{b}_{\mathbf{k}} + \phi(\mathbf{k})^* \hat{b}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}} \right). \quad (3.1.9)$$

Now, as ϕ is a complex function, it can be written as $|\phi(\mathbf{k})| e^{i\theta(\mathbf{k})}$, where θ is a real function. The Hamiltonian (3.1.9) can be diagonalized by the operators

$$\hat{c}_{\mathbf{k}\pm}^\dagger = \frac{1}{\sqrt{2}} \left(e^{i\theta(\mathbf{k})} \hat{a}_{\mathbf{k}}^\dagger \pm \hat{b}_{\mathbf{k}}^\dagger \right), \quad (3.1.10)$$

giving the form of the Hamiltonian as

$$\mathcal{H} = -t \sum_{\mathbf{k}} |\phi(\mathbf{k})| \left(\hat{c}_{\mathbf{k}+}^\dagger \hat{c}_{\mathbf{k}+} - \hat{c}_{\mathbf{k}-}^\dagger \hat{c}_{\mathbf{k}-} \right). \quad (3.1.11)$$

The dispersion of graphene can be read from this equation as $\varepsilon_{\mathbf{k}} = \pm t |\phi(\mathbf{k})|$. This dispersion is shown in the Brillouin zone in Fig. 3.1(b). The dispersion is found to be linear at zero energy around two points $\mathbf{K}_\pm$ in the Brillouin zone. These points are called the Dirac points. They lie at wavevectors

$$\mathbf{K}_\pm = \frac{2\pi}{3a} \left(1, \pm \frac{1}{\sqrt{3}} \right). \quad (3.1.12)$$

As such, low energy excitations of graphene will have a wavevector that differs only slightly from the Dirac point wavevector, $\mathbf{k} = \mathbf{K}_\pm + \mathbf{q}$, $q = |\mathbf{q}| \ll |\mathbf{K}_\pm|$. The function ϕ can then be expanded in orders of $\mathbf{q}$ around $\mathbf{K}_\pm$ to find the dispersion around the Dirac point. Straightforward evaluation of the ϕ function shows that around the

positive Dirac point

$$\phi(\mathbf{K}_+ + \mathbf{q}) \approx -\frac{3\sqrt{3}}{4}q_x a - \frac{3}{4}q_y a + i \left(\frac{3}{4}q_x a - \frac{3\sqrt{3}}{4}q_y a \right), \quad (3.1.13)$$

and the energy of small excitations is

$$\varepsilon_{\mathbf{q}} = \pm t |\phi(\mathbf{K}_+ + \mathbf{q})| \approx \pm \frac{3ta}{2} \sqrt{q_x^2 + q_y^2} = \pm \hbar v_F q. \quad (3.1.14)$$

Here the Fermi velocity $v_F = 3ta/2\hbar$ is defined in analogy with how the speed of light enters in the relativistic energy-momentum relationship of light. The Fermi velocity is approximately $v_F \approx 10^6$ m/s.

For low energy excitations, the Dirac cones around the two Dirac points act as independent excitations, referred to as the valley degree of freedom. As such in the low energy approximation graphene should, when accounting for spin and the sublattice A and B, be described by a 8-dimensional Hamiltonian. In the following treatment however we neglect both the spin and the valley degrees of freedom (see e.g. [161]). As such the low energy graphene excitations are described by the single particle Hamiltonian

$$\hat{\mathcal{H}} = \hbar v_F \hat{\boldsymbol{\sigma}} \cdot \hat{\mathbf{k}} = \hbar v_F \begin{pmatrix} 0 & \hat{k}_x - i\hat{k}_y \\ \hat{k}_x + i\hat{k}_y & 0 \end{pmatrix}. \quad (3.1.15)$$

Here $\hat{\boldsymbol{\sigma}} = (\hat{\sigma}_x, \hat{\sigma}_y)$ is the 2D vector of the Pauli matrices, and $\hat{\mathbf{k}} = -i\nabla$ is the wavevector operator. The fundamental reason why the sublattice degree of freedom cannot be similarly neglected is mathematical in nature. A linear dispersion cannot be analytical at the $k = 0$ point unless it extends to negative energy as well.

3.2 Theory of dressed graphene

The goal of this section is to find the stationary transport properties of graphene dressed with a strong electromagnetic wave. The first step will be to find how the dressing field modifies eigenfunctions and the dispersion of graphene. Armed with

Figure 3.2: Sketch of the graphene hexagonal carbon lattice with an electromagnetic field propagating in the z direction, with electric field intensity $\mathcal{E}_0$. The system is treated separately for the (a) circular and (b) linear polarization of the irradiating field.

the complete quantum mechanical description of the dressed system, the scattering rates can be calculated, and from them the DC conductivity can be found with Boltzmann theory.

3.2.1 Model

As the properties that are investigated, stationary transport, are single particle properties, the wave function of graphene interacting with a strong electromagnetic field should be found by solving the single particle Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} \psi = \hat{\mathcal{H}} \psi. \quad (3.2.1)$$

For the sake of definiteness, the graphene sheet is specified as lying in the x - y plane, and the electromagnetic field as a uniform plane wave propagating in the positive z direction (see Fig. 3.2). The Hamiltonian of the graphene electrons is obtained from (3.1.15) by replacing the canonical momentum operator with the kinetic momentum in the presence of a vector potential:

$$\hat{\mathcal{H}}(t) = v \hat{\boldsymbol{\sigma}} \cdot (\hbar \hat{\mathbf{k}} - e \mathbf{A}(t)). \quad (3.2.2)$$

Here e is the electron charge, the subscript F is dropped from the Fermi velocity and $\mathbf{A}$ is the vector potential of the electromagnetic wave propagating through the

graphene plane, evaluated in the graphene plane. As such the vector potential does not depend on the spatial x, y coordinates. As the Hamiltonian has explicit time-dependence, there are no true energy eigenstates. Instead due to the periodicity of the Hamiltonian, Floquet theory admits solutions characterized by quasienergies, analogous to the quasimomentum of crystal Bloch states.

The cases of circular and linear polarization of the irradiating wave are non-trivially different, and are treated separately in the two following subsections.

3.2.2 Dispersion and wavefunctions for circular polarization

For circular polarization of the wave the vector potential can be written as

$$\mathbf{A}_c = \left(\frac{\mathcal{E}_0}{\omega} \cos \omega t, \frac{\mathcal{E}_0}{\omega} \sin \omega t \right), \quad (3.2.3)$$

where $\mathcal{E}_0$ is the electric field amplitude of the wave, and ω is the wave frequency. The frequency ω of the field is assumed not to be close to any interband resonances.

The Hamiltonian can be written as

$$\hat{\mathcal{H}}(t) = \hat{\mathcal{H}}_0(t) + \hat{\mathcal{H}}_{\mathbf{k}}, \quad (3.2.4)$$

where

$$\hat{\mathcal{H}}_0(t) = -\frac{ve\mathcal{E}_0}{\omega} \begin{pmatrix} 0 & e^{-i\omega t} \\ e^{i\omega t} & 0 \end{pmatrix}, \quad (3.2.5)$$

$$\hat{\mathcal{H}}_{\mathbf{k}} = \hbar v \begin{pmatrix} 0 & k_x - ik_y \\ k_x + ik_y & 0 \end{pmatrix}. \quad (3.2.6)$$

The nonstationary Schrödinger equation accounting only for the effect of the dressing field (3.2.5),

$$i\hbar \frac{\partial \psi_0}{\partial t} = \hat{\mathcal{H}}_0(t) \psi_0, \quad (3.2.7)$$

describes the dynamics of the zero wavevector state. Solutions to the above equation

can be found with the ansatz

$$\psi_0 = e^{-i\alpha t/\hbar}(A\Phi'_1(\mathbf{r})e^{-i\omega t/2} + B\Phi'_2(\mathbf{r})e^{i\omega t/2}), \quad (3.2.8)$$

where $\Phi'_{1,2}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}\Phi_{1,2}(\mathbf{r})/\sqrt{S}$ are the graphene Bloch states that make up the two-dimensional sublattice basis, $\Phi_{1,2}(\mathbf{r})$ are sublattice-confined periodic functions, S is the system area, and α , A and B are parameters. Substituting the wave function (3.2.8) into the full Schrödinger equation (3.2.7) yields the linear algebra equality

$$\begin{aligned} A\left(\alpha + \frac{\hbar\omega}{2}\right) + B\left(\frac{ve\mathcal{E}_0}{\omega}\right) &= 0 \\ A\left(\frac{ve\mathcal{E}_0}{\omega}\right) + B\left(\alpha - \frac{\hbar\omega}{2}\right) &= 0. \end{aligned} \quad (3.2.9)$$

A solution to this system of equations exists if and only if

$$\begin{vmatrix} \alpha + \frac{\hbar\omega}{2} & \frac{ve\mathcal{E}_0}{\omega} \\ \frac{ve\mathcal{E}_0}{\omega} & \alpha - \frac{\hbar\omega}{2} \end{vmatrix} = 0. \quad (3.2.10)$$

This gives two different solutions for the eigenvalue, $\alpha = \pm\Omega/2$, where

$$\Omega = \sqrt{(\hbar\omega)^2 + \left(\frac{2ve\mathcal{E}_0}{\omega}\right)^2}. \quad (3.2.11)$$

The two corresponding solutions for the other two parameters, subject to the normalization condition $|A|^2 + |B|^2 = 1$, can be written

$$\psi_0^\pm = e^{\pm i\Omega t/2\hbar} \left[\sqrt{\frac{\Omega \pm \hbar\omega}{2\Omega}} \Phi'_1(\mathbf{r})e^{-i\omega t/2} \pm \frac{e}{|e|} \sqrt{\frac{\Omega \mp \hbar\omega}{2\Omega}} \Phi'_2(\mathbf{r})e^{i\omega t/2} \right]. \quad (3.2.12)$$

Now, since the two wave functions (3.2.12) form a complete state basis for any fixed time t , we can seek the solution of the full Schrödinger equation as a linear combination of the two functions.

$$\psi_{\mathbf{k}} = a^+(t)\psi_0^+ + a^-(t)\psi_0^-. \quad (3.2.13)$$

Substituting the expression above into the full Schrödinger equation yields a two coupled differential equations for the functions $a^\pm(t)$,

$$i\dot{a}^+(t) = v \frac{e}{|e|} \left[\frac{W_0}{\Omega} (k_x \cos \omega t + k_y \sin \omega t) a^+(t) - (k_x - ik_y) \left(\frac{\Omega + \hbar\omega}{2\Omega} \right) \right. \\ \left. \times e^{-i(\Omega/\hbar - \omega)t} a^-(t) + (k_x + ik_y) \left(\frac{\Omega - \hbar\omega}{2\Omega} \right) e^{-i(\Omega/\hbar + \omega)t} a^-(t) \right], \quad (3.2.14)$$

$$i\dot{a}^-(t) = -v \frac{e}{|e|} \left[\frac{W_0}{\Omega} (k_x \cos \omega t + k_y \sin \omega t) a^-(t) + (k_x + ik_y) \left(\frac{\Omega + \hbar\omega}{2\Omega} \right) \right. \\ \left. \times e^{i(\Omega/\hbar - \omega)t} a^+(t) - (k_x - ik_y) \left(\frac{\Omega - \hbar\omega}{2\Omega} \right) e^{i(\Omega/\hbar + \omega)t} a^+(t) \right], \quad (3.2.15)$$

where

$$W_0 = \frac{2ve\mathcal{E}_0}{\omega}. \quad (3.2.16)$$

Now the assumptions is made that the dressing field frequency ω satisfies the inequality

$$\hbar\omega \gg W_0. \quad (3.2.17)$$

From this follows that $\Omega \approx \hbar\omega$, and

$$\frac{W_0}{\Omega} \approx 0, \quad \frac{\Omega + \hbar\omega}{2\Omega} \approx 1, \quad \frac{\Omega - \hbar\omega}{2\Omega} \approx 0. \quad (3.2.18)$$

The dynamic equations of the coefficients can then be greatly simplified as

$$i\dot{a}^+(t) = -v \frac{e}{|e|} (k_x - ik_y) e^{-i(\Omega - \hbar\omega)t/\hbar} a^-(t), \\ i\dot{a}^-(t) = -v \frac{e}{|e|} (k_x + ik_y) e^{i(\Omega - \hbar\omega)t/\hbar} a^+(t). \quad (3.2.19)$$

Solutions of these equations are now sought in the form $a_c^\pm(t) = C^\pm e^{-i(\pm\Omega/2 \mp \hbar\omega/2 + \varepsilon_{\mathbf{k}})t/\hbar}$, where $C^\pm$ and $\varepsilon_{\mathbf{k}}$ are free parameters. Solving the system of equations (3.2.19), we arrive at

$$\varepsilon_c^\pm(k) = \pm \sqrt{(\varepsilon_g/2)^2 + (\hbar vk)^2}, \quad (3.2.20)$$

where

$$\varepsilon_g = \sqrt{(\hbar\omega)^2 + \left(\frac{2ve\mathcal{E}_0}{\omega}\right)^2} - \hbar\omega. \quad (3.2.21)$$

In the context of Floquet theory, the quantity (3.2.20) is referred to as the quasienergy of the graphene state dressed by a circularly polarized field. The sign identifies the conduction (+) and valence (-) band. The two wave functions (3.2.13) can be written as

$$\begin{aligned} \psi_{\mathbf{k}}^{\pm} = & e^{-i\varepsilon_{\mathbf{k}}t/\hbar} \left[\pm \sqrt{\frac{|\varepsilon_{\mathbf{k}}| \mp \varepsilon_g/2}{2|\varepsilon_{\mathbf{k}}|}} e^{-i\theta/2} \left(\frac{e}{|e|} \sqrt{\frac{\Omega + \hbar\omega}{2\Omega}} \Phi'_1(\mathbf{r}) + \sqrt{\frac{\Omega - \hbar\omega}{2\Omega}} \Phi'_2(\mathbf{r}) e^{i\omega t} \right) \right. \\ & \left. - \sqrt{\frac{|\varepsilon_{\mathbf{k}}| \pm \varepsilon_g/2}{2|\varepsilon_{\mathbf{k}}|}} e^{i\theta/2} \left(\sqrt{\frac{\Omega - \hbar\omega}{2\Omega}} \Phi'_1(\mathbf{r}) e^{-i\omega t} - \frac{e}{|e|} \sqrt{\frac{\Omega + \hbar\omega}{2\Omega}} \Phi'_2(\mathbf{r}) \right) \right], \end{aligned} \quad (3.2.22)$$

where $\theta = \arctan(k_y/k_x)$ is the polar angle of the wave vector. Applying the conditions (3.2.18), and writing out the Bloch functions, the dressed wave functions for circular polarization can be written as

$$\psi_{c\mathbf{k}}^{\pm}(\mathbf{r}, t) = \frac{e^{i(\mathbf{k}\cdot\mathbf{r} - \varepsilon_c^{\pm}t/\hbar)}}{\sqrt{S}} \left[\sqrt{\frac{|\varepsilon_c^{\pm}(k)| \mp \varepsilon_g/2}{2|\varepsilon_c^{\pm}(k)|}} e^{-i\theta/2} \Phi_1(\mathbf{r}) \pm \sqrt{\frac{|\varepsilon_c^{\pm}(k)| \pm \varepsilon_g/2}{2|\varepsilon_c^{\pm}(k)|}} e^{i\theta/2} \Phi_2(\mathbf{r}) \right]. \quad (3.2.23)$$

As should be expected, the energy spectrum (3.2.20) exactly coincides with the electron spectrum of graphene dressed by a quantized circularly polarized field in the limit of large photon occupation numbers [148]. Thus, the chief influence of circularly polarized light on the electronic properties of graphene is renormalization of energy spectrum (3.2.20), which leads to the energy gap (3.2.21) and a isotropic parabolic electron dispersion, shown in Fig. 3.3(a).

3.2.3 Dispersion and wavefunctions for linear polarization

For an dressing wave polarized linearly in the x direction, the vector potential reads

$$\mathbf{A}_l = \left(\frac{\mathcal{E}_0}{\omega} \cos \omega t, 0 \right). \quad (3.2.24)$$

Figure 3.3: Dispersion of graphene electrons dressed by light. The Fermi energy is indicated with ϵ_F . The dispersion of bare graphene is shown in dotted lines. **(a)** For circular polarization of the dressing field a gap ϵ_g opens in the dispersion, inducing an insulator transition. The Fermi surface remains a circle, with a slightly smaller radius. **(b)** For linear polarization of the dressing field the dispersion remains linear, but the Fermi velocity becomes anisotropic, decreasing in the direct perpendicular to the polarization direction. The Fermi surface becomes an ellipse.

