Analysis of semiconductor microcavity exciton-polaritons with coupled harmonic oscillators

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Abstract

Terahertz (THz) time-domain spectroscopy provides insight into electron dynamics in semiconductor heterostructures. High-field THz spectroscopy probes the excitonic nonlinear response of GaAs quantum well (QW) systems and enables the measurement of its coherent dynamics in the time-domain. Consequently, THz spectroscopy allows one to explore the fundamental properties of many-body interactions as well as the potentials of semiconductor nanodevice technology. This work analyzes the light-matter interaction in a semiconductor microcavity with a computational approach. When an exciton in a QW microcavity strongly couples with a cavity photon, a new quasiparticle known as exciton-polariton forms. This thesis shows that classical coupled harmonic oscillators with optical and THz excitations can be used as a model to simulate the dynamics of exciton-polariton and its quantum coherent phenomena. The time evolution of the exciton-photon coupled system is demonstrated by employing the time-dependent damping of the exciton mode and varying the delay between optical and THz pulses. The normal mode splitting is observed in the frequency spectra as a result of strong light-matter hybridization. Finally, computed exciton-polariton oscillation of this work is compared to the computed result from a reference which was obtained using a semiconductor Bloch equations.
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Chapter 1

Introduction

1.1 Motivation

Electrons can be confined in one, two, or three dimensions using semiconductors with different band gaps. A quantum-well (QW) confines electrons in one dimension so that their motion is allowed only in two dimensions. QW is a layer formed in semiconductors such as gallium arsenide (GaAs), with thickness roughly equal to the exciton Bohr radius, sandwiched between two barrier layers with a wider band gap, such as aluminium gallium arsenide (AlGaAs). Due to the confinement in lower dimensions, electron energy levels are quantized, and these systems with reduced dimensionality show optical and electronic properties that are completely different from bulk semiconductors. There is also much attention on quantum wires (1D systems) and quantum dots (3D systems). Quantum confinement in additional dimension yields changes in many properties of semiconductors. Understanding the optical and electrical properties of semiconductor nanostructures and their coupling dynamics provides paths to pursue their applications to nanodevices as well as to the techniques to manipulate their quantum states. Besides expanding the opportunities in technologies, it is interesting to study the physics of many body interactions in semiconductors. Nonlinear optical effects [1,2,3,4], light emissions [5], electronic transport [6], and exciton-polariton Bose-Einstein condensation [7] are examples that led to fundamental understandings of semiconductor nanostructures and their interaction with light.

The objective of this work is to demonstrate the coupling dynamics of QW excitons with microcavity photons using the combination of optical and THz excitations, and to study their quantum coherence. The dynamics of an exciton-polariton in semiconductor microcavity are investigated with a computational approach. It is well-known that photons are describable by quantum harmonic oscillators. Excitons are formed in semiconductors excited with a well-defined energy, and their characteristics can also be described by harmonic oscillators when dealing with their optical response. In QWs in an optical microcavity, these two oscillators are coupled through light-matter interactions. In order to demonstrate this interaction between exciton mode and photon mode, one can use coupled harmonic oscillator model where the two oscillators represent the exciton mode and the cavity photon mode, and they are coupled by a spring. When they oscillate in phase, i.e. in resonance, energy transfers periodically between
the two oscillators, indicating that they are strongly coupled. This partially light and partially matter state is known as the exciton-polariton. The exciton mode is excited by the electric field of an optical pulse. Because of the exciton-cavity photon coupling, energy is coherently exchanged between the two modes. This periodic energy exchange between the two modes is called the excitonic normal-mode coupling because the optical response of the resonator yields two normal modes in its transmission spectrum [2].

1.2 Light-matter interactions

In light-matter interactions, the electric dipole of the atoms interacts with the oscillating electric field of the light. The electromagnetic field, which is often described as a propagating wave, also has characteristics as particles known as photons. Quantum mechanics has allowed us to investigate the nature of particles in a microscopic scale. Quantum optics is an architecture in which quantum mechanics is used to explore the properties of photons and their interactions with matter. Many interesting phenomena of light-matter interactions in microscopic scale have been observed in microcavities where photons are reflected back and forth by the mirrors a number of times, enhancing the interaction strength by confining photons in a small volume of space.