The Hamiltonian can be rewritten as in the preceeding subsection as

$$\hat{\mathcal{H}}(t) = \hat{\mathcal{H}}_0(t) + \hat{\mathcal{H}}_{\mathbf{k}}, \quad (3.2.25)$$

where now

$$\hat{\mathcal{H}}_0(t) = -\frac{ve\mathcal{E}_0}{\omega} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \cos \omega t. \quad (3.2.26)$$

and

$$\hat{\mathcal{H}}_{\mathbf{k}} = \hbar v \begin{pmatrix} 0 & k_x - ik_y \\ k_x + ik_y & 0 \end{pmatrix}. \quad (3.2.27)$$

Again the Schrödinger equation is solved for the explicitly time-dependent part of the Hamiltonian:

$$i\hbar \frac{\partial \psi_0}{\partial t} = \hat{\mathcal{H}}_0(t) \psi_0. \quad (3.2.28)$$

Solutions are searched for in the form

$$\psi_0 = \left(A\Phi'_1(\mathbf{r}) + B\Phi'_2(\mathbf{r}) \right) \exp \left[i\alpha \frac{ve\mathcal{E}_0}{\hbar\omega^2} \sin \omega t \right], \quad (3.2.29)$$

where α , A and B are free parameters. Inserting the ansatz above into the nonstationary Schrödinger equation (3.2.28) produces coupled equations for the parameters as

$$\begin{aligned} A\alpha - B &= 0 \\ A - B\alpha &= 0. \end{aligned} \quad (3.2.30)$$

These equations give solutions if and only if

$$\begin{vmatrix} \alpha & -1 \\ 1 & -\alpha \end{vmatrix} = 0,$$

or for $\alpha = \pm 1$. The two possible solutions determine the other two parameters, subject to the normalization condition $|A|^2 + |B|^2 = 1$, as

$$\begin{aligned} A^+ &= B^+ = \frac{1}{\sqrt{2}}, \\ A^- &= -B^- = \frac{1}{\sqrt{2}}. \end{aligned} \quad (3.2.31)$$

As a result, there are two wave functions

$$\psi_0^\pm = \frac{1}{\sqrt{2}} [\Phi'_1(\mathbf{r}) \pm \Phi'_2(\mathbf{r})] \exp \left[\pm i \frac{ve\mathcal{E}_0}{\hbar\omega^2} \sin \omega t \right], \quad (3.2.32)$$

that characterize the zero-wavevector state of graphene dressed by a linearly polarized field. Since the two functions form a complete basis for a given time t , we can write the solutions to the Schrödinger equation with the full Hamiltonian (3.2.25) as a linear combination

$$\psi_{\mathbf{k}} = a^+(t)\psi_0^+ + a^-(t)\psi_0^-. \quad (3.2.33)$$

By substitution coupled equations for coefficients $a^\pm(t)$ are found as,

$$\begin{aligned} i\dot{a}^+(t) &= vk_x a^+(t) + ivk_y a^-(t) \exp \left[-i \frac{2ve\mathcal{E}_0}{\hbar\omega^2} \sin \omega t \right], \\ i\dot{a}^-(t) &= -ivk_y a^+(t) \exp \left[i \frac{2ve\mathcal{E}_0}{\hbar\omega^2} \sin \omega t \right] - vk_x a^-(t). \end{aligned} \quad (3.2.34)$$

The expansion coefficients are now written in the form

$$a^\pm(t) = e^{-i\varepsilon_{\mathbf{k}}t/\hbar} \tilde{a}^\pm(t), \quad (3.2.35)$$

where $\varepsilon_{\mathbf{k}}$ is the electron quasienergy and $\tilde{a}^\pm(t)$ are periodic functions with period $T = 2\pi/\omega$. As such they can be replaced by their Fourier series

$$\tilde{a}^\pm(t) = \sum_{n=-\infty}^{\infty} c_n^\pm e^{in\omega t}. \quad (3.2.36)$$

The exponents in the right side of Eqs. (3.2.34) can then be rewritten using the Jacobi-Anger expansion

$$e^{iz \sin \gamma} = \sum_{n=-\infty}^{\infty} J_n(z) e^{in\gamma}, \quad (3.2.37)$$

The coupled equations for the time-dependent coefficients can then be recast as algebraic equations

$$\begin{aligned} \left(vk_x - \frac{\varepsilon_{\mathbf{k}}}{\hbar} - n\omega \right) c_n^+ + ivk_y \sum_{m=-\infty}^{\infty} c_{n-m}^- J_m \left(-\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) &= 0, \\ \left(vk_x + \frac{\varepsilon_{\mathbf{k}}}{\hbar} + n\omega \right) c_n^- + ivk_y \sum_{m=-\infty}^{\infty} c_{n-m}^+ J_m \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) &= 0, \end{aligned} \quad (3.2.38)$$

where $J_m(z)$ is the Bessel function of the first kind. By considering the high-frequency case, characterized by the conditions

$$\hbar\omega \gg \varepsilon_{\mathbf{k}}, \quad \omega \gg vk, \quad (3.2.39)$$

the equation can be approximated by

$$c_n^\pm \approx i \frac{v k_y}{n\omega} \sum_{m=-\infty}^{\infty} c_{n-m}^\mp J_m \left(\mp \frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right). \quad (3.2.40)$$

Keeping in mind that $|c_n^\pm| \leq 1$ and $|J_n(z)| \leq 1$, Eq. (3.2.40) leads to the evident solution $c_{n \neq 0}^\pm \approx 0$. After substitution of this solution into Eqs. (3.2.38), the expressions (3.2.38) are reduced to the two equations

$$\begin{aligned} \left(vk_x - \frac{\varepsilon_{\mathbf{k}}}{\hbar} \right) c_0^+ + ivk_y c_0^- J_0 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) &= 0, \\ \left(vk_x + \frac{\varepsilon_{\mathbf{k}}}{\hbar} \right) c_0^- + ivk_y c_0^+ J_0 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) &= 0. \end{aligned} \quad (3.2.41)$$

Solving the above system of two equations we find both the coefficients $c_0^\pm$ and the quasienergy spectrum, as shown in Fig. 3.3(b), of the dressed states

$$\varepsilon_l^\pm(\mathbf{k}) = \varepsilon_{\mathbf{k}} = \pm \sqrt{k_x^2 + J_0^2 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) k_y^2} = \pm \hbar v k f(\theta), \quad (3.2.42)$$

where

$$f(\theta) = \sqrt{\cos^2 \theta + J_0^2 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) \sin^2 \theta}. \quad (3.2.43)$$

Effectively the Fermi velocity becomes anisotropic, with the velocity in the direction perpendicular to the dressing field polarization obtaining a factor of the Bessel function. The wavefunction of graphene dressed by a linearly polarized field can finally be expressed as

$$\begin{aligned} \psi_{l\mathbf{k}}^\pm(\mathbf{r}, t) = & \frac{e^{i(\mathbf{k} \cdot \mathbf{r} - \varepsilon_l^\pm t/\hbar)}}{\sqrt{S}} \sqrt{\frac{\cos \theta + f(\theta)}{4f(\theta)}} \left([\Phi_1(\mathbf{r}) \pm \Phi_2(\mathbf{r})] e^{\pm i(ve\mathcal{E}_0/\hbar\omega^2) \sin \omega t} \right. \\ & \left. - i \frac{\sin \theta}{\cos \theta + f(\theta)} J_0 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) [\Phi_1(\mathbf{r}) \mp \Phi_2(\mathbf{r})] e^{\mp i(ve\mathcal{E}_0/\hbar\omega^2) \sin \omega t} \right). \end{aligned} \quad (3.2.44)$$

Note that the dressed wavefunction are obtained non-perturbatively. As expected, in the absence of the irradiation ($\mathcal{E}_0 = 0$), both wavefunctions (3.2.23) and (3.2.44) exactly coincide with known wave function of unirradiated graphene. Finally, we note that the non-zero wavevector energy gaps observed in the works [150–152] do not appear here as we assumed the energy $\hbar\omega$ not to be close to any interband resonances.

3.2.4 Scattering rates and Boltzmann transport equation

To address stationary transport properties, the scattering properties of the dressed electrons should be found. We consider the interaction of the graphene electrons with a total scattering potential, $U(\mathbf{r})$, which arises from macroscopically large number of scatterers in the graphene lattice. The scattering potential is assumed to vary slowly on the scale of the graphene lattice cell size. The modified Schrödinger equation governing the scattered electron wave function Ψ reads:

$$i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r},t) = \left(\hat{\mathcal{H}}(t) + U(\mathbf{r})\right)\Psi(\mathbf{r},t), \quad (3.2.45)$$

The scattering potential is assumed to be small, and is considered as a perturbation. The scattering rate of an electron in the conduction band (band index +) from the initial state characterized by wavevector $\mathbf{k}$ to the final state $\mathbf{k}'$ can then be defined as

$$w_{\mathbf{k}'\mathbf{k}} = \frac{d}{dt} |\langle\psi_{\mathbf{k}'}^+|\Psi\rangle|^2, \quad (3.2.46)$$

where the index identifying the dressing field polarization has been dropped for simplicity, and the initial conditions of the Schrödinger equation (3.2.45) is chosen as $\Psi(\mathbf{r},0) = \psi_{\mathbf{k}}^+(\mathbf{r},0)$. According to the conventional Born scattering theory, the scattering rate can be with first-order time-dependent perturbation theory as [162]

$$w_{\mathbf{k}'\mathbf{k}} = \frac{2\pi}{\hbar} |\langle\psi_{\mathbf{k}'}^+|U(\mathbf{r})|\psi_{\mathbf{k}}^+\rangle|^2 \delta(\varepsilon_{\mathbf{k}'} - \varepsilon_{\mathbf{k}}), \quad (3.2.47)$$

where $\varepsilon_{\mathbf{k}}$ refers to the positive branches of (3.2.20, 3.2.42). As the scattering potential $U(\mathbf{r})$ is smooth over the lattice cell, we can write the scattering matrix elements

in Eq. (3.2.47) as

$$\left\langle \Phi_i(\mathbf{r}) \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{S}} \middle| U(\mathbf{r}) \middle| \Phi_j(\mathbf{r}) \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{S}} \right\rangle \approx U_{\mathbf{k}'\mathbf{k}} \delta_{ij}, \quad (3.2.48)$$

where $U_{\mathbf{k}'\mathbf{k}} = S^{-1} \int e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{r}} U(\mathbf{r}) d\mathbf{r}$ is the Fourier space representation of the potential. As a result, the scattering probability (3.2.47) takes the form

$$w_{\mathbf{k}'\mathbf{k}} = \frac{2\pi}{\hbar} |\chi_{\mathbf{k}'\mathbf{k}}|^2 |U_{\mathbf{k}'\mathbf{k}}|^2 \delta(\varepsilon_{\mathbf{k}'} - \varepsilon_{\mathbf{k}}), \quad (3.2.49)$$

where the quantity $\chi_{\mathbf{k}'\mathbf{k}}$ depends on the polarization of the dressing field. For a circularly polarized wave it reads

$$\chi_{\mathbf{k}'\mathbf{k}} = \sqrt{\frac{|\varepsilon_{\mathbf{k}'}| - \varepsilon_g/2}{2|\varepsilon_{\mathbf{k}'}|}} \sqrt{\frac{|\varepsilon_{\mathbf{k}}| - \varepsilon_g/2}{2|\varepsilon_{\mathbf{k}}|}} e^{i\frac{\theta' - \theta}{2}} + \sqrt{\frac{|\varepsilon_{\mathbf{k}'}| + \varepsilon_g/2}{2|\varepsilon_{\mathbf{k}'}|}} \sqrt{\frac{|\varepsilon_{\mathbf{k}}| + \varepsilon_g/2}{2|\varepsilon_{\mathbf{k}}|}} e^{-i\frac{\theta' - \theta}{2}}, \quad (3.2.50)$$

while for the linearly polarized one it reads

$$\begin{aligned} \chi_{\mathbf{k}'\mathbf{k}} = & \sqrt{\frac{\cos(\theta') + f(\theta')}{2f(\theta')}} \sqrt{\frac{\cos(\theta) + f(\theta)}{2f(\theta)}} \\ & \times \left[1 + \frac{\sin(\theta')}{\cos(\theta') + f(\theta')} \frac{\sin(\theta)}{\cos(\theta) + f(\theta)} J_0^2 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) \right]. \end{aligned} \quad (3.2.51)$$

Finally the potential is assumed to be of the form $|U_{\mathbf{k}'\mathbf{k}}|^2 = N_i U_0^2 / S$.

One last condition is now imposed on the dressing field frequency, that $\omega \gg 1/\tau_0$, where τ_0 is heuristic relaxation time of the graphene electrons in the absence of the dressing field. It can be seen for example in Refs. [141, 163] that under this condition the process of collisional absorption can be neglected. Thus both interband and intraband absorption of photons are forbidden, and the classical description of the field and its non-absorptive interaction with the graphene electrons remains complete. Transports properties of the dressed graphene is therefore fully described by how the dressing field alters the scattering rates and the dispersion.

Due to the form of scattering rates (3.2.49), electrons scatter only between equal

energy states. Assuming all states below the Fermi energy ε_F to be occupied, only states at the Fermi line are involved in scattering processes. Assuming positive bias in the system, i.e. positive Fermi energy, only electrons in the conductivity band enter into the conductivity calculations. We employ the conventional Boltzmann equation for relaxation time, which reads [164]

$$\frac{1}{\tau(\mathbf{k})} = \frac{S}{4\pi^2} \int \left\{ 1 - \frac{\tau(\mathbf{k}') \mathbf{E} \cdot \mathbf{v}(\mathbf{k}')}{\tau(\mathbf{k}) \mathbf{E} \cdot \mathbf{v}(\mathbf{k})} \right\} w_{\mathbf{k}'\mathbf{k}} d^2\mathbf{k}', \quad (3.2.52)$$

where S is the area of the system, $\mathbf{E}$ is a small electric field that produces current, and $\mathbf{v}(\mathbf{k}) = \nabla_{\mathbf{k}} \varepsilon_{\mathbf{k}} / \hbar$ is the electron group velocity. As already stated, the form of the scattering rates means we only calculate scattering times of electrons on the Fermi line. After calculating the wavevector dependent relaxation time the current can subsequently be found as

$$\mathbf{J} = \frac{\nu e^2}{4\pi^2} \int \mathbf{v}(\mathbf{k}) [\mathbf{E} \cdot \mathbf{v}(\mathbf{k})] \tau(\mathbf{k}) \delta(\varepsilon_{\mathbf{k}} - \varepsilon_F) d^2\mathbf{k}, \quad (3.2.53)$$

where $\nu = 4$ is the graphene spin-valley degeneracy number. Conductivity is consequently found as the ratio $\sigma_{ij} = J_i / E_j$.

Note that since the scattering rates $w_{\mathbf{k}'\mathbf{k}}$ are proportional to the delta function $\delta(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'})$, and we only calculate the relaxation time $\tau(\mathbf{k})$ for wavevectors on the Fermi line $\varepsilon_{\mathbf{k}} = \varepsilon_F$, the integrals in the right hand side of eqns. (3.2.52) and (3.2.53) are in fact both of the form

$$I = \int g(\mathbf{k}) \delta(\varepsilon_{\mathbf{k}} - \varepsilon_F) d^2\mathbf{k}. \quad (3.2.54)$$

The correct way to solve this integral, as given in Ref. [165], is to separate the integration in the direction perpendicular to constant energy surfaces of $\varepsilon_{\mathbf{k}}$, and the direction(s) along the constant energy surfaces, such that

$$\int d\mathbf{k} = \int_{\varepsilon_{\mathbf{k}} \text{ constant}} d\Sigma \int dk_{\perp}, \quad (3.2.55)$$

where $\int d\Sigma$ is a $d - 1$ dimensional integral. As $d = 2$ for graphene, this is a one

dimensional path integral over the values of $\mathbf{k}$ that give the same value for the dispersion $\varepsilon_{\mathbf{k}}$. For the component of the wavevector perpendicular to the constant energy surface we can make the change of variables $\varepsilon = \varepsilon_{\mathbf{k}}$, and we get $dk_{\perp} = d\varepsilon/|\nabla_{\mathbf{k}}\varepsilon_{\mathbf{k}}|$. The integration in the ε variable then resolves the delta function to the Fermi energy, and the integral evaluates as

$$I = \frac{1}{\hbar} \oint \frac{g(\Sigma)}{|\mathbf{v}(\Sigma)|} d\Sigma, \quad (3.2.56)$$

where the path integral goes over the Fermi line. Note the abuse of notation, where $g(\Sigma)$ is shorthand for $g(\mathbf{k})$ where $\mathbf{k}$ is on the Fermi line ($\varepsilon_{\mathbf{k}} = \varepsilon_F$), parametrized by Σ .

Again we proceed by treating separately the case of circular and linear polarization of the dressing field.

3.2.5 Conductivity for circular polarization

In the case of circular polarization of the dressing field, and Fermi energy outside the gap; $\varepsilon_F > \varepsilon_g/2$, the Fermi line remains a circle, with radius $k_F = \sqrt{\varepsilon_F^2 - (\varepsilon_g/2)^2}/\hbar v$. The integration in eq. (3.2.52) therefore goes over states with $|\mathbf{k}'| = \sqrt{k_x'^2 + k_y'^2} = k_F$. The most natural choice of integration is in circular coordinates, with $k_x' = k_F \cos \theta'$, $k_y' = k_F \sin \theta'$, with θ' integrated from 0 to 2π . The integration measure then reads $d\Sigma' = k_F d\theta'$.

Symmetry dictates that the relaxation rates be independent of the direction of the wavevector, so $\tau_c(\mathbf{k}) = \tau_c$ for all $\mathbf{k}$ on the Fermi circle. The integro-differential equation (3.2.52) then becomes a straight-forward integral. As $\mathbf{k}'$ and $\mathbf{k}$ both correspond to the Fermi energy we get from (3.2.50) that

$$|\chi_{\mathbf{k}'\mathbf{k}}|^2 = \frac{\varepsilon_F^2 + \varepsilon_g^2/4}{2\varepsilon_F^2} + \frac{\varepsilon_F^2 - \varepsilon_g^2/4}{2\varepsilon_F^2} \cos(\theta' - \theta). \quad (3.2.57)$$

From the dispersion (3.2.20) we find the group velocity on the Fermi circle as

$$\mathbf{v}(\mathbf{k}) = \frac{\nabla_{\mathbf{k}} \varepsilon_c(k)}{\hbar} = v \frac{\sqrt{\varepsilon_F^2 - \varepsilon_g^2/4}}{\varepsilon_F} \left(\cos \theta \mathbf{e}_x + \sin \theta \mathbf{e}_y \right). \quad (3.2.58)$$

The circular symmetry further implies the result is independent of the choice of probe field $\mathbf{E}$ direction, so $\mathbf{E} = E_x \mathbf{e}_x$ can be chosen to make the calculation explicit. In a similar vein we chose $\theta = 0$. Then the integral becomes

$$\begin{aligned} \frac{1}{\tau_c} &= \frac{N_i U_0^2}{4\pi \hbar^3 v^2 \varepsilon_F} \int_0^{2\pi} d\theta' \left\{ 1 - \cos \theta' \right\} \left[\varepsilon_F^2 + \varepsilon_g^2/4 + (\varepsilon_F^2 - \varepsilon_g^2/4) \cos \theta' \right] \\ &= \frac{1}{\tau_0} \frac{1}{\pi \varepsilon_F^2} 2\pi \left[\varepsilon_F^2 + \varepsilon_g^2/4 - (\varepsilon_F^2 - \varepsilon_g^2/4) \cdot \frac{1}{2} \right] = \frac{1}{\tau_0} \left(1 + \frac{3\varepsilon_g^2}{4\varepsilon_F^2} \right). \end{aligned} \quad (3.2.59)$$

Here terms proportional to $\cos \theta'$ disappeared when integrated around a full circle, and $\tau_0 = 4\hbar^3 v^2 / N_i U_0^2 \varepsilon_F$ was identified as the relaxation time of unirradiated graphene.

The current can now be calculated from the relaxation time using eq. (3.2.53) as

$$\begin{aligned} \mathbf{J} &= \frac{\nu e^2 \tau_0 \varepsilon_F}{4\pi^2 \hbar^2} \frac{1 - \varepsilon_g^2/4\varepsilon_F}{1 + 3\varepsilon_g^2/4\varepsilon_F^2} \int_0^{2\pi} d\theta \left(\cos \theta \mathbf{e}_x + \sin \theta \mathbf{e}_y \right) \mathbf{E} \cdot \left(\cos \theta \mathbf{e}_x + \sin \theta \mathbf{e}_y \right) \\ &= E_x \frac{\sigma_0}{\pi} \frac{1 - \varepsilon_g^2/4\varepsilon_F}{1 + 3\varepsilon_g^2/4\varepsilon_F^2} 2\pi \cdot \frac{1}{2} \mathbf{e}_x = E_x \sigma_0 \frac{1 - \varepsilon_g^2/4\varepsilon_F}{1 + 3\varepsilon_g^2/4\varepsilon_F^2} \mathbf{e}_x, \end{aligned} \quad (3.2.60)$$

where the term $\sin \theta \cos \theta$ integrated to zero, and $\sigma_0 = \nu e^2 \tau_0 \varepsilon_F / 4\pi \hbar^2$ is the conductivity in undressed graphene. From this expression it is seen that the off-diagonal terms σ_{ij} , $i \neq j$ of the conductivity are zero, and that the diagonal terms, shown in Fig. 3.5(a) as a function of radiation intensity, equal

$$\frac{\sigma_c}{\sigma_0} = \frac{1 - \varepsilon_g^2/4\varepsilon_F^2}{1 + 3\varepsilon_g^2/4\varepsilon_F^2}. \quad (3.2.61)$$

When the Fermi energy lies in the gap, $\varepsilon_F < \varepsilon_g/2$, the graphene acts as an insulator, and conductivity is zero.

3.2.6 Conductivity for linear polarization

In the case of linear polarization the Fermi line is an ellipse with minor axis $k_{Fx} = \varepsilon_F/\hbar v$ in k_x -direction, and major axis $k_{Fy} = [\hbar v |J_0 (2ve\varepsilon_0/\hbar\omega^2)|]^{-1}\varepsilon_F$ in k_y -direction. As such, it can be parametrized as

$$k_F(\theta) = \frac{\varepsilon_F}{\hbar v} \sqrt{\cos^2 \theta + \frac{\sin^2 \theta}{J_0^2 \left(\frac{2ve\varepsilon_0}{\hbar\omega^2} \right)}}, \quad (3.2.62)$$

with θ going from 0 to 2π [see Fig. (3.4)]. We determine the integration measure $d\Sigma$ by geometric arguments. First we observe that for infinitesimal $d\theta$ we have $k_F(\theta + d\theta) - k_F(\theta) = k'_F(\theta)d\theta$ by definition. Then from Fig. (3.4) we see that

$$d\Sigma = \sqrt{\left(k_F(\theta)d\theta\right)^2 + \left(k'_F(\theta)d\theta\right)^2} = \frac{\varepsilon_F}{\hbar v} \sqrt{\frac{\cos^2 \theta + \sin^2 \theta / J_0^4 \left(\frac{2ve\varepsilon_0}{\hbar\omega^2} \right)}{\cos^2 \theta + \sin^2 \theta / J_0^2 \left(\frac{2ve\varepsilon_0}{\hbar\omega^2} \right)}} d\theta \quad (3.2.63)$$

Now the relaxation time should be calculated from eq. (3.2.52). We only calculate the relaxation time on the Fermi line, so it can be uniquely identified by parameter θ , and we write $\tau_l(\theta)$. In performing the integration it is useful to write

Figure 3.4: The Fermi line of a graphene electron dressed by linearly polarized light is an ellipse, with the major axis in the k_y -direction. The radius is given in polar coordinates by $k_F(\theta)$ from eq. (3.2.62). The integration measure is indicated by $d\Sigma$.

in Cartesian coordinates the $\mathbf{k}$ -values as parametrized by θ :

$$k_x = \frac{\varepsilon_F}{\hbar v} \cos \theta, \quad k_y = \frac{\varepsilon_F}{\hbar v} \frac{\sin \theta}{J_0\left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2}\right)}. \quad (3.2.64)$$

Please note that the θ parametrizing the integration over the Fermi ellipse is distinct from the θ in equations (3.2.42), (3.2.43) and (3.2.51), where it is the orientation angle of the wavevector in polar coordinates. It is therefore necessary to first rewrite the equation (3.2.51) in Cartesian coordinates:

$$\chi_{\mathbf{k}'\mathbf{k}} = \sqrt{\frac{k'_x + \varepsilon_{\mathbf{k}'}/\hbar v}{2\varepsilon_{\mathbf{k}'}/\hbar v}} \sqrt{\frac{k_x + \varepsilon_{\mathbf{k}}/\hbar v}{2\varepsilon_{\mathbf{k}}/\hbar v}} \left[1 + \frac{k'_y}{k'_x + \varepsilon_{\mathbf{k}'}/\hbar v} \frac{k_y}{k_x + \varepsilon_{\mathbf{k}}/\hbar v} J_0^2\left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2}\right) \right] \quad (3.2.65)$$

As we are on the Fermi ellipse we have $\varepsilon_{\mathbf{k}} = \varepsilon_{\mathbf{k}'} = \varepsilon_F$. Now we insert from eq. (3.2.64) and get

$$|\chi_{\mathbf{k}'\mathbf{k}}|^2 = \frac{\cos \theta' + 1}{2} \frac{\cos \theta + 1}{2} \left[1 + \frac{\sin \theta'}{\cos \theta' + 1} \frac{\sin \theta}{\cos \theta + 1} \right]^2 = \cos^2 \left(\frac{\theta' - \theta}{2} \right), \quad (3.2.66)$$

where θ and θ' are parameters of integration along the Fermi ellipse. Finally, from the dispersion (3.2.42) we find the group velocity on the Fermi line as

$$\mathbf{v}(\mathbf{k}) = \frac{v}{\sqrt{k_x^2 + J_0^2\left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2}\right)}} \left(k_x, J_0^2\left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2}\right) k_y \right) = v \left(\cos \theta, J_0\left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2}\right) \sin \theta \right). \quad (3.2.67)$$

Here it can be seen directly what was already highlighted about the effect of the linearly polarized dressing field. The Fermi velocity effectively obtains a factor of the Bessel function in the direction perpendicular to the polarization direction of the field.

In calculating the relaxation time it must be distinguished between the case where the probe field $\mathbf{E}$ is parallel to the dressing field, and where it is perpendicular. In the parallel case we have $\mathbf{E} = E_x \mathbf{e}_x$. Inserting every expression above into eq. (3.2.52) and performing straightforward elementary algebra we get the dimensionless integral

equation

$$\begin{aligned} \frac{\tau_0}{\tau_{\parallel}(\theta)} = \frac{2}{\pi} \int_0^{2\pi} d\theta' \sqrt{\frac{\cos^2 \theta' + \sin^2 \theta' / J_0^4 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right)}{\cos^2 \theta' + \sin^2 \theta' / J_0^2 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right)}} \left\{ 1 - \frac{\tau_{\parallel}(\theta') \cos \theta'}{\tau_{\parallel}(\theta) \cos \theta} \right\} \cos^2 \left(\frac{\theta' - \theta}{2} \right) \\ \times \left(\cos^2 \theta' + J_0^2 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) \sin^2 \theta' \right)^{-\frac{1}{2}}. \end{aligned} \quad (3.2.68)$$

This equation must be solved numerically. In the case where the probe field is perpendicular to the dressing field, $\mathbf{E} = E_y \mathbf{e}_y$, the integral equation for $\tau_{\perp}(\theta)$ is analogous to eq. (3.2.68), and is obtained by exchanging the cosines with sines in the fraction containing the relaxation time in the integral.

To calculate the conductivity we proceed as for circular polarization, applying eq. (3.2.53). Again performing elementary algebra we find for the conductivity parallel to the dressing field that

$$\begin{aligned} \frac{\sigma_{\parallel}}{\sigma_0} = \frac{1}{\pi} \int_0^{2\pi} d\theta \sqrt{\frac{\cos^2 \theta + \sin^2 \theta / J_0^4 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right)}{\cos^2 \theta + \sin^2 \theta / J_0^2 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right)}} \times \\ \frac{\cos^2 \theta}{\sqrt{\cos^2 \theta + J_0^2 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) \sin^2 \theta}} \frac{\tau_{\parallel}(\theta)}{\tau_0}. \end{aligned} \quad (3.2.69)$$

The conductivity $\sigma_{\perp}$ in the direction perpendicular to the dressing field is obtained by replacing the product $\tau_{\parallel}(\theta) \cos^2 \theta$ with $\tau_{\perp}(\theta) J_0^2 \left(\frac{2ve\mathcal{E}_0}{\hbar\omega^2} \right) \sin^2 \theta$. These equations should again be solved numerically, using the numerical solution for $\tau(\theta)$. The conductivity is then found straightforwardly by dividing with the probe field strength. The result is shown in Fig. 3.5(b,c). Note that as circular symmetry is broken we should distinguish between the conductivity $\sigma_{xx} = J_x/E_x$ in the direction where the probe field is parallel to the polarization of the dressing field, and $\sigma_{yy} = J_y/E_y$ where the probe field is perpendicular to the dressing field.

Figure 3.5: **(a)** Conductivity of graphene dressed by circularly polarized field, as a function of dressing field irradiation intensity. The frequency of the field is $\hbar\omega = 4$ meV, and ε_F is the Fermi energy. **(b)** Conductivity of graphene dressed by field linearly polarized in the x direction, as a function of dressing field irradiation intensity. The inset shows the anisotropy of the conductivity. The unphysical values that are calculated in regions where the condition $\omega \gg vk$ is not fulfilled are shown in dashed lines. **(c)** Conductivity of graphene dressed by linearly polarized field as a function of dressing field intensity and energy. Darker unphysical areas of the plot that do not fulfill the condition $\omega \gg vk$ are surrounded by white borders.

3.3 Results and comments

The figure 3.5 constitutes the main results of this chapter. Parts (a) and (b) show the calculated conductivity depends on the irradiation intensity $I_0 = \varepsilon_0 \mathcal{E}_0^2 c/2$, for circular and linear polarization of the dressing field respectively. For circular polarization the conductivity decreases monotonically, until the irradiation induced gap energy $\varepsilon_g/2$ reaches the Fermi energy, at which point the dressed graphene becomes insulating, and conductivity goes to zero. The decrease can be understood qualitatively by looking at electron group velocity on the Fermi circle:

$$|\mathbf{v}(\mathbf{k})| = \frac{\nabla_{\mathbf{k}} \varepsilon_{\mathbf{k}}|_{k=k_F}}{\hbar} = v \frac{\sqrt{\varepsilon_F^2 - (\varepsilon_g/2)^2}}{\varepsilon_F}. \quad (3.3.1)$$

Note that the energy gap (3.2.21) increases with the intensity I_0 . As ε_g increases the group velocity decreases, until it goes to zero at $\varepsilon_g/2 = \varepsilon_F$. It follows by eq. (3.3.1) that the group velocity, and consequently the conductivity, decreases slower when the Fermi energy is higher. This is confirmed by the Fig. 3.5(a). The field energy was here chosen as $\hbar\omega = 4$ meV, corresponding to a frequency of $\omega/2\pi \approx 0.97$ THz. Such a high frequency is chosen such that the intra-band scattering condition $\omega \gg 1/\tau_0$ is satisfied.

For linear polarization, the picture is more complicated, but simultaneously richer. The chief complication is the Fermi surface. As stated earlier it is an ellipse with minor axis $k_{Fx} = \varepsilon/\hbar v$, and major axis $k_{Fy} = \varepsilon_F/(\hbar v |J_0(2ve\mathcal{E}_0)|)$. The theory developed in this section assumes in the case of linear polarization that $k = \sqrt{k_x^2 + k_y^2} \ll \omega/v$. However, as the Bessel function J_0 takes zeros, the y component of the wavevector goes to infinity in two points of the Fermi surface. As such for certain ranges of the dressing field intensity the theory becomes inaccurate. In Fig. 3.5(b) this range of inapplicability is marked with a dashed line.

The behavior of the conductivity for the case of linearly polarized dressing field is as follows. The conductivity σ_{yy} in the direction perpendicular to the dressing field polarization direction decreases in an oscillatory manner to zero, reminiscent of the absolute value of a Bessel function. Meanwhile the conductivity σ_{xx} in the direction

parallel to the dressing field, while also decreasing in an oscillatory manner, does not go to zero, at least in the range calculated. This results in a large anisotropy between the perpendicular directions, reaching up to a factor of $\sigma_{xx}/\sigma_{yy} \approx 10$ in the range of variables where the theory is valid. This can be qualitatively explained by referencing the fact that from the dispersion we see that the Fermi velocity in the x direction remains constant v , while in the y direction it acquires a factor of the Bessel function, decreasing it. This does not tell the whole story, as also the conductivity in the direction parallel to the polarization of the dressing field decreases, to a lesser degree. However, it can offer a heuristic understanding of the phenomena of conductivity anisotropy that arises.

For the preceding analysis, the dressing field energy was chosen as $\hbar\omega = 1$ meV, corresponding to a frequency of $\omega/2\pi \approx 0.24$ THz. Figure 3.5(c) shows a density plot of the conductivity in the case of linearly polarized dressing field, as a function of dressing field intensity, and also dressing field energy. The thin, darker regions where the theory is unphysical are here surrounded by white borders. The behavior of the conductivity as a function of irradiation intensity remains the same for different values of the dressing field energy, with the difference that the scale of the oscillation and decay in the intensity scales as the fourth power of the energy. This is easy to understand, as the argument of the Bessel function that enters the dispersion and scattering rates can be rewritten as $2\sqrt{2}ve\hbar/\sqrt{\varepsilon_0 c} \times \sqrt{I_0}/(\hbar\omega)^2$. So for this quantity to remain constant the dressing field intensity should be proportional to the fourth power of the dressing field energy.

Here the intuitive explanation for the modification of conductivity due to the dressing field has been observed from the modification of the dispersion relation and the group velocity derived therefrom. This is in contrast to the case of the effect of the dressing field on conductivity in 2DEG [142], where the increase in conductivity is intuitively explained from the form of the scattering matrix element. For 2DEG the quantity $\chi_{\mathbf{k}'\mathbf{k}}$ takes the form of a Bessel function, suppressing the rate of backscattering, which consequently increases conductivity [141]. The dispersion of the dressed 2DEG, as seen from the Volkov wavefunction (1.4.8), is only shifted by a fixed energy and does not contribute to the change in conductivity.

4. Conclusions

In the second chapter of this thesis the effect of nonlinearities on the dipolariton system were investigated. Multistability curves were analyzed, and the stability of the solutions investigated. Conditions of parametric instability were identified, under which mode occupations oscillated under continuous wave excitation. In particular the oscillations of indirect excitons, that correspond to oscillations of dipole moment, were argued to result in emission of electromagnetic waves in the terahertz range. The emission is superradiant, increasing as the square of the area of the device. The frequency of the emission was shown to be tunable by altering the strength of the applied electric field.

In the second section of this chapter embedding the dipolariton system in a THz cavity was considered. It was shown that the IX oscillations persist when the THz cavity is added, and emission power is enhanced. The dependence of the emission power on the quality factor Q of the THz cavity was analyzed. Optimal emission power is achieved at relatively low values of the quality factors, in the range of 15-30. A high- Q oscillating behavior was also found, imitating the amplification mechanism of a Q -switched laser. Finally, we demonstrated that by simultaneously altering the THz cavity eigenfrequency, for example by using stress to deform the cavity, the emission frequency can be tuned by the applied electric field.

In the third chapter the effect of dressing by nonresonant, low THz electromagnetic wave on stationary transport properties of graphene was investigated. First modified eigenfunctions and dispersion of the “electron+photon” system were found separately for circular and linear polarization of the dressing field. For circular polarization a gap in the dispersion is found to open at zero wavevector, inducing an

insulator transition, in agreement with earlier works. For linear polarization the dispersion remains gapless, while creating anisotropy, lowering the effective Fermi velocity in the direction perpendicular to the polarization direction.

The modified wavefunctions are then used to calculate the scattering rates and Boltzmann conductivity of the dressed electrons. Conductivity of graphene dressed by circularly polarized field is found to decrease monotonically with increased field strength, until the gap energy passes the Fermi energy, at which point it reaches zero. Conductivity of graphene dressed by linearly polarized field shows a richer behavior, where conductivity in the direction perpendicular to the polarization direction is suppressed more heavily than in the direction parallel to the polarization. The subsequent conductivity anisotropy is estimated to be of up to a factor of 10.