Cavity quantum electrodynamics (QED) explores a coherent interaction of photons confined within a resonant cavity with an atom placed in the cavity. Fig.1.1 (a) describes a cavity QED system. A two-level atom consisting of ground \(|g\rangle\) and excited \(|e\rangle\) states is placed inside a resonator and is coupled to the cavity photon field at a rate \(\Omega\). The cavity has a resonance frequency \(\omega_c\) and a decay rate at which the light inside escapes from the cavity. The atom has a transition energy \(\hbar\omega_a\), and a decoherence rate, which is the rate at which the coherence of the atom’s superposition states is lost due to its interaction with the environment. Decoherence also may be caused when the atom decays to other energy levels and emits a photon having a frequency not resonant with the cavity mode. Total loss of a cavity QED system is described by the cavity decay rate and the decoherence rate of the atom. When a single atom is coupled to the electromagnetic field in free space and put into the excited state, it will spontaneously decay to its ground state while emitting a photon. However, when the resonance frequency of the cavity is tuned to the frequency of this atom, and the cavity has a high quality factor, the atom and the cavity can exchange energy, and a strong coupling can be reached. Interaction is in the strong coupling limit when \(\Omega\) exceeds the larger of the two decay rates of the system.

When the atom and the cavity are in resonance, i.e. when they have the same frequency, the eigenstates of the system split, as shown in Fig.1.1 (b). In this resonant regime of the system, the atom and the cavity are hybridized, and the excited atom can oscillate between the \(|g\rangle\) and \(|e\rangle\) at a rate \(2\Omega\) before its coherence is lost, while periodically emitting and absorbing a photon. Every time the atom is excited, it decays by spontaneously emitting a photon into the cavity, at a rate characterized by the decoherence rate of the atom. This cycle is called a vacuum Rabi oscillation, and the spectroscopic investigation allows the observation of a splitting known as vacuum Rabi splitting in the frequency domain [8]. This splitting has a separation \(\Omega_R = 2\Omega\), which is called the vacuum Rabi frequency. The simplest model to demonstrate the strong coupling effect is the classical coupled harmonic oscillators in which energy is ex-
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changed [9]. Their dynamics are fully classical, but they are capable of explaining the key ideas of the cavity QED effects, which are quantum mechanical. The light-matter hybridization in a semiconductor QW system is demonstrated using this model in the later sections of this thesis.

Figure 1.1: (a) A cavity QED system. A two level atom with frequency $\omega_a$ placed inside the cavity with frequency $\omega_c$ is coherently coupled to the cavity electromagnetic field, at a rate $\Omega$. (b) Vacuum Rabi splitting. Resonant coupling resulting in dressed states with energy separation $\hbar \Omega_R$.

Light-matter interaction has been actively studied in many different systems governed by the cavity QED. One of the early demonstrations of strong light-matter interaction in cavity QED systems were demonstrated in experiments where a cloud of cesium atoms are trapped above a Fabry-Perot optical cavity, and they are then allowed to pass through the cavity one by one. Transmission of the laser through the cavity is measured to count the number of atoms in the cavity and to determine the state of the atoms. The vacuum Rabi splitting was first demonstrated in these atomic systems [10].

Cavity QED has also been realized in solid-state systems [11]. Self-assembled quantum dots behaving as artificial atoms are fabricated inside distributed Bragg reflectors (DBRs) [12]. The most successful implementation of cavity QED is the superconducting quantum-bits (qubits), which led to its own field referred to as circuit QED. In these systems, superconducting circuits composed of capacitors, inductors, and the Josephson junctions behave as non-linear oscillators, serving as artificial atoms, or qubits, and a co-planar waveguide resonators in microwave frequency are coupled to the qubits [13]. Cavity QED has been playing a critical role in quantum information processing because strong coupling is a key to coherently manipulate the qubits.

A polariton is a quasi-particle resulting from a strong coupling of electromagnetic field with electric dipole excitations existing within a crystal lattice [14]. Besides the exciton-polariton discussed in this thesis, there are multiple types of polaritons: phonon-polaritons, inter-subband-polaritons, and surface plasmon-polaritons. These types of polaritons are briefly explained in chapter 9 of Ref.[14]. The confinement of electron-hole pairs (excitons) in semiconductor microcavities was first demonstrated in 1980s [15], and the exciton-photon normal mode coupling was first experimentally shown by Weisbuch et al. in 1992 [16]. The microcavity
exciton-polaritons have long been studied using GaAs heterostructures, and they have served as a platform to study linear and nonlinear effects of polaritons. Exciton-polaritons have also been observed in ZnO nanowires [17], transition metal dichalcogenide (TMDs) [18], plasmonic lattices [19] etc.