List of publications

Relevant to this thesis

- [A1] K. Kristinsson, O. Kyriienko, T. C. H. Liew, and I. A. Shelykh, “Continuous terahertz emission from dipolaritons”, *Phys. Rev. B* **88**, 245303 (2013).
- [A2] K. Kristinsson, O. Kyriienko, and I. A. Shelykh, “Terahertz laser based on dipolaritons”, *Phys. Rev. A* **89**, 023836 (2014).
- [A3] K. Kristinsson, O. Kibis, S. Morina, and I. A. Shelykh, “Control of electronic transport in a graphene by electromagnetic dressing”, *Sci. Rep.* **6**, 20082 (2016).

Other publications

- [A4] A. C. Seridonio, K. Kristinsson, M. de Souza, F. M. Souza, L. H. Guessi, R. S. Machado and I. A. Shelykh, “Effect of inter-adatoms correlations on the local density of states of graphene”, *EPL* **108**, 47006 (2014).
- [A5] L. H. Guessi, R. S. Machado, Y. Marques, L. S. Ricco, K. Kristinsson, M. Yoshida, I. A. Shelykh, M. de Souza, and A. C. Seridonio, “Catching the bound states in the continuum of a phantom atom in graphene”, *Phys. Rev. B* **92**, 045409 (2015).
- [A6] L. H. Guessi, Y. Marques, R. S. Machado, K. Kristinsson, L. S. Ricco, I. A. Shelykh, M. S. Figueira, M. de Souza, and A. C. Seridonio, “Quantum phase transition triggering magnetic bound states in the continuum in graphene”, *Phys. Rev. B* **92**, 245107 (2015).

Appendix A: Higher order mean field theory

This Appendix is taken directly from publication [A1].

In the following section we derive dynamical equations for the relevant expectation values to higher order mean field theory using the density matrix formalism. The higher order equations are then shown not to yield observable increase in accuracy.

The total Hamiltonian of the system can be written as a sum of terms describing coherent and incoherent processes, $\hat{H} = \hat{H}_{coh} + \hat{H}_{dec}$. The master equation for the density matrix ρ reads:

$$i\hbar \frac{\partial \rho}{\partial t} = [\hat{H}_{coh}, \rho] + \hat{\mathcal{L}}\rho, \quad (\text{A.1})$$

where $\hat{\mathcal{L}}$ is the Lindblad superoperator. [?] It accounts for dissipation processes such as non-radiative recombination of excitons, and leakage of cavity mode photons due to the non-zero transmission resonator mirrors. The explicit form of the term is

$$\begin{aligned} \hat{\mathcal{L}} = & \frac{i\gamma_C}{2}(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a) + \frac{i\gamma_{DX}}{2}(2b\rho b^\dagger - b^\dagger b\rho \\ & - \rho b^\dagger b) + \frac{i\gamma_{IX}}{2}(2c\rho c^\dagger - c^\dagger c\rho - \rho c^\dagger c). \end{aligned} \quad (\text{A.2})$$

Here a is the annihilation operator for the cavity mode (C), b is the annihilation operator of the direct exciton (DX), and c that of the indirect exciton (IX), and we omit hats of operators for simplicity. The coefficients $\gamma_i = \hbar/\tau_i$ are the decay rates of those modes ($i = C, DX, IX$).

The equation of motion for the expectation value of an operator A reads

$$i\hbar \frac{\partial \langle A \rangle}{\partial t} = i\hbar \frac{\partial}{\partial t} \text{Tr}\{A\rho\} = \langle [A, \hat{H}_{coh}] \rangle + \text{Tr}\{A\hat{\mathcal{L}}\rho\}. \quad (\text{A.3})$$

Equations of motion can be derived using simple operator algebra, and applying the mean field approximation. For illustration purposes, let us derive the terms corresponding to the nonlinear interaction part of the Hamiltonian,

$$H_{nonl} = \frac{V_{DD}}{2} b^\dagger b^\dagger b b + \frac{V_{II}}{2} c^\dagger c^\dagger c c + V_{DI} b^\dagger c^\dagger b c, \quad (\text{A.4})$$

where V_{DD} is the scattering matrix element for the interaction between two direct excitons, V_{II} is the indirect-indirect exciton interaction matrix element, and V_{DI} is the matrix element for scattering of a direct and an indirect exciton, calculated in Appendix B. Using the equation (A.3) the nonlinear part of dynamics for operator A reads

$$\{\partial_t \langle A \rangle\}_{nonl} = \frac{1}{i\hbar} \langle [A, H_{nonl}] \rangle. \quad (\text{A.5})$$

Now, all number operators of the system commute with H_{nonl} . Applying this equation to the particle numbers operators N_i , $i = C, DX, IX$ we get

$$\{\partial_t \langle N_C \rangle\}_{nonl} = \{\partial_t \langle N_{DX} \rangle\}_{nonl} = \{\partial_t \langle N_{IX} \rangle\}_{nonl} = 0. \quad (\text{A.6})$$

Let us now apply Eq. (A.5) to the correlator $\alpha = \langle a^\dagger b \rangle$:

$$\begin{aligned} \{\partial_t \alpha\}_{nonl} &= \frac{1}{2i\hbar} \langle [a^\dagger b; V_{DD} b^\dagger b^\dagger b b + V_{II} c^\dagger c^\dagger c c + 2V_{DI} b^\dagger c^\dagger b c] \rangle \\ &= \frac{1}{2i\hbar} V_{DD} \langle a^\dagger [b, b^\dagger b^\dagger] b b \rangle + \frac{1}{i\hbar} V_{DI} \langle a^\dagger [b, b^\dagger] c^\dagger b c \rangle \\ &= -\frac{i}{\hbar} \left(V_{DD} \langle a^\dagger b^\dagger b b \rangle + V_{DI} \langle a^\dagger c^\dagger b c \rangle \right) \\ &\approx -\frac{i}{\hbar} \left(V_{DD} \langle a^\dagger b \rangle \langle b^\dagger b \rangle + V_{DI} \langle a^\dagger b \rangle \langle c^\dagger c \rangle \right) \\ &= -\frac{i}{\hbar} \left(V_{DD} N_{DX} + V_{DI} N_{IX} \right) \alpha. \end{aligned} \quad (\text{A.7})$$

In the second to last step we used second order mean field theory to close the

system of equations. Equations of motion for correlators $\beta = \langle b^\dagger c \rangle$ and $\gamma = \langle c^\dagger a \rangle$ can similarly be calculated as

$$\{\partial_t \beta\}_{nonl} = \frac{i}{\hbar} \left((V_{DD} - V_{DI})N_{DX} + (V_{DI} - V_{II})N_{IX} \right) \beta \quad (\text{A.8})$$

$$\{\partial_t \gamma\}_{nonl} = \frac{i}{\hbar} \left(V_{DI}N_{DX} + V_{II}N_{IX} \right) \gamma. \quad (\text{A.9})$$

Next, we derive the dynamic equations for expectation values of the annihilation operators. Inserting the operator b in Eq. (A.5) we get

$$\begin{aligned} \{\partial_t \langle b \rangle\}_{nonl} &= \frac{1}{2i\hbar} \langle [b; V_{DD}b^\dagger b^\dagger bb + V_{II}c^\dagger c^\dagger cc + 2V_{DI}b^\dagger c^\dagger bc] \rangle \\ &= \frac{1}{2i\hbar} V_{DD} \langle [b, b^\dagger b^\dagger] bb \rangle + \frac{1}{i\hbar} V_{DI} \langle [b, b^\dagger] c^\dagger bc \rangle \\ &= -\frac{i}{\hbar} \left(V_{DD} \langle b^\dagger bb \rangle + V_{DI} \langle c^\dagger bc \rangle \right) \\ &\approx -\frac{i}{\hbar} \left(V_{DD} \langle b \rangle \langle b^\dagger b \rangle + V_{DI} \langle b \rangle \langle c^\dagger c \rangle \right) \\ &= -\frac{i}{\hbar} \left(V_{DD}N_{DX} + V_{DI}N_{IX} \right) \langle b \rangle, \end{aligned} \quad (\text{A.10})$$

and in the same way derive equation for c ,

$$\{\partial_t \langle c \rangle\}_{nonl} = -\frac{i}{\hbar} \left(V_{DI}N_{DX} + V_{II}N_{IX} \right) \langle c \rangle. \quad (\text{A.11})$$

Note that creation or annihilation operators for the cavity mode does not appear in H_{nonl} , and

$$\{\partial_t \langle a \rangle\}_{nonl} = 0. \quad (\text{A.12})$$

Repeating this procedure for the other terms in the Hamiltonian leads to a closed set of equations. We perform a change of variables $a_i \rightarrow e^{-i\omega_c t} a_i$, $i = C, DX, IX$. This preserves particle numbers and correlators, and removes reference to absolute mode energies. Equations can then be expressed using relative energies $\delta_\Omega = \omega_C -$

ω_{DX} , $\delta_J = \omega_{IX} - \omega_{DX}$, and read

$$\partial_t N_C = \Omega \text{Im}(\alpha) - \frac{2}{\hbar} \text{Im}(\tilde{P}(t)^* \langle a \rangle) - \frac{1}{\tau_C} N_C, \quad (\text{A.13})$$

$$\partial_t N_{DX} = -\Omega \text{Im}(\alpha) - J \text{Im}(\beta) - \frac{1}{\tau_{DX}} N_{DX}, \quad (\text{A.14})$$

$$\partial_t N_{IX} = J \text{Im}(\beta) - \frac{1}{\tau_{IX}} N_{IX}, \quad (\text{A.15})$$

$$\partial_t \alpha = i\delta_\Omega \alpha + i\frac{\Omega}{2}(N_{DX} - N_C) + i\frac{J}{2}\gamma^* - \frac{i}{\hbar}(V_{DD}N_{DX} + V_{DI}N_{IX})\alpha \quad (\text{A.16})$$

$$+ i\tilde{P}(t)^* \langle b \rangle - \frac{1}{2} \left(\frac{1}{\tau_C} + \frac{1}{\tau_{DX}} \right) \alpha, \quad (\text{A.17})$$

$$\partial_t \beta = -i\delta_J \beta + i\frac{\Omega}{2}\gamma^* + i\frac{J}{2}(N_{DX} - N_{IX}) - \frac{1}{2} \left(\frac{1}{\tau_{DX}} + \frac{1}{\tau_{IX}} \right) \beta \quad (\text{A.18})$$

$$+ \frac{i}{\hbar} \left[(V_{DD} - V_{DI})N_{DX} + (V_{DI} - V_{II})N_{IX} \right] \beta, \quad (\text{A.19})$$

$$\partial_t \gamma = i(\delta_J - \delta_\Omega)\gamma - i\frac{\Omega}{2}\beta^* - i\frac{J}{2}\alpha^* + \frac{i}{\hbar}(V_{II}N_{IX} + V_{DI}N_{DX})\gamma \quad (\text{A.20})$$

$$- i\tilde{P}(t)\langle c \rangle^* - \frac{1}{2} \left(\frac{1}{\tau_{IX}} + \frac{1}{\tau_C} \right) \gamma, \quad (\text{A.21})$$

$$\partial_t \langle a \rangle = -i\frac{\Omega}{2}\langle b \rangle - i\tilde{P}(t) - \frac{1}{2\tau_C} \langle a \rangle, \quad (\text{A.22})$$

$$\partial_t \langle b \rangle = i\delta_\Omega \langle b \rangle - i\frac{\Omega}{2}\langle a \rangle + i\frac{J}{2}\langle c \rangle - \frac{i}{\hbar}(V_{DD}N_{DX} + V_{DI}N_{IX})\langle b \rangle - \frac{1}{2\tau_{DX}} \langle b \rangle, \quad (\text{A.23})$$

$$\partial_t \langle c \rangle = i(\delta_\Omega - \delta_J)\langle c \rangle + i\frac{J}{2}\langle b \rangle - \frac{i}{\hbar}(V_{II}N_{IX} + V_{DI}N_{DX})\langle c \rangle - \frac{1}{2\tau_{IX}} \langle c \rangle, \quad (\text{A.24})$$

where $\tilde{P}(t) = e^{i\omega_C t} P(t)$.

For accuracy comparison, numerical modelling was done in two ways. First the system of the nine coupled equations written for second order mean-field approximation were solved for certain conditions. Second the three first order equations for expectation values $\langle a \rangle$, $\langle b \rangle$ and $\langle c \rangle$ were solved for the same conditions, setting $N_i = |\langle a_i \rangle|^2$. The latter correspond to Eqns. (2.1.7)–(2.1.9) in the main text. The results of this comparison is shown in Fig. A.1. The second order theory is shown only to yield observable increase in accuracy for pumping strengths many orders of magnitude smaller than the ones treated in the paper. For this reason the first order theory was chosen to be presented in the paper, and used for obtaining the results

shown.

Figure A.1: Relative difference between stable population numbers, obtained from equations (2.1.7)-(2.1.9) and equations (A.13)-(A.24), as a function of pumping strength. The scale of the pumping strength is relative to a typical strength $|P_0|^2 = 10^{29} \text{ s}^{-1}$. The difference is negligible at the typical pumping strength. The inset shows a comparison of solutions using the first and second order mean field theory, for a short turning on time of the CW pump. Even at large times there is no observable difference in the solutions, confirming there is no increase in accuracy.

Appendix B: DX-IX interaction constant

This Appendix is taken directly from publication [A1].

The calculation of the matrix element of the interaction between the direct and indirect exciton follows the derivation of IX-IX interaction constant performed in Ref. [157]. However, the difference comes from difference of wave function of interacting particles corresponding to direct and indirect exciton, and distinct geometry of the Coulomb interaction between carriers. The matrix element can be written as the sum $V_{DX-IX} = V_{DX-IX}^{dir} + V_{DX-IX}^{X-exch} + V_{DX-IX}^{e-exch} + V_{DX-IX}^{h-exch}$, where

$$V_{DX-IX}^{dir}(\mathbf{Q}, \mathbf{Q}', \mathbf{q}) = \int d^2\mathbf{r}_e d^2\mathbf{r}_h d^2\mathbf{r}_{e'} d^2\mathbf{r}_{h'} \Psi_{\mathbf{Q}}^{DX}(\mathbf{r}_e, \mathbf{r}_h)^* \Psi_{\mathbf{Q}'}^{IX}(\mathbf{r}_{e'}, \mathbf{r}_{h'})^* \\ \times V_I(\mathbf{r}_e, \mathbf{r}_h, \mathbf{r}_{e'}, \mathbf{r}_{h'}) \Psi_{\mathbf{Q}+\mathbf{q}}^{DX}(\mathbf{r}_e, \mathbf{r}_h) \Psi_{\mathbf{Q}'-\mathbf{q}}^{IX}(\mathbf{r}_{e'}, \mathbf{r}_{h'}), \quad (\text{B.1})$$

$$V_{DX-IX}^{X-exch}(\mathbf{Q}, \mathbf{Q}', \mathbf{q}) = \int d^2\mathbf{r}_e d^2\mathbf{r}_h d^2\mathbf{r}_{e'} d^2\mathbf{r}_{h'} \Psi_{\mathbf{Q}}^{DX}(\mathbf{r}_e, \mathbf{r}_h)^* \Psi_{\mathbf{Q}'}^{IX}(\mathbf{r}_{e'}, \mathbf{r}_{h'})^* \\ \times V_I(\mathbf{r}_e, \mathbf{r}_h, \mathbf{r}_{e'}, \mathbf{r}_{h'}) \Psi_{\mathbf{Q}+\mathbf{q}}^{DX}(\mathbf{r}_{e'}, \mathbf{r}_{h'}) \Psi_{\mathbf{Q}'-\mathbf{q}}^{IX}(\mathbf{r}_e, \mathbf{r}_h), \quad (\text{B.2})$$

$$V_{DX-IX}^{e-exch}(\mathbf{Q}, \mathbf{Q}', \mathbf{q}) = - \int d^2\mathbf{r}_e d^2\mathbf{r}_h d^2\mathbf{r}_{e'} d^2\mathbf{r}_{h'} \Psi_{\mathbf{Q}}^{DX}(\mathbf{r}_e, \mathbf{r}_h)^* \Psi_{\mathbf{Q}'}^{IX}(\mathbf{r}_{e'}, \mathbf{r}_{h'})^* \\ \times V_I(\mathbf{r}_e, \mathbf{r}_h, \mathbf{r}_{e'}, \mathbf{r}_{h'}) \Psi_{\mathbf{Q}+\mathbf{q}}^{DX}(\mathbf{r}_{e'}, \mathbf{r}_h) \Psi_{\mathbf{Q}'-\mathbf{q}}^{IX}(\mathbf{r}_e, \mathbf{r}_{h'}), \quad (\text{B.3})$$

$$V_{DX-IX}^{h-exch}(\mathbf{Q}, \mathbf{Q}', \mathbf{q}) = - \int d^2\mathbf{r}_e d^2\mathbf{r}_h d^2\mathbf{r}_{e'} d^2\mathbf{r}_{h'} \Psi_{\mathbf{Q}}^{DX}(\mathbf{r}_e, \mathbf{r}_h)^* \Psi_{\mathbf{Q}'}^{IX}(\mathbf{r}_{e'}, \mathbf{r}_{h'})^* \\ \times V_I(\mathbf{r}_e, \mathbf{r}_h, \mathbf{r}_{e'}, \mathbf{r}_{h'}) \Psi_{\mathbf{Q}+\mathbf{q}}^{DX}(\mathbf{r}_e, \mathbf{r}_{h'}) \Psi_{\mathbf{Q}'-\mathbf{q}}^{IX}(\mathbf{r}_{e'}, \mathbf{r}_h). \quad (\text{B.4})$$

Here V_I is the Coulomb interaction between one direct exciton (e, h) and one

indirect exciton (e', h') . It can be expressed as

$$V_I(\mathbf{r}_e, \mathbf{r}_h, \mathbf{r}_{e'}, \mathbf{r}_{h'}) = \frac{e^2}{4\pi\epsilon\epsilon_0} \left[\frac{1}{\sqrt{(\mathbf{r}_e - \mathbf{r}_{e'})^2 + L^2}} + \frac{1}{|\mathbf{r}_h - \mathbf{r}_{h'}|} - \frac{1}{|\mathbf{r}_e - \mathbf{r}_{h'}|} - \frac{1}{\sqrt{(\mathbf{r}_h - \mathbf{r}_{e'})^2 + L^2}} \right],$$

where L is a separation distance between centers of coupled QWs and we used the narrow QW approximation.

Going to the center of mass coordinates of the excitons we set $\mathbf{R} = \beta_e \mathbf{r}^e + \beta_h \mathbf{r}^h$, and $\mathbf{r} = \mathbf{r}^e - \mathbf{r}^h$, where $\beta_e = m_e/(m_e + m_h)$, $\beta_h = m_h/(m_e + m_h)$. Separating the center of mass motion and the relative motion, we can write

$$\Psi_{\mathbf{Q}}^{DX/IX}(\mathbf{r}_e, \mathbf{r}_h) = \frac{1}{\sqrt{A}} e^{i\mathbf{Q}\cdot\mathbf{R}} \phi_{DX/IX}(\mathbf{r}), \quad (\text{B.5})$$

where $\mathbf{Q}$ corresponds to the center-of-mass momentum of the exciton. The relative motion part of wave function $\phi(r)$ can be written using the variational procedure, where trial functions can be represented in several different forms, for example [167]

$$\phi_1(\mathbf{r}) = \sqrt{\frac{2}{\pi}} \frac{1}{a} \exp\left(-\frac{|\mathbf{r}|}{a}\right), \quad (\text{B.6})$$

$$\phi_2(\mathbf{r}) = \frac{1}{\sqrt{2\pi b(b+l)}} \exp\left(-\frac{\sqrt{r^2 + l^2} - l}{2b}\right). \quad (\text{B.7})$$

Here a and $2b$ are quantities associated with indirect exciton Bohr radii and l is a variational parameter reminiscent of the separation distance between the quantum wells.

Direct DX-IX interaction. First we calculate the matrix element for the direct interaction. We choose the second trial function, setting

$$\phi_{DX}(\mathbf{r}) = \frac{1}{\sqrt{2\pi b_D(b_D + l_D)}} \exp\left(-\frac{\sqrt{r^2 + l_D^2} - l_D}{2b_D}\right), \quad (\text{B.8})$$

$$\phi_{IX}(\mathbf{r}) = \frac{1}{\sqrt{2\pi b_I(b_I + l_I)}} \exp\left(-\frac{\sqrt{r^2 + l_I^2} - l_I}{2b_I}\right). \quad (\text{B.9})$$

Dropping the index $DX - IX$ for the sake of convenience, the integral (B.1) reads

$$\begin{aligned}
V_{dir}(\mathbf{Q}, \mathbf{Q}', \mathbf{q}) &= \frac{1}{S^2} \frac{\exp(l_D/b_D)}{2\pi b_D(b_D + l_D)} \frac{\exp(l_I/b_I)}{2\pi b_I(b_I + l_I)} \int d^2\mathbf{r}_e d^2\mathbf{r}_h d^2\mathbf{r}_{e'} d^2\mathbf{r}_{h'} \\
&\times V_I(\mathbf{r}_e, \mathbf{r}_h, \mathbf{r}_{e'}, \mathbf{r}_{h'}) \exp[-i\mathbf{Q}(\beta_e \mathbf{r}_e + \beta_h \mathbf{r}_h) - i\mathbf{Q}'(\beta_e \mathbf{r}_{e'} + \beta_h \mathbf{r}_{h'}) \\
&+ i(\mathbf{Q} + \mathbf{q})(\beta_e \mathbf{r}_e + \beta_h \mathbf{r}_h) + i(\mathbf{Q}' - \mathbf{q})(\beta_e \mathbf{r}_{e'} + \beta_h \mathbf{r}_{h'})] \\
&\times \exp\left(-\frac{\sqrt{(\mathbf{r}_e - \mathbf{r}_h)^2 + l_D^2}}{b_D}\right) \exp\left(-\frac{\sqrt{(\mathbf{r}_{e'} - \mathbf{r}_{h'})^2 + l_I^2}}{b_I}\right). \quad (\text{B.10})
\end{aligned}$$

Rewriting the integral using center-of-mass coordinates for both excitons, with $\mathbf{R} = \beta_e \mathbf{r}_e + \beta_h \mathbf{r}_h$, $\mathbf{R}' = \beta_e \mathbf{r}_{e'} + \beta_h \mathbf{r}_{h'}$, $\mathbf{r} = \mathbf{r}_e - \mathbf{r}_h$ and $\mathbf{r}' = \mathbf{r}_{e'} - \mathbf{r}_{h'}$, and defining the constant $C = \frac{e^2}{4\pi\epsilon_0 A} \frac{\exp(l_D/b_D)}{2\pi b_D(b_D + l_D)} \frac{\exp(l_I/b_I)}{2\pi b_I(b_I + l_I)}$, we get

$$\begin{aligned}
V_{dir}(\mathbf{q}) &= \frac{C}{S} \int d^2\mathbf{r} d^2\mathbf{r}' d^2\mathbf{R} d^2\mathbf{R}' \exp\left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D}\right) \exp\left(-\frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I}\right) \\
&\times \exp[i\mathbf{q} \cdot (\mathbf{R} - \mathbf{R}')] \left[\frac{1}{\sqrt{(\beta_h(\mathbf{r} - \mathbf{r}') + \mathbf{R} - \mathbf{R}')^2 + L^2}} + \frac{1}{|-\beta_e(\mathbf{r} - \mathbf{r}') + \mathbf{R} - \mathbf{R}'|} \right. \\
&\left. - \frac{1}{|\beta_h \mathbf{r} + \beta_e \mathbf{r}' + \mathbf{R} - \mathbf{R}'|} - \frac{1}{\sqrt{(-\beta_e \mathbf{r} - \beta_h \mathbf{r}' + \mathbf{R} - \mathbf{R}')^2 + L^2}} \right], \quad (\text{B.11})
\end{aligned}$$

where one can note that complex exponents with $\mathbf{Q}$ and $\mathbf{Q}'$ cancel each other. It is convenient to use the substitutions $\boldsymbol{\xi} = \mathbf{R} - \mathbf{R}'$, $\boldsymbol{\sigma} = (\mathbf{R} + \mathbf{R}')/2$. Then the integration over $\boldsymbol{\sigma}$ yields the area S of the system. The integral (B.11) rewritten in a new variables reads

$$\begin{aligned}
V_{dir}(\mathbf{q}) &= C \int d^2\boldsymbol{\xi} d^2\mathbf{r} d^2\mathbf{r}' \exp\left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D}\right) \exp\left(-\frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I}\right) \exp[i\mathbf{q} \cdot \boldsymbol{\xi}] \\
&\times \left[\frac{1}{\sqrt{(\boldsymbol{\xi} + \beta_h(\mathbf{r} - \mathbf{r}'))^2 + L^2}} + \frac{1}{|\boldsymbol{\xi} - \beta_e(\mathbf{r} - \mathbf{r}')|} \right. \\
&\left. - \frac{1}{|\boldsymbol{\xi} + \beta_h \mathbf{r} + \beta_e \mathbf{r}'|} - \frac{1}{\sqrt{(\boldsymbol{\xi} - \beta_e \mathbf{r} - \beta_h \mathbf{r}')^2 + L^2}} \right]. \quad (\text{B.12})
\end{aligned}$$

The expression (B.12) represents a sum of four integrals for electron-electron, hole-

hole and electron-hole mixed interaction,

$$V_{dir} = \left[\mathcal{J}_{ee'} + \mathcal{J}_{hh'} + \mathcal{J}_{eh'} + \mathcal{J}_{he'} \right]. \quad (\text{B.13})$$

These integrals can be calculated separately using the standard integrals

$$\int_0^{2\pi} e^{ix \sin \theta} d\theta = 2\pi J_0(x), \quad \int_0^\infty \frac{x}{\sqrt{a^2 + x^2}} J_0(bx) dx = \frac{e^{-ab}}{b}.$$

We start with $\mathcal{J}_{ee'}$, and perform the change of variables $\boldsymbol{\chi} = \boldsymbol{\xi} + \beta_h(\mathbf{r} - \mathbf{r}')$:

$$\begin{aligned} \frac{\mathcal{J}_{ee'}(\mathbf{q})}{C} &= \int d^2\mathbf{r} d^2\mathbf{r}' d^2\boldsymbol{\xi} \exp\left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I}\right) \frac{\exp[i\mathbf{q} \cdot \boldsymbol{\xi}]}{\sqrt{(\boldsymbol{\xi} + \beta_h(\mathbf{r} - \mathbf{r}'))^2 + L^2}} \\ &= \int d^2\mathbf{r} d^2\mathbf{r}' d^2\boldsymbol{\chi} \exp\left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I}\right) \frac{\exp[i\mathbf{q} \cdot \boldsymbol{\chi} - i\beta_h\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}')] }{\sqrt{\boldsymbol{\chi}^2 + L^2}} \\ &= \int d^2\mathbf{r} d^2\mathbf{r}' \exp\left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I}\right) \exp[-i\beta_h\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}')] \\ &\quad \times \int_0^\infty d\chi \frac{\chi}{\sqrt{\chi^2 + L^2}} \int_0^{2\pi} d\theta e^{iq\chi \cos \theta} \\ &= 2\pi \int d^2\mathbf{r} d^2\mathbf{r}' \exp\left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I}\right) \exp[-i\beta_h\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}')] \\ &\quad \times \int_0^\infty d\chi \frac{\chi}{\sqrt{\chi^2 + L^2}} J_0(q\chi) \\ &= 2\pi \frac{\exp(-qL)}{q} \int d^2\mathbf{r} \exp\left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D}\right) \exp[-i\beta_h\mathbf{q} \cdot \mathbf{r}] \\ &\quad \times \int d^2\mathbf{r}' \exp\left(-\frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I}\right) \exp[i\beta_h\mathbf{q} \cdot \mathbf{r}'] \\ &= 2\pi \frac{\exp(-qL)}{q} I_D(\mathbf{q}) I_I(\mathbf{q}). \end{aligned} \quad (\text{B.14})$$

The integral is decomposed into a product of two integrals, which are evaluated as

$$\begin{aligned}
I_{D/I}(\mathbf{q}) &= \int_0^\infty dr \cdot r \exp\left(-\frac{\sqrt{r^2 + l_{D/I}^2}}{b_{D/I}}\right) \int_0^{2\pi} d\theta e^{\mp i\beta_h q r \cos\theta} \\
&= 2\pi \int_0^\infty dr \cdot r \exp\left(-\frac{\sqrt{r^2 + l_{D/I}^2}}{b_{D/I}}\right) J_0(\beta_h q r). \tag{B.15}
\end{aligned}$$

It is not possible to calculate integral (B.15) analytically in the general case, but we are interested in $q \rightarrow 0$ limit. In this limit the zeroeth order Bessel function equals one. We make the change of variables $x = \sqrt{r^2 + r_0^2}$ and integrate by parts:

$$\begin{aligned}
I_{D/I}(\mathbf{q} \rightarrow 0) &= 2\pi \int_0^\infty dr \cdot r \exp\left(-\frac{\sqrt{r^2 + l_{D/I}^2}}{b_{D/I}}\right) = 2\pi \int_{l_{D/I}}^\infty dx \cdot x \exp\left(-\frac{x}{b_{D/I}}\right) \\
&= 2\pi \exp\left(-\frac{l_{D/I}}{b_{D/I}}\right) b_{D/I} (b_{D/I} + l_{D/I}). \tag{B.16}
\end{aligned}$$

Finally, the integral $\mathcal{J}_{ee'}$ in the $\mathbf{q} \rightarrow 0$ reads

$$\begin{aligned}
\mathcal{J}_{ee'}^{\mathbf{q} \rightarrow 0} &= 2\pi \frac{e^{-qL}}{q} \frac{e^2}{4\pi\epsilon\epsilon_0 S} \frac{\exp(l_D/b_D)}{2\pi b_D(b_D + l_D)} \frac{\exp(l_I/b_I)}{2\pi b_I(b_I + l_I)} (2\pi)^2 \exp\left(-\frac{l_I}{b_I} - \frac{l_D}{b_D}\right) \\
&\quad \times b_D(b_D + l_D) b_I(b_I + l_I) \\
&= \frac{e^2}{2\epsilon\epsilon_0 S} \frac{e^{-qL}}{q}. \tag{B.17}
\end{aligned}$$

The three remaining integrals can be calculated in the same way, using different change of variables. For the integral $\mathcal{J}_{hh'}$ we employ the substitution $\chi = \xi - \beta_e(\mathbf{r} - \mathbf{r}')$, yielding the result

$$\mathcal{J}_{hh'}^{\mathbf{q} \rightarrow 0} = \frac{e^2}{2\epsilon\epsilon_0 S} \frac{1}{q}.$$

The integral $\mathcal{J}_{eh'}$, using $\chi = \xi + \beta_h \mathbf{r} + \beta_e \mathbf{r}'$, results in

$$\mathcal{J}_{eh'}^{\mathbf{q} \rightarrow 0} = -\frac{e^2}{2\epsilon\epsilon_0 S} \frac{1}{q},$$

and $\mathcal{J}_{he'}$ calculated with $\chi = \xi - \beta_e \mathbf{r} - \beta_h \mathbf{r}'$ gives

$$\mathcal{J}_{he'}^{\mathbf{q} \rightarrow 0} = -\frac{e^2}{2\epsilon\epsilon_0 S} \frac{e^{-qL}}{q}.$$

Finally, the sum of the four integrals is

$$V_{dir}^{q \rightarrow 0} = \frac{e^2}{2\epsilon\epsilon_0 S} \left[\frac{e^{-qL}}{q} + \frac{1}{q} - \frac{1}{q} - \frac{e^{-qL}}{q} \right] = 0. \quad (\text{B.18})$$

This corresponds to the fact that *direct interaction between direct and indirect exciton vanishes* in $q \rightarrow 0$ limit, on the contrary to IX-IX case, where dipole-dipole interaction gives the main contribution. [157, 168]

Exciton exchange DX-IX interaction. The interaction matrix element due to simultaneous exchange of electron and hole between two excitons, which is referred as exciton exchange V_X^{exh} , can be calculated using same trial functions as for the direct matrix element. It reads

$$\begin{aligned}
V_X^{exch}(\mathbf{Q}, \mathbf{Q}', \mathbf{q}) &= \frac{1}{S^2} \frac{\exp(l_D/b_D)}{2\pi b_D(b_D + l_D)} \frac{\exp(l_I/b_I)}{2\pi b_I(b_I + l_I)} \int d^2\mathbf{r}_e d^2\mathbf{r}_h d^2\mathbf{r}_{e'} d^2\mathbf{r}_{h'} \\
&\times V_I(\mathbf{r}_e, \mathbf{r}_h, \mathbf{r}_{e'}, \mathbf{r}_{h'}) \exp[-i\mathbf{Q}(\beta_e \mathbf{r}_e + \beta_h \mathbf{r}_h) - i\mathbf{Q}'(\beta_e \mathbf{r}_{e'} + \beta_h \mathbf{r}_{h'}) \\
&\quad + i(\mathbf{Q} + \mathbf{q})(\beta_e \mathbf{r}_{e'} + \beta_h \mathbf{r}_{h'}) + i(\mathbf{Q}' - \mathbf{q})(\beta_e \mathbf{r}_e + \beta_h \mathbf{r}_h)] \\
&\times \exp\left(-\frac{\sqrt{(\mathbf{r}_e - \mathbf{r}_h)^2 + l_D^2}}{b_D} - \frac{\sqrt{(\mathbf{r}_{e'} - \mathbf{r}_{h'})^2 + l_I^2}}{b_I} \right. \\
&\quad \left. - \frac{\sqrt{(\mathbf{r}_{e'} - \mathbf{r}_{h'})^2 + l_D^2}}{b_D} - \frac{\sqrt{(\mathbf{r}_e - \mathbf{r}_h)^2 + l_I^2}}{b_I}\right) \\
&= C \int d^2\boldsymbol{\xi} d^2\mathbf{r} d^2\mathbf{r}' \exp\left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I} - \frac{\sqrt{\mathbf{r}^2 + l_I^2}}{b_I} \right. \\
&\quad \left. - \frac{\sqrt{\mathbf{r}'^2 + l_D^2}}{b_D}\right) \exp[-i(\mathbf{q} + \Delta\mathbf{Q}) \cdot \boldsymbol{\xi}] \left[\frac{1}{\sqrt{(\boldsymbol{\xi} + \beta_h(\mathbf{r} - \mathbf{r}'))^2 + L^2}} \right. \\
&\quad \left. + \frac{1}{|\boldsymbol{\xi} - \beta_e(\mathbf{r} - \mathbf{r}')|} - \frac{1}{|\boldsymbol{\xi} + \beta_h \mathbf{r} + \beta_e \mathbf{r}'|} - \frac{1}{\sqrt{(\boldsymbol{\xi} - \beta_e \mathbf{r} - \beta_h \mathbf{r}')^2 + L^2}} \right],
\end{aligned} \tag{B.19}$$

where $C = \frac{e^2}{4\pi\epsilon\epsilon_0 S} \frac{\exp(l_D/b_D)}{2\pi b_D(b_D + l_D)} \frac{\exp(l_I/b_I)}{2\pi b_I(b_I + l_I)}$ and $\boldsymbol{\xi} = \mathbf{R} - \mathbf{R}'$. Additionally, we defined exchanged momentum between excitons as $\Delta\mathbf{Q} = \mathbf{Q} - \mathbf{Q}'$. We are interested in small values of $\Delta\mathbf{Q}$, taking the limit as it approaches zero. It is therefore convenient to define momentum $\mathbf{K} = \Delta\mathbf{Q} + \mathbf{q}$, and study its long wavelength limit.