1.3 Optical properties of semiconductor nanostructures

1.3.1 Exciton

Figure 1.2 shows the band structure in semiconductors. Band structures of solids are the most fundamental and important concepts of solid state physics. Band structures represent the allowed energy levels within the crystals, known as bands, where each band is a continuous function of a wave vector (dispersion relation). Band structures provide information about electrical and optical properties of solids. The energy gap within the band structure where no electron states can exist is known as the band gap. It is the difference in energy between the lowest energy state in the conduction band and the highest energy state in the valence band of the semiconducting materials [20]. Different material has its unique band structure, and thus band gap is a material-dependent value. GaAs is a III-IV semiconductor with a band gap of 1.424eV.

Exciton is a bound state of an electron-hole pair existing in optically excited semiconductors. When an electron confined in a QW is optically excited (green arrows) and absorbs a photon energy lower than the material’s band gap, electron leaves behind a “hole” (blank circles) with charge $+e$ in the valence band. Excited electron in the conduction band (orange circles) is attracted to the hole by Coulomb force, and the attracted pair is called an exciton. This is analogous to a hydrogen atom in which an electron is bound to a proton. The bound electron-hole pair forms a electric dipole and its dipole moment interacts with the electric field of light. Excitons have discrete energy levels just below the conduction band. Energy-level structure of excitons in QW is analogous to that of hydrogen, but their binding energy is $\sim$10meV in GaAs QW, which is much less than that of hydrogen. The smaller binding energy is due to substantially lighter effective mass of an electron-hole pair compared to that of electron and proton. The background dielectric constant of semiconductor also reduces the strength of Coulomb interaction [4,21].
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Figure 1.2: **Electronic band structure.** Exciton binding energy is $E_{ex} = 10\text{meV}$. For GaAs, band gap is $E_g = 1.424\text{eV}$.

1.3.2 Quantum-well microcavity polaritons

Fig.1.3 (a) shows a typical structure of QW microcavity. An optical microcavity is formed by the DBRs, consisting of layers of low and high refractive indices, each with thickness of $\lambda/4$, where $\lambda$ is the wavelength of the optical pulse in a medium. This structure leads to all reflections in phase, creating constructive interference at a specific wavelength and thus a non-absorbing reflector. The reflectivity achieved by the cavity follows the concept of Fresnel reflection and it is determined by the number of pairs of low/high refractive index layers or by the contrast in the refractive indices. In an experimental work, an optical pulse (shown as a pump laser) irradiates the sample and forms excitons in QWs. Each QW is placed at the antinodes of cavity’s electric field to achieve the maximum exciton-photon coupling strength, and the microcavity is designed to have a thickness of $\lambda/2$ so that the cavity field amplitude is at its maximum at the center of the cavity [7]. DBRs acting as highly reflective mirrors confine the cavity photon field, and QW excitons inside the cavity interact with the photons. Achieving a high quality factor and a small mode volume of a resonator is a key to and has been a challenge to strongly couple light to matter in QED systems.

A Bragg-mirror microcavity encloses the QWs which confine electrons and positively charged holes in a two-dimensional region. Therefore, they are free to move in the planes shown as red layers in Fig.1.3 (a), but their motion in the direction normal to the layers is restricted by the potential discontinuities. Fig.1.3 (b) shows a band diagram of a QW made of GaAs and AlGaAs layers. The discontinuities in the band gap at the interfaces of the GaAs/AlGaAs layers results in the confinement of excitons. Since the motion of excitons is quantized in $z$ direction, their energy levels are discrete, as shown by the dashed lines in the QW. In a system with reduced dimensions such as a QW, quantum confinement effect becomes effective when the thickness of a QW is close to the de Broglie wavelength of electrons or holes [22].
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Figure 1.3: (a) A microcavity quantum well structure. QWs are placed between distributed Bragg reflectors (DBRs). The spontaneous absorption and emission of photons repeat. Figure taken from Ref.[23]. (b) A quantum-well. Schematic diagram of a QW formed with GaAs/AlGaAs layers and their energy bands.

When an electron relaxes and recombines to the hole, it emits a photon with the same energy used to excite the electron. When the resonance frequency of the cavity is tuned to this frequency of excitation, the emitted photon can be trapped into the cavity, excites an electron and creates an exciton again. When energy is coherently exchanged between the exciton and the cavity field, the resulting coupled state is the exciton-polariton. Thus, exciton-polariton is a half-light half-matter quasiparticle resulting from strong light-matter interactions in a QW microcavity.