We proceed in the similar way as with the direct interaction, separating the integral into four parts for different interaction terms:

$$V_X^{exch}(\mathbf{K}) = [\mathcal{J}'_{ee'} + \mathcal{J}'_{hh'} + \mathcal{J}'_{eh'} + \mathcal{J}'_{he'}].$$

We start by calculating $\mathcal{J}'_{ee'}$, using the same change of variables $\boldsymbol{\chi} = \boldsymbol{\xi} + \beta_h(\mathbf{r} - \mathbf{r}')$

as before:

$$\begin{aligned}
\mathcal{J}'_{ee'} &= C \int d^2 \boldsymbol{\xi} d^2 \mathbf{r} d^2 \mathbf{r}' \exp \left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I} - \frac{\sqrt{\mathbf{r}^2 + l_I^2}}{b_I} - \frac{\sqrt{\mathbf{r}'^2 + l_D^2}}{b_D} \right) \\
&\quad \times e^{-i\mathbf{K} \cdot \boldsymbol{\xi}} \frac{1}{\sqrt{(\boldsymbol{\xi} + \beta_h(\mathbf{r} - \mathbf{r}'))^2 + L^2}} \\
&= C \int d^2 \boldsymbol{\chi} d^2 \mathbf{r} d^2 \mathbf{r}' \exp \left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I} - \frac{\sqrt{\mathbf{r}^2 + l_I^2}}{b_I} - \frac{\sqrt{\mathbf{r}'^2 + l_D^2}}{b_D} \right) \\
&\quad \times \exp[-i\mathbf{K} \cdot \boldsymbol{\chi}] \exp[i\beta_h \mathbf{K} \cdot (\mathbf{r} - \mathbf{r}')] \frac{1}{\sqrt{\boldsymbol{\chi}^2 + L^2}} \\
&= C \int d^2 \mathbf{r} d^2 \mathbf{r}' \exp \left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I} - \frac{\sqrt{\mathbf{r}^2 + l_I^2}}{b_I} - \frac{\sqrt{\mathbf{r}'^2 + l_D^2}}{b_D} \right) \\
&\quad \times \exp[i\beta_h \mathbf{K} \cdot (\mathbf{r} - \mathbf{r}')] \int_0^\infty d\chi \frac{\chi}{\sqrt{\chi^2 + L^2}} \int_0^{2\pi} d\theta \exp(-i\beta_h K \chi \cos \theta) \\
&= 2\pi C \int d^2 \mathbf{r} d^2 \mathbf{r}' \exp \left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I} - \frac{\sqrt{\mathbf{r}^2 + l_I^2}}{b_I} - \frac{\sqrt{\mathbf{r}'^2 + l_D^2}}{b_D} \right) \\
&\quad \times \exp[i\beta_h \mathbf{K} \cdot (\mathbf{r} - \mathbf{r}')] \int_0^\infty d\chi \frac{\chi}{\sqrt{\chi^2 + L^2}} J_0(\beta_h K \chi) \\
&= 2\pi C \frac{e^{-KL}}{K} \int d^2 \mathbf{r} e^{i\beta_h \mathbf{K} \cdot \mathbf{r}} \exp \left(-\frac{\sqrt{\mathbf{r}^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}^2 + l_I^2}}{b_I} \right) \\
&\quad \times \int d^2 \mathbf{r}' e^{-i\beta_h \mathbf{K} \cdot \mathbf{r}'} \exp \left(-\frac{\sqrt{\mathbf{r}'^2 + l_D^2}}{b_D} - \frac{\sqrt{\mathbf{r}'^2 + l_I^2}}{b_I} \right) \\
&= 2\pi C \frac{e^{-KL}}{K} I_+(\mathbf{K}) I_-(\mathbf{K}), \tag{B.20}
\end{aligned}$$

and we have written the total integral as a product of two integrals, which can be written as

$$\begin{aligned}
I_{+/-}(\mathbf{K}) &= \int_0^\infty dr r \exp \left(-\frac{\sqrt{r^2 + l_D^2}}{b_D} \right) \exp \left(-\frac{\sqrt{r^2 + l_I^2}}{b_I} \right) \int_0^{2\pi} d\theta e^{\pm i\beta_h K r \cos \theta} \\
&= 2\pi \int_0^\infty dr r \exp \left(-\frac{\sqrt{r^2 + l_D^2}}{b_D} \right) \exp \left(-\frac{\sqrt{r^2 + l_I^2}}{b_I} \right) J_0(\beta_h K r) \\
&\rightarrow 2\pi \int_0^\infty dr r \exp \left(-\frac{\sqrt{r^2 + l_D^2}}{b_D} \right) \exp \left(-\frac{\sqrt{r^2 + l_I^2}}{b_I} \right) := I_X
\end{aligned}$$

in the limit $K \rightarrow 0$. As $\mathbf{K} = \mathbf{q} + \Delta\mathbf{Q}$ we can write

$$\mathcal{J}'_{ee'}(\mathbf{q} \rightarrow 0, \Delta\mathbf{Q} \rightarrow 0) = 2\pi C I_X^2 \frac{e^{-KL}}{K}.$$

Repeating the procedure for other three terms we find

$$\begin{aligned}\mathcal{J}'_{hh'}(\mathbf{q} \rightarrow 0, \Delta\mathbf{Q} \rightarrow 0) &= 2\pi C I_X^2 \frac{1}{K}, \\ \mathcal{J}'_{eh'}(\mathbf{q} \rightarrow 0, \Delta\mathbf{Q} \rightarrow 0) &= -2\pi C I_X^2 \frac{1}{K}, \\ \mathcal{J}'_{he'}(\mathbf{q} \rightarrow 0, \Delta\mathbf{Q} \rightarrow 0) &= -2\pi C I_X^2 \frac{e^{-KL}}{K}.\end{aligned}$$

The total exciton exchange term becomes

$$V_X(K \rightarrow 0) = 2\pi C I_X^2 \left[\frac{e^{-KL}}{K} + \frac{1}{K} - \frac{1}{K} - \frac{e^{-KL}}{K} \right] = 0.$$

Electron and hole exchange DX-IX interaction. The general form of the electron exchange interaction between direct and indirect exciton is given by Eq. (B.3). For the calculations it is more convenient to choose the indirect exciton wave function written in the form (B.6). Thus the exchange interaction matrix element reads

$$\begin{aligned}V_e^{exch}(\mathbf{Q}, \mathbf{Q}', \mathbf{q}) &= -\frac{e^2}{4\pi\epsilon\epsilon_0} \frac{1}{S^2} \frac{4}{\pi^2} \frac{1}{a_{DX}^2} \frac{1}{a_{IX}^2} \int d^2\mathbf{r}_e d^2\mathbf{r}_h d^2\mathbf{r}_{e'} d^2\mathbf{r}_{h'} \\ &\times \exp\left(-\frac{|\mathbf{r}_e - \mathbf{r}_h| + |\mathbf{r}_{e'} - \mathbf{r}_h|}{a_{DX}} - \frac{|\mathbf{r}_{e'} - \mathbf{r}_{h'}| + |\mathbf{r}_e - \mathbf{r}_{h'}|}{a_{IX}}\right) \\ &\times \exp[-i\mathbf{Q}(\beta_e\mathbf{r}_e + \beta_h\mathbf{r}_h) - i\mathbf{Q}'(\beta_e\mathbf{r}_{e'} + \beta_h\mathbf{r}_{h'}) \\ &+ i(\mathbf{Q} + \mathbf{q})(\beta_e\mathbf{r}_e + \beta_h\mathbf{r}_h) + i(\mathbf{Q}' - \mathbf{q})(\beta_e\mathbf{r}_{e'} + \beta_h\mathbf{r}_{h'})] \\ &\times \left[\frac{1}{\sqrt{(\mathbf{r}_e - \mathbf{r}_{e'})^2 + L^2}} + \frac{1}{|\mathbf{r}_h - \mathbf{r}_{h'}|} - \frac{1}{|\mathbf{r}_e - \mathbf{r}_{h'}|} - \frac{1}{\sqrt{(\mathbf{r}_h - \mathbf{r}_{e'})^2 + L^2}} \right].\end{aligned}\tag{B.21}$$

The exact calculation of exchange integral is straightforward but tedious. Starting with the same substitutions as for direct interaction calculation, and defining

new variables

$$\mathbf{y}_1 = (\boldsymbol{\xi} - \beta_e \mathbf{r} - \beta_h \mathbf{r}')/a_{DX}, \quad \mathbf{y}_2 = (\boldsymbol{\xi} + \beta_h \mathbf{r} + \beta_e \mathbf{r}')/a_{DX}, \quad \mathbf{x} = \mathbf{r}/a_{DX},$$

one gets the final expression of electron exchange interaction

$$V_e^{exch} = -\frac{e^2}{\pi^3 \epsilon \epsilon_0 S} \frac{a_{DX}^3}{a_{IX}^2} \mathcal{J}_e^{exch}(\Delta Q, q, \Theta, \beta_e, a_{DX}, \alpha). \quad (\text{B.22})$$

Here $\alpha = a_{DX}/a_{IX}$ is the ratio between the direct and indirect exciton Bohr radii, and Θ is the angle between $\Delta \mathbf{Q}$ and $\mathbf{q}$. The integral is given by

$$\begin{aligned} \mathcal{J}_e^{exch}(\Delta Q, q, \Theta, \beta_e, a_{DX}, \alpha) = & \int_0^\infty dx \int_0^{2\pi} d\theta_x \int_0^\infty dy_1 \int_0^{2\pi} d\theta_1 \int_0^\infty dy_2 \int_0^{2\pi} d\theta_2 \\ & \times xy_1 y_2 \cos \left[a_{DX} \beta_e \Delta Q [x \cos(\Theta - \theta_x) + y_1 \cos(\Theta - \theta_1)] + a_{DX} q [x \cos \theta_x \right. \\ & \left. + \beta_e y_1 \cos \theta_1 - (1 - \beta_e) y_2 \cos \theta_2] \right] \exp(-x - y_1 - \alpha y_2) \\ & \times \exp \left[-\alpha \sqrt{(y_2 \cos \theta_2 - y_1 \cos \theta_1 - x \cos \theta_x)^2 + (y_2 \sin \theta_2 - y_1 \sin \theta_1 - x \sin \theta_x)^2} \right] \\ & \times \left[\frac{1}{\sqrt{y_1^2 + x^2 + 2y_1 x \cos(\theta_1 - \theta_x) + \tilde{L}^2}} + \frac{1}{\sqrt{y_2^2 + x^2 - 2y_2 x \cos(\theta_2 - \theta_x)}} - \frac{1}{y_2} \right. \\ & \left. - \frac{1}{\sqrt{y_1^2 + \tilde{L}^2}} \right], \end{aligned} \quad (\text{B.23})$$

where $\tilde{L} = L/a_{DX}$. The calculation of exchange integral was performed with a multidimensional Monte Carlo algorithm.

The same procedure for the hole exchange term yields

$$V_h^{exch} = -\frac{e^2}{\pi^3 \epsilon \epsilon_0 S} \frac{a_{DX}^3}{a_{IX}^2} \mathcal{J}_h^{exch}(\Delta Q, q, \Theta, \beta_e, a_{DX}, \alpha), \quad (\text{B.24})$$

where one needs to evaluate numerically the expression

$$\begin{aligned}
\mathcal{F}_h^{exch}(\Delta Q, q, \Theta, \beta_e, a_{DX}, \alpha) = & \int_0^\infty dx \int_0^{2\pi} d\theta_x \int_0^\infty dy_1 \int_0^{2\pi} d\theta_1 \int_0^\infty dy_2 \int_0^{2\pi} d\theta_2 \\
& \times xy_1y_2 \cos \left[a_{DX}(1 - \beta_e)\Delta Q[-x \cos(\Theta - \theta_x) + y_2 \cos(\Theta - \theta_2)] + a_{DX}q[-x \cos \theta_x \right. \\
& \left. - \beta_e y_1 \cos \theta_1 + (1 - \beta_e)y_2 \cos \theta_2] \right] \exp(-x - \alpha y_1 - y_2) \\
& \times \exp \left[-\alpha \sqrt{(y_2 \cos \theta_2 - y_1 \cos \theta_1 - x \cos \theta_x)^2 + (y_2 \sin \theta_2 - y_1 \sin \theta_1 - x \sin \theta_x)^2} \right] \\
& \times \left[\frac{1}{\sqrt{y_1^2 + x^2 + 2y_1x \cos(\Theta_1 - \Theta_x) + \tilde{L}^2}} + \frac{1}{\sqrt{y_2^2 + x^2 - 2y_2x \cos(\Theta_2 - \Theta_x)}} - \frac{1}{y_2} \right. \\
& \left. - \frac{1}{\sqrt{y_1^2 + \tilde{L}^2}} \right]. \tag{B.25}
\end{aligned}$$

The total direct-indirect exciton interaction constant can be written as $V_{DI} = V_e^{exch} + V_h^{exch}$. The two non-zero terms are shown as a function of quantum well separation in Fig. B.1.

Figure B.1: Electron and hole exchange terms of the DX-IX interaction constant plotted as a function of separation between centers of quantum wells. The hole exchange term is larger than the electron exchange term for all values of well separation L calculated.

Appendix C: Calculation of overlap dipole matrix element

This Appendix is taken directly from publication [A2].

In this section we calculate the dipole matrix element $d_{di} = -e\langle DX|\hat{z}|IX\rangle$. This can be done by calculating the exchange integral between the ground states of two finite potential quantum wells. The time-independent Schrödinger equations for each separate well read as

$$E\psi(z) = -\frac{\hbar^2}{2m^*}\psi''(z) + V(z)\psi(z), \quad (\text{C.1})$$

where

$$V(z) = V_0\left(\theta(-z - d/2) + \theta(z - d/2)\right), \quad (\text{C.2})$$

V_0 is the QW depth, and d is the well width. The ground state solution to Eq. (C.1) reads

$$\psi(z) = \begin{cases} Ae^{\alpha z} & \text{if } z < -L/2 \\ B \cos(kz) & \text{if } |z| < L/2, \\ Ae^{-\alpha z} & \text{if } z > L/2 \end{cases} \quad (\text{C.3})$$

where

$$\alpha = \sqrt{2m^*(V_0 - E)}/\hbar,$$

$$k = \sqrt{2m^*E}/\hbar,$$

are parameters found from the lowest k solution to the equation

$$\sqrt{2m^*V_0 - \hbar^2k^2} = \hbar^2k^2 \tan(kd/2). \quad (\text{C.4})$$

Normalization of the states gives coefficients

$$B = \left(\frac{kd + \sin(kd)}{2k} + \frac{\cos(kd/2)^2}{\alpha} \right)^{-1/2}, \quad (\text{C.5})$$

$$A = \cos(kd/2)e^{\alpha d/2}B. \quad (\text{C.6})$$

In addition to the parameters in the main text, the valence band offset between the GaAs bulk and the $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}/\text{In}_{0.08}\text{Ga}_{0.92}\text{As}$ QW is $V_0 = 55$ meV [169]. Finally, the exchange integral can be straightforwardly calculated as

$$d_{di} = \langle DX | (-e\hat{z}) | IX \rangle = -e \int \psi(z) z \psi(z - D) dz \approx -0.17eL, \quad (\text{C.7})$$

where $D = 14$ nm is the distance between the centers of the two QWs.

Bibliography

- [1] M. O. Scully and M. S. Zubairy, *Quantum Optics* (University Press, Cambridge) 2001.
- [2] H. Carmichael, *An Open Systems Approach to Quantum Optics* (Springer-Verlag, Berlin) 1991.
- [3] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions: Basic Processes and Applications* (Wiley, Weinheim) 2004.
- [4] Y.-S. Lee, *Principles of Terahertz Science and Technology* (Springer, U.S.A.) 2009.
- [5] X.-C. Zhang and J. Xu, *Introduction to THz Wave Photonics* (Springer, U.S.A.) 2010.
- [6] A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, “The electronic properties of graphene”, *Rev. Mod. Phys.* **81**, 109 (2009).
- [7] S. Das Sarma, S. Adam, E. H. Hwang, and E. Rossi, “Electronic transport in two-dimensional graphene”, *Rev. Mod. Phys.* **83**, 407 (2011).
- [8] J. F. Federici, B. Schulkin, F. Huang, D. Gary, R. Barat, F. Oliveira, and D. Zimdars, “THz imaging and sensing for security applications – Explosives, weapons and drugs”, *Semicond. Sci. Technol.* **20**, S266 (2005).
- [9] A. J. Fitzgerald, E. Berry, N. N. Zinovev, G. C. Walker, M. A. Smith, and J. M. Chamberlain, “An Introduction to Medical Imaging with Coherent Terahertz Frequency Radiation”, *Phys. Med. Biol.* **47**, R67 (2002).

- [10] C. Yu, S. Fan, Y. Sun, and E. Pickwell-MacPherson, “The potential of terahertz imaging for cancer diagnosis: A review of investigations to date”, *Quant. Imaging Med. Surg.* **2**(1), 33 (2012).
- [11] C. Kulesa, “Terahertz Spectroscopy for Astronomy: From Comets to Cosmology”, *IEEE Trans. Terahertz Sci. Technol.* **1**(1), 232 (2011).
- [12] P. F. Taday, I. V. Bradley, D. D. Arnone, and M. Pepper, “Using Terahertz pulse spectroscopy to study the crystalline structure of a drug: a case study of the polymorphs of ranitidine hydrochloride”, *J. Pharm. Sci.* **92**(4), 831 (2003).
- [13] C. Gmachl, F. Capasso, D. L. Sivco, and A. Y. Cho, “Recent progress in quantum cascade lasers and applications”, *Rep. Prog. Phys.* **64**, 1533 (2001).
- [14] R. F. Kazarinov, and R. A. Suris, “Possibility of amplification of electromagnetic waves in a semiconductor with a superlattice”, *Sov. Phys. Semicond.* **5**, 207 (1971).
- [15] J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A. L. Hutchinson, and A. Y. Cho, “Quantum Cascade Laser”, *Science* **264**, 553 (1994).
- [16] S. Kumar and A. W. M. Lee, “Resonant-phonon terahertz quantum-cascade lasers and video-rate terahertz imaging”, *IEEE J. Sel. Top. Quantum Electron.* **14**, 333 (2008).
- [17] J. Shan and T. F. Heinz, “Terahertz radiation from semiconductors”, *Topics Appl. Phys.* **92**, 59 (2004).
- [18] Y. J. Ma, M. C. Huang, S. Ryu, C. W. Bark, C. B. Eom, P. Irvin, and J. Levy, “Broadband Terahertz Generation and Detection at 10 nm Scale”, *Nano Lett.* **13**, 2884 (2013).
- [19] M. Nagel, A. Michalski, T. Botzem, and H. Kurz, “Near-field investigation of THz surface-wave emission from optically excited graphite flakes”, *Opt. Express* **19**, 4667 (2011).

- [20] G. Ramakrishnan, N. Kumar, G. K. P. Ramanandan, A. J. L. Adam, R. W. A. Hendrikx, and P. C. M. Planken, “Plasmon-enhanced terahertz emission from a semiconductor/metal interface”, *Appl. Phys. Lett.* **104**, 071104 (2014).
- [21] M. Wächter, C. Matheisen, M. Waldow, T. Wahlbrink, J. Bolten, M. Nagel, and H. Kurz, “Optical generation of terahertz and second-harmonic light in plasma-activated silicon nanophotonic structures”, *Appl. Phys. Lett.* **97**, 161107 (2010).
- [22] H. Eisele, A. Rydberg, and G. I. Haddad, “Recent advances in the performance of InP Gunn devices and GaAs TUNNETT diodes for the 100-300-GHz frequency range and above”, *IEEE Trans. Microw. Theory Techn.* **48**, 626 (2000).
- [23] Z. Huang and K.-J. Kim, “Review of x-ray free-electron laser theory”, *Phys. Rev. ST Accel. Beams* **10**, 034801 (2007).
- [24] B. A. Knyazeb, G. N. Kulipanov, and N. A. Vinokurov, “Novosibirsk terahertz free electron laser: instrumentation development and experimental achievements”, *Meas. Sci. Technol.* **21**, 054017 (2010).
- [25] A. V. Kavokin, J. J. Baumberg, G. Malpuech, and F. P. Laussy, *Microcavities* (Oxford University Press, Oxford) 2007.
- [26] J. P. Reithmaier, G. Sek, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskii, T. L. Reinecke, and A. Forchel. *Nature* **432**, 197-200 (2004).
- [27] J. Vučković and Y. Yamamoto, “Photonic crystal microcavities for cavity quantum electrodynamics with a single quantum dot”, *Appl. Phys. Lett.* **82**, 2374 (2003).
- [28] T. Yoshie, A. Scherer, J. Hendrickson, G. Khitrova, H. M. Gibbs, G. Rupper, C. Ell, O. B. Shchekin, and D. G. Deppe, “Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity”, *Nature* **432**, 200 (2004).

- [29] D. Englund, D. Fattal, E. Waks, G. Solomon, B. Zhang, T. Nakaoka, Y. Arakawa, Y. Yamamoto, and J. Vučković, “Controlling the Spontaneous Emission Rate of Single Quantum Dots in a Two-Dimensional Photonic Crystal”, *Phys. Rev. Lett.* **95**, 013904 (2005).
- [30] M. Cai, O. Painter, and K. J. Vahala, “Observation of Critical Coupling in a Fiber Taper to a Silica-Microsphere Whispering-Gallery Mode System”, *Phys. Rev. Lett.* **85**, 74 (2000).
- [31] E. Peter, P. Senellart, D. Martrou, A. Lemaître, J. Hours, J. M. Gérard, and J. Bloch, “Exciton-Photon Strong-Coupling Regime for a Single Quantum Dot Embedded in a Microcavity”, *Phys. Rev. Lett.* **95**, 067401 (2005).
- [32] L. Sun, Z. Chen, Q. Ren, K. Yu, L. Bai, W. Zhou, H. Xiong, Z. Q. Zhu, and X. Shen, “Direct Observation of Whispering Gallery Mode Polaritons and their Dispersion in a ZnO Tapered Microcavity”, *Phys. Rev. Lett.* **100**, 156403 (2008).
- [33] L. M. Surhone, M. T. Timpelton, and S. F. Marseken (Eds.), *Transfer-Matrix Method (Optics)* (VDM Publishing) 2010.
- [34] A. C. Jones, M. L. Hitchman, *Chemical Vapour Deposition: Precursors, Processes and Applications* (Royal Society of Chemistry) 2009.
- [35] G. S. Huang, T. C. Lu, H. H. Yao, H. C. Kuo, S. C. Wang, C.-W. Lin, and L. Chang, “Crack-free GaN/AlN distributed Bragg reflectors incorporated with GaN/AlN superlattices grown by metalorganic chemical vapor deposition”, *Appl. Phys. Lett.* **88**, 061904 (2006).
- [36] K. E. Waldrip, J. Han, J. J. Figiel, H. Zhou, E. Makarona, and A. V. Nurmiikko, “Stress engineering during metalorganic chemical vapor deposition of AlGaIn/GaN distributed Bragg reflectors”, *Appl. Phys. Lett.* **78**, 3205 (2001).
- [37] T. C. Lu, J. Y. Tsai, J. T. Chu, Y. S. Chang, and S. C. Wang, “InP/InGaAlAs distributed Bragg reflectors grown by low-pressure metal organic chemical vapor deposition”, *J. Cryst. Growth.* **250**, 305 (2003).

- [38] M. Cho, J.-H. Seo, J. Lee, D. Zhao, H. Mi, X. Yin, M. Kim, X. Wang, W. Zhou, and Z. Ma, “Ultra-thin distributed Bragg reflectors via stacked single-crystal silicon nanomembranes”, *Appl. Phys. Lett.* **106**, 181107 (2015).
- [39] C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, “Observation of the coupled exciton-photon mode splitting in a semiconductor quantum microcavity”, *Phys. Rev. Lett.* **69**, 3314 (1992).
- [40] H. Deng, H. Haug, and Y. Yamamoto, “Exciton-polariton Bose-Einstein condensation”, *Rev. Mod. Phys.*, **82**, 1489 (2010).
- [41] I. Carusotto and C. Ciuti, “Quantum fluids of light”, *Rev. Mod. Phys.* **85**, 299 (2013).
- [42] J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. M. J. Keeling, F. M. Marchetti, M. H. Szymańska, R. André, J. L. Staehli, V. Savona, P. B. Littlewood, B. Deveaud, and Le Si Dang, “Bose-Einstein condensation of exciton polaritons”, *Nature* **443**, 409 (2006).
- [43] R. Balili, V. Hartwell, D. Snoke, L. Pfeiffer, and K. West, “Bose-Einstein Condensation of Microcavity Polaritons in a Trap”, *Science* **316**, 1007 (2007).
- [44] M. H. Anderson, J. R. Ensher, M. R. Matthews, C. E. Wieman, and E. A. Cornell, “Observation of Bose-Einstein Condensation in a Dilute Atomic Vapor”, *Science* **269**, 198 (1995).
- [45] J. J. Baumberg, A. V. Kavokin, S. Christopoulos, A. J. D. Grundy, R. Butté, G. Christmann, D. D. Solnyshkov, G. Malpuech, G. Baldassarri Höger von Högersthal, E. Feltin, J.-F. Carlin, and N. Grandjean, “Spontaneous Polarization Buildup in a Room-Temperature Polariton Laser”, *Phys. Rev. Lett.* **101**, 136409 (2008).
- [46] J. D. Plumhof, T. Stöferle, L. Mai, U. Scherf, and R. F. Mahrt, “Room-temperature Bose-Einstein condensation of cavity exciton–polaritons in a polymer”, *Nature Materials* **13**, 247 (2014).

- [47] A. Imamoglu, R. J. Ram, S. Pau, and Y. Yamamoto, “Nonequilibrium condensates and lasers without inversion: Exciton-polariton lasers”, *Phys. Rev. A* **53**, 4250 (1996).
- [48] S. Christopoulos, G. Bandassarri Höger von Högersthal, A. J. D. Grundy, P. G. Lagoudakis, A. V. Kavokin, J. J. Baumberg, G. Christmann, R. Butté, E. Feltin, J.-F. Carlin, and N. Grandjean, “Room-Temperature Polariton Lasing in Semiconductor Microcavities”, *Phys. Rev. Lett.* **98**, 126405 (2007).
- [49] C. Schneider, A. Rahimi-Imam, N. Y. Kim, J. Fischer, I. G. Savenko, M. Amthor, M. Lerner, A. Wolf, L. Worschech, V. D. Kulakovskii, E. A. She-lykh, M. Kamp, S. Reitzenstein, A. Forchel, Y. Yamamoto, and S. Höfling, “An electrically pumped polariton laser”, *Nature* **497**, 348 (2013).
- [50] P. Bhattacharya, B. Xiao, A. Das, S. Bhowmick, and J. Heo, “Solid State Electrically Injected Exciton-Polariton Laser”, *Phys. Rev. Lett.* **110**, 206403 (2013).
- [51] P. Bhattacharya, T. Frost, S. Deshpande, Md Z. Baten, A. Hazari, and A. Das, “Room Temperature Electrically Injected Polariton Laser”, *Phys. Rev. Lett.* **112**, 236802 (2014).
- [52] L. Davidovich, “Sub-Poissonian processes in quantum optics”, *Rev. Mod. Phys.* **68**, 127 (1996).
- [53] A. J. Shields, “Semiconductor quantum light sources”, *Nature Photonics* **1**, 215 (2007).
- [54] V. Scarani, H. Bechmann-Pasquinucci, N. J. Cerf, M. Dušek, N. Lütkenhaus, and M. Peev, “The security of practical quantum key distribution”, *Rev. Mod. Phys.* **81**, 1301 (2009).
- [55] A. Verger, C. Ciuti, and I. Carusotto, “Polariton quantum blockade in a photonic dot”, *Phys. Rev. B* **73**, 193306 (2006).

- [56] F. Tassone and Y. Yamamoto, “Exciton-exciton scattering dynamics in a semiconductor microcavity and stimulated scattering into polaritons”, *Phys. Rev. B* **59**, 10830 (1999).
- [57] A. Baas, J. Ph. Karr, H. Eleuch, and E. Giacobino, “Optical bistability in semiconductor microcavities”, *Phys. Rev. A* **69**, 023809 (2004).
- [58] A. Baas, J. Ph. Karr, M. Romanelli, A. Bramati, and E. Giacobino, “Optical bistability in semiconductor microcavities in the nondegenerate parametric oscillation regime: Analogy with the optical parametric oscillator”, *Phys. Rev. B* **70**, 161307(R) (2004).
- [59] N. A. Gippius, S. G. Tikhodeev, V. D. Kulakovskii, D. N. Krizhanovskii, and A. I. Tartakovskii, “Nonlinear dynamics of polariton scattering in semiconductor microcavity: Bistability vs. stimulated scattering”, *EPL* **67**, 997 (2004).
- [60] D. M. Whittaker, “Effects of polariton-energy renormalization in the microcavity optical parametric oscillator”, *Phys. Rev. B* **71**, 115301 (2005).
- [61] N. A. Gippius, I. A. Shelykh, D. D. Solnyshkov, S. S. Gavrilov, Y. G. Rubo, A. V. Kavokin, S. G. Tikhodeev, and G. Malpuech, “Polarization Multistability of Cavity Polaritons”, *Phys. Rev. Lett.* **98**, 236401 (2007).
- [62] T. K. Paraïso, M. Wouters, Y. Léger, F. Morier-Genoud, and B. Deveaud-Plédran, “Multistability of a coherent spin ensemble in a semiconductor microcavity”, *Nat. Mater.* **9**, 655 (2010).
- [63] I. A. Shelykh, T. C. H. Liew, and A. V. Kavokin, “Spin Rings in Semiconductor Microcavities”, *Phys. Rev. Lett.* **100**, 116401 (2008).
- [64] C. Adrados, A. Amo, T. C. H. Liew, R. Hivet, R. Houdré, E. Giacobino, A. V. Kavokin, and A. Bramati, “Spin Rings in Bistable Planar Semiconductor Microcavities”, *Phys. Rev. Lett.* **105**, 216403 (2010).

- [65] C. Adrados, T. C. H. Liew, A. Amo, M. D. Martín, D. Sanvitto, C. Antón, E. Giacobino, A. Kavokin, A. Bramati, and L. Viña, “Motion of Spin Polariton Bullets in Semiconductor Microcavities”, *Phys. Rev. Lett.* **107**, 146402 (2011).
- [66] T. Gao, P. S. Eldridge, T. C. H. Liew, S. I. Tsintzos, G. Stavrinidis, G. Deligeorgis, Z. Hatzopoulos, and P. G. Savvidis, “Polariton condensate transistor switch”, *Phys. Rev. B* **85**, 235102 (2012).
- [67] D. Ballarini, M. De Giorgi, E. Cancellieri, R. Houdré, E. Giacobino, R. Cingolani, A. Bramati, G. Gigli, and D. Sanvitto, “All-optical polariton transistor”, *Nat. Commun.* **4**, 1778 (2013).
- [68] A. Amo, T. C. H. Liew, C. Adrados, R. Houdré, E. Giacobino, A. V. Kavokin, and A. Bramati, “Exciton-polariton spin switches”, *Nature Photon.* **4**, 361 (2010).
- [69] M. De Giorgi, D. Ballarini, E. Cancellieri, F. M. Marchetti, M. H. Szymanska, C. Tejedor, R. Cingolani, E. Giacobino, A. Bramati, G. Gigli, and D. Sanvitto, “Control and Ultrafast Dynamics of a Two-Fluid Polariton Switch”, *Phys. Rev. Lett.* **109**, 266407 (2012).
- [70] T. C. H. Liew, A. V. Kavokin, and I. A. Shelykh, “Optical Circuits Based on Polariton Neurons in Semiconductor Microcavities”, *Phys. Rev. Lett.* **101**, 016402 (2008).
- [71] T. Espinosa-Ortega and T. C. H. Liew, “Complete architecture of integrated photonic circuits based on AND and NOT logic gates of exciton polaritons in semiconductor microcavities”, *Phys. Rev. B* **87**, 195395 (2013).
- [72] K. V. Kavokin, M. A. Kaliteevski, R. A. Abram, A. V. Kavokin, S. Sharkova, and I. A. Shelykh, “Stimulated emission of terahertz radiation by exciton-polariton lasers”, *Appl. Phys. Lett.* **97**, 201111 (2010).
- [73] F. Barachati, S. De Liberato, and S. Kéna-Cohen. “Generation of Rabi-frequency radiation using exciton-polaritons”, *Phys. Rev. A* **92**, 033828 (2015).

- [74] E. del Valle and A. V. Kavokin, “Terahertz lasing in a polariton system: Quantum theory”, *Phys. Rev. B* **83**, 193303 (2011).
- [75] I. G. Savenko, I. A. Shelykh, and M. A. Kaliteevski, “Nonlinear Terahertz Emission in Semiconductor Microcavities”, *Phys. Rev. Lett.* **107**, 027401 (2011).
- [76] A. V. Kavokin, I. A. Shelykh, T. Taylor, and M. M. Glazov, “Vertical Cavity Surface Emitting Terahertz Laser”, *Phys. Rev. Lett.* **108**, 197401 (2012).
- [77] J. L. Tomaino, A. D. Jameson, Y.-S. Lee, G. Khitrova, H. M. Gibbs, A. C. Klettke, M. Kira, and S. W. Koch, “Terahertz Excitation of a Coherent Λ -Type Three-Level System of Exciton-Polariton Modes in a Quantum-Well Microcavity”, *Phys. Rev. Lett.* **108**, 267402 (2012).
- [78] T. C. H. Liew, M. M. Glazov, K. V. Kavokin, I. A. Shelykh, M. A. Kaliteevski, and A. V. Kavokin, “Proposal for a Bosonic Cascade Laser”, *Phys. Rev. Lett.* **110**, 047402 (2013).
- [79] S. De Liberato, C. Ciuti, and C. C. Phillips, “Terahertz lasing from intersubband polariton-polariton scattering in asymmetric quantum wells”, *Phys. Rev. B* **87**, 241304(R) (2013).
- [80] D. Dini, R. Köhler, A. Tredicucci, G. Biasiol, and L. Sorba, “Microcavity Polariton Splitting of Intersubband Transitions”, *Phys. Rev. Lett.* **90**, 116401 (2003).
- [81] Yu. E. Lozovik and V. I. Yudson, “A new mechanism for superconductivity: pairing between spatially separated electrons and holes”, *Zh. Eksp. Teor. Fiz.* **71**, 738 (1976) [*Sov. Phys. JETP* **44**, 389 (1976)].
- [82] T. Fukuzawa, E. E. Mendez, and J. M. Hong, “Phase transition of an exciton system in GaAs coupled quantum wells”, *Phys. Rev. Lett.* **64**, 3066 (1990).
- [83] X. Zhu, P. B. Littlewood, M. S. Hybertsen, and T. M. Rice, “Exciton Condensate in Semiconductor Quantum Well Structures”, *Phys. Rev. Lett.* **74**, 1633 (1995).

- [84] Yu. E. Lozovik and O. L. Berman, “Phase transitions in a system of spatially separated electrons and holes”, JETP Lett. **84**, 1027 (1997).
- [85] T. Iida and M. Tsubota, “Order formation and superfluidity of excitons in type-II semiconductor quantum wells”, Phys. Rev. B **60**, 5802 (1999).
- [86] A. B. Dzyubenko and A. L. Yablonskii, “Intrawell and interwell magnetoexcitons in $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ coupled double quantum wells”, Phys. Rev. B **53**, 16355 (1996).
- [87] V. I. Yudson, “Charged “Few-Electron-Single Spatially Separated Hole” Complexes in a Double Quantum Well near a Metal Plate”, Phys. Rev. Lett. **77**, 1564 (1996).
- [88] J. Fernández-Rossier and C. Tejedor, “Spin Degree of Freedom in Two Dimensional Exciton Condensates”, Phys. Rev. Lett. **78**, 4809 (1997).
- [89] L. V. Butov and A. I. Filin, “Anomalous transport and luminescence of indirect excitons in AlAs/GaAs coupled quantum wells as evidence for exciton condensation”, Phys. Rev. B **58**, 1980 (1998).
- [90] V. V. Krivolapchuk, E. S. Moskalenko, A. L. Zhmodikov, T. S. Cheng, and C. T. Foxon, “Collective properties of spatially indirect excitons in asymmetric $\text{GaAs}/\text{AlGaAs}$ double quantum wells”, Solid State Commun. **111**, 49 (1999).
- [91] A. V. Larionov and V. B. Timofeev, “Condensation of interwell excitons in $\text{GaAs}/\text{AlGaAs}$ double quantum wells”, JETP Lett. **73**, 342 (2001).
- [92] L. V. Butov, A. L. Ivanov, A. Imamoglu, P. B. Littlewood, A. A. Shashkin, V. T. Dolgoplov, K. L. Campman, and A. C. Gossard, “Stimulated Scattering of Indirect Excitons in Coupled Quantum Wells: Signature of a Degenerate Bose-Gas of Excitons”, Phys. Rev. Lett. **86**, 5608 (2001).
- [93] L. V. Butov, C. W. Lai, A. L. Ivanov, A. C. Gossard, and D. S. Chemla, “Towards Bose-Einstein condensation of excitons in potential traps”, Nature **417**, 47 (2002).

- [94] L. V. Butov, A. C. Gossard, and D. S. Chemla, “Macroscopically ordered state in an exciton system”, *Nature* **418**, 751 (2002).
- [95] A. A. High, J. R. Leonard, A. T. Hammack, M. M. Fogler, L. V. Butov, A. V. Kavokin, K. L. Campman, and A. C. Gossard, “Spontaneous coherence in a cold exciton gas”, *Nature* **483**, 584 (2012).
- [96] V. Negoita, D. W. Snoke, and K. Eberl, “Harmonic-potential traps for indirect excitons in coupled quantum wells”, *Phys. Rev. B* **60**, 2661 (1999).
- [97] V. Negoita, D. W. Snoke, and K. Eberl, “Stretching quantum wells: A method for trapping free carriers in GaAs heterostructures”, *Appl. Phys. Lett.* **75**, 2059 (1999).
- [98] L. V. Butov, A. Zrenner, G. Abstreiter, G. Böhm, and G. Weimann, “Condensation of Indirect Excitons in Coupled AlAs/GaAs Quantum Wells”, *Phys. Rev. Lett.* **73**, 304 (1994).
- [99] V. Negoita, D. Hackworth, D. W. Snoke, and K. Eberl, “Sub-Hz spectral fluctuations from high-density excitons in biased coupled quantum wells”, *Opt. Lett.* **25**, 572 (2000).
- [100] A. L. Ivanov, “Quantum diffusion of dipole-oriented indirect excitons in coupled quantum wells”, *Europhys. Lett.* **59**, 586 (2002).
- [101] D. Snoke, S. Denev, Y. Liu, L. Pfeiffer, and K. West, “Long-range transport in excitonic dark states in coupled quantum wells”, *Nature* **418**, 754 (2002).
- [102] G. Christmann, C. Coulson, J. J. Baumberg, N. T. Pelekanos, Z. Hatzopoulos, S. I. Tsintzos, and P. G. Savvidis, “Control of polariton scattering in resonant-tunneling double-quantum-well semiconductor microcavities”, *Phys. Rev. B* **82**, 113308 (2010).
- [103] P. Cristofolini, G. Christmann, S. I. Tsintzos, G. Deligeorgis, G. Konstantinidis, Z. Hatzopoulos, P. G. Savvidis, and J. J. Baumberg, “Coupling Quantum Tunneling with Cavity Photons”, *Science* **336**, 704 (2012).

- [104] G. Christmann, A. Askitopoulos, G. Deligeorgis, Z. Hatzopoulos, S. I. Tsintzos, P. G. Savvidis, and J. J. Baumberg, “Oriented polaritons in strongly-coupled asymmetric double quantum well microcavities”, *Appl. Phys. Lett.* **98**, 081111 (2011).
- [105] K. Sivalertporn, L. Mouchliadis, A. L. Ivanov, R. Philp, and E. A. Muljarov, “Direct and indirect excitons in semiconductor coupled quantum wells in an applied electric field”, *Phys. Rev. B* **85**, 045207 (2012).
- [106] O. Kyriienko, A. V. Kavokin, and I. A. Shelykh, “Superradiant terahertz emission by dipolaritons”, *Phys. Rev. Lett.* **111**, 176401 (2013).
- [107] S. H. Autler and C. H. Townes, “Stark Effect in Rapidly Varying Fields”, *Phys. Rev.* **100**, 703 (1955).
- [108] F. Schuda, C. R. Stroud, Jr., and M. Hercher, “Observation of the resonant Stark effect at optical frequencies”, *J. Phys. B* **7**, L198 (1974).
- [109] F. Y. Wu, R. E. Grove, and S. Ezekiel, “Investigation of the Spectrum of Resonance Fluorescence Induced by a Monochromatic Field”, *Phys. Rev. Lett.* **35**, 1426 (1975).
- [110] W. Hartig, W. Rasmussen, R. Schieder, and H. Walther, “Study of the Frequency Distribution of the Fluorescent Light Induced by Monochromatic Radiation”, *Z. Phys. A* **278**, 205 (1976).
- [111] M. Chini, B. Zhao, H. Wang, Y. Cheng, S. X. Hu, and Z. Chang, “Subcycle ac Stark Shift of Helium Excited States Probed with Isolated Attosecond Pulses”, *Phys. Rev. Lett.* **109**, 073601 (2012).
- [112] P. V. Demekhin and L. S. Cederbaum, “Dynamic Interference of Photoelectrons Produced by High-Frequency Laser Pulses”, *Phys. Rev. Lett.* **108**, 253001 (2012).
- [113] C. Yu, N. Fu, T. Hu, G. Zhang, and J. Yao, “Dynamic Stark effect and interference photoelectron spectra of H_2^+ ”, *Phys. Rev. A* **88**, 043408 (2013).

- [114] P. V. Demekhin and L. S. Cederbaum, “ac Stark effect in the electronic continuum and its impact on the photoionization of atoms by coherent intense short high-frequency laser pulses”, *Phys. Rev. A* **88**, 043414 (2013).
- [115] N. B. Delone and V. P. Krainov, “AC Stark shift of atomic energy levels”, *Sov. Phys. Usp.* **42**, 669 (1999).
- [116] M. Chini, B. Zhao, H. Wang, Y. Cheng, S. X. Hu, and Z. Chang, “Subcycle ac Stark Shift of Helium Excited States Probed with Isolated Attosecond Pulses”, *Phys. Rev. Lett.* **109**, 073601 (2012).
- [117] M. S. P. Eastham, *The Spectral Theory of Periodic Differential Equations* (Scottish Academic Press, Edinburgh) 1973.
- [118] N. J. Mason, “Laser-assisted electron-atom collisions”, *Rep. Prog. Phys.* **56**, 1275 (1993).
- [119] F. Ehlotzky, A. Jaroń, and J. Z. Kamiński, “Electron–atom collisions in a laser yield”, *Phys. Rep.* **297**, 63 (1998).
- [120] N. M. Kroll and K. M. Watson, “Charged-Particle Scattering in the Presence of a Strong Electromagnetic Wave”, *Phys. Rev. A* **8**, 804 (1973).
- [121] A. Weingartshofer, J. K. Holmes, G. Caudle, E. M. Clarke, and H. Krüger, “Direct Observation of Multiphoton Processes in Laser-Induced Free-Free Transitions”, *Phys. Rev. Lett.* **39**, 269 (1977).
- [122] F. W. Byron, Jr. and C. J. Joachain, “Electron–atom collisions in a strong laser field”, *J. Phys. B* **17**, L295 (1984).
- [123] F. W. Byron, Jr., P. Francken, and C. J. Joachain, “Laser-assisted elastic electron–atom collisions”, *J. Phys. B* **20**, 5487 (1987).
- [124] A. Dubois, A. Maquet, and S. Jetzke, “Electron–H-atom collisions in the presence of a laser field: One-photon free-free transitions”, *Phys. Rev. A* **34**, 1888 (1986).

- [125] P. Francken and C. J. Joachain, “Electron–atomic-hydrogen elastic collisions in the presence of a laser field”, *Phys. Rev. A* **35**, 1590 (1987).
- [126] P. Francken, C. J. Joachain, “Electron-impact excitation of atoms in the presence of a nearly resonant laser field”, *Phys. Rev. A* **41**, 3770 (1990).
- [127] M. Dörr, C. J. Joachain, R. M. Potvliege, and S. Vučić, “Born-Floquet theory of laser-assisted electron-atom collisions”, *Phys. Rev. A* **49**, 4852 (1994).
- [128] R. Kanya, Y. Morimoto, and K. Yamanouchi, “Observation of Laser-Assisted Electron–Atom Scattering in Femtosecond Intense Laser Fields”, *Phys. Rev. Lett.* **105**, 123202 (2010).
- [129] Y. Morimoto, R. Kanya, and K. Yamanouchi, “Light-Dressing Effect in Laser-Assisted Elastic Electron Scattering by Xe”, *Phys. Rev. Lett.* **115**, 123201 (2015).
- [130] B. A. deHarak, L. Ladino, K. B. MacAdam, and N. L. S. Martin, “High-energy electron-helium scattering in a Nd:YAG laser field”, *Phys. Rev. A* **83**, 022706 (2011).
- [131] B. A. deHarak, B. Nosarzewski, M. Siavashpouri, and N. L. S. Martin, “Electron-helium scattering in a 1.17 eV laser field: The effect of polarization direction”, *Phys. Rev. A* **90**, 032709 (2014).
- [132] F. Quochi, G. Bongiovanni, A. Mura, J. L. Staehli, B. Deveaud, R. P. Stanley, U. Oesterle, and R. Houdré, “Strongly Driven Semiconductor Microcavities: From the Polariton Doublet to an ac Stark Triplet”, *Phys. Rev. Lett.* **80**, 4733 (1998).
- [133] A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migus, W. T. Masselink, and H. Morkoç, ““Dressed Excitons” in a Multiple-Quantum-Well Structure: Evidence for an Optical Stark Effect with Femtosecond Response Time”, *Phys. Rev. Lett.* **56**, 2748 (1986).

- [134] A. Von Lehmen, D. S. Chemla, J. E. Zucker, and J. P. Heritage, “Optical Stark effect on excitons in GaAs quantum wells”, *Opt. Lett.* **11**, 609 (1986).
- [135] N. Peyhambarian, S. W. Koch, M. Lindberg, B. Fluegel, M. Joffe, “Dynamic Stark effect of exciton and continuum states in CdS”, *Phys. Rev. Lett.* **62**, 1185 (1989).
- [136] G. Platero and R. Aguado, “Photon-assisted transport in semiconductor nanostructures”, *Phys. Rep.* **395**, 1 (2004).
- [137] M. H. Pedersen and M. Büttiker, “Scattering theory of photon-assisted electron transport”, *Phys. Rev. B* **58**, 12993 (1998).
- [138] M. Moskalets and M. Büttiker, “Floquet scattering theory of quantum pumps”, *Phys. Rev. B* **66**, 205320 (2002).
- [139] O. V. Kibis, “Dissipationless Electron Transport in Photon-Dressed Nanostructures”, *Phys. Rev. Lett.* **107**, 106802 (2011).
- [140] O. V. Kibis, “Persistent current induced by quantum light”, *Phys. Rev. B* **86**, 155108 (2012).
- [141] O. V. Kibis, “How to suppress the backscattering of conduction electrons?”, *EPL* **107**, 57003 (2014).
- [142] S. Morina, O. V. Kibis, A. A. Pervishko, and I. A. Shelykh, “Transport properties of a two-dimensional electron gas dressed by light”, *Phys. Rev. B* **91**, 155312 (2015).
- [143] D. V. Volkov, “On a class of solutions of the Dirac equation”, *Z. Phys.* **94**, 250 (1935).
- [144] A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, “The electronic properties of graphene”, *Rev. Mod. Phys.* **81**, 109 (2009).
- [145] S. Das Sarma, S. Adam, E. H. Hwang, and E. Rossi, “Electronic transport in two-dimensional graphene”, *Rev. Mod. Phys.* **83**, 407 (2011).

- [146] F. J. López-Rodríguez and G. G. Naumis, “Analytic solution for electrons and holes in graphene under electromagnetic waves: Gap appearance and nonlinear effects”, *Phys. Rev. B* **78**, 201406(R) (2008).
- [147] T. Oka and H. Aoki, “Photovoltaic Hall effect in graphene”, *Phys. Rev. B* **79**, 081406(R) (2009).
- [148] O. V. Kibis, “Metal-insulator transition in graphene induced by circularly polarized photons”, *Phys. Rev. B* **81**, 165433 (2010).
- [149] S. E. Savel’ev and A. S. Alexandrov, “Massless Dirac fermions in a laser field as a counterpart of graphene superlattices”, *Phys. Rev. B* **84**, 035428 (2011).
- [150] S. V. Syzranov, M. V. Fistul, and K. B. Efetov, “Effect of radiation on transport in graphene”, *Phys. Rev. B* **78**, 045407 (2008).
- [151] H. L. Calvo, H. M. Pastawski, S. Roche, and L. E. F. Foa Torres, “Tuning laser-induced band gaps in graphene”, *Appl. Phys. Lett.* **98**, 232103 (2011).
- [152] H. L. Calvo, P. M. Perez-Piskunow, H. M. Pastawski, S. Roche, and L. E. F. Foa Torres, “Non-perturbative effects of laser illumination on the electrical properties of graphene nanoribbons”, *J. Phys. Condens. Matter* **25**, 144202 (2013).
- [153] Y. Zhou and M. W. Wu, “Optical response of graphene under intense terahertz fields”, *Phys. Rev. B* **83**, 245436 (2011).
- [154] A. Iurov, G. Gumbs, O. Roslyak, and D. Huang, “Photon dressed electronic states in topological insulators: tunneling and conductance”, *J. Phys. Condens. Matter* **25**, 135502 (2013).
- [155] G. Usaj, P. M. Perez-Piskunow, L. E. F. Foa Torres, and C. A. Balseiro, “Irradiated graphene as a tunable Floquet topological insulator”, *Phys. Rev. B* **90**, 115423 (2014).

- [156] D. Sarchi, I. Carusotto, M. Wouters, and V. Savona, “Coherent dynamics and parametric instabilities of microcavity polaritons in double-well systems”, *Phys. Rev. B* **77**, 125324 (2008).
- [157] O. Kyriienko, E. B. Magnússon, and I. A. Shelykh, “Spin dynamics of cold exciton condensates”, *Phys. Rev. B* **86**, 115324 (2012).
- [158] L. D. Landau and E. M. Lifshitz, *The Classical Theory of Fields* (Butterworth-Heinemann) 1980.
- [159] R. H. Dicke, “Coherence in Spontaneous Radiation Processes”, *Phys. Rev.* **93**, 99 (1954).
- [160] E. M. Purcell, “Spontaneous Emission Probabilities at Radio Frequencies”, *Phys. Rev.* **69**, 674 (1946).
- [161] O. V. Kibis, O. Kyriienko, and I. A. Shelykh, “Band gap in graphene induced by vacuum fluctuations”, *Phys. Rev. B* **84**, 195413 (2011).
- [162] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics: Non-Relativistic Theory* (Pergamon Press, Oxford) 1991.
- [163] N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Saunders College, Philadelphia) 1976.
- [164] R. S. Sorbello, “On the anisotropic relaxation time”, *J. Phys. F* **4**, 503 (1974).
- [165] M. P. Marder, *Condensed matter physics* (John Wiley and Sons, Inc., New York) 2000.
- [166] C. Grossmann, G. Christmann, J. J. Baumberg, I. Farrer, H. Beere, and D. A. Ritchie, “Strong coupling at room temperature in ultracompact flexible metallic microcavities”, *Appl. Phys. Lett.* **102**, 011118 (2013).
- [167] S. de-Leon and B. Laikhtman, “Exciton wave function, binding energy, and lifetime in InAs/GaSb coupled quantum wells”, *Phys. Rev. B* **61**, 2874 (2000).

- [168] S. de-Leon and B. Laikhtman, “Exciton-exciton interactions in quantum wells: Optical properties and energy and spin relaxation”, *Phys. Rev. B* **63**, 125306 (2001).
- [169] H.-P. Komsa, E. Arola, and T. T. Rantala, “Band offset of InGaAs(N)/GaAs interfaces from first principles”, *Appl. Phys. Lett.* **92**, 262101 (2008).