1.4 THz spectroscopy

1.4.1 Optical and THz excitations

Linear and nonlinear optical properties of semiconductor nanostructures have been studied using ultrafast lasers. The approach of coherent excitation using lasers is widely used to explore the quantum physics of many-body systems [19]. Pump-probe spectroscopy is the simplest and the most common technique of ultrafast spectroscopy. In this technique, output laser is split into two: one of the laser pulse trains (pump) is used to excite the sample, and the other (probe) detects the changes induced in the sample by the pump pulse, with some time delay with respect to the pump pulse.

The THz spectroscopy is used as a method to identify transitions between excitonic eigenstates because exciton binding energy in GaAs QW ranges up to 10meV, which corresponds to the THz frequency range (4.14meV at 1THz) [3]. THz spectroscopy can be used to probe the change in optical response of microcavity through light transmission, reflection, and absorption. In this work, optical pulse is applied to the system, creating polaritons, followed by a single-cycle THz pulse, removing their exciton component. When these pulses are applied to the coupled exciton-photon state, each mode can be thought of as a harmonic oscillator driven by the time dependent electric field of the pulses. Fig.1.4 shows the intraexciton transition implemented by
the optical and THz excitation. We can form 1s excitons by exciting electrons with an optical pulse with photon energy lower than the band gap of GaAs. Frequency of the THz pulse can be tuned so that the process essentially destroys the exciton-photon coupling by exciting the excitonic 1s state to higher excitonic states [1]. The THz pulse essentially converts the coherent s-type polarization into p-type incoherent exciton populations which cannot couple to light.

There are two normal mode resonances in the reflection spectrum of the optical pump pulse as a result of the exciton-photon coupling, and the reflection changes depending on the delay between the optical and THz excitations. Therefore, time-evolution of exciton-photon coupling in QW microcavity can be studied by recording the change in reflection spectrum of the optical pulse while varying the delay. The effects of THz field propagating in semiconductor microcavities possess nonlinearities and provide insights into the quantum coherence in semiconductor nanostructures [3].

Figure 1.4: **Exciton energy level diagram and the resonant pulsed excitation of the 1s exciton state.** 1s exciton forms when an electron is excited from the ground state to the 1s excited state with an applied optical pump pulse. THz pulse applied after some time delay transfers most of the polarization to the higher energy states [1]. Conduction band is represented as the continuum, and the exciton energy levels are just below the conduction band.
Chapter 2

Theory

2.1 Coupled harmonic oscillators

The time-evolution of exciton-polariton dynamics is simulated using a classical model of coupled harmonic oscillators driven by optical pulse and THz pulse. Fig.2.1 shows the model used to demonstrate the exciton-photon coupling in a QW microcavity.

Figure 2.1: The classical model of system. Two harmonic oscillators with spring constant $k_e$ and $k_c$ represent the exciton mode and cavity photon mode, respectively. Coupling constant $\Omega = \sqrt{k/m}$ is determined by the middle spring. Springs are anchored to the fixed boundaries.

Harmonic oscillator with mass $m_1$ and spring constant $k_e$ represents the exciton mode, and the oscillator with $m_2$ and $k_c$ represents the photon mode. To make the calculation simpler, I use the same mass $m_1 = m_2 = m$ for both exciton mode and photon mode. The two modes oscillate at frequencies $\omega_e = \sqrt{k_e/m}$ and $\omega_c = \sqrt{k_c/m}$. Displacement of each mass is denoted by $x_i(t)$. The relative strength of spring constants determines the energy separation of the two normal modes. We will call $\Omega = \sqrt{k/m}$ as coupling constant. Optical pulse (blue) excites the exciton mode, and $m_1$ starts to oscillate. Energy is then coherently exchanged between the two modes. A single-cycle THz pulse (red) is applied to the system after some time delay $\Delta t$, removing the exciton component when it’s strong.
2.2 Equations of Motion

We start by briefly going over the coupled oscillators and normal modes to understand the normal mode coupling. The equations of motion for each harmonic oscillator in Fig.2.1 are given by

\[ \ddot{x}_1 = -\frac{k_e}{m}x_1 - \frac{k}{m}(x_1 - x_2) \]  
\[ \ddot{x}_2 = -\frac{k_e}{m}x_2 - \frac{k}{m}(x_2 - x_1). \]  

To solve equations (2.1) and (2.2), it is useful to use the matrix form

\[ M\ddot{x} = -Kx \]  

where

\[ x = \begin{bmatrix} x_1 \\ x_2 \end{bmatrix}, \quad M = \begin{bmatrix} m & 0 \\ 0 & m \end{bmatrix} \quad \text{and} \quad K = \begin{bmatrix} k_e + k & -k \\ -k & k_c + k \end{bmatrix}. \]  

We can make an ansatz to (2.3) as

\[ x_j(t) = A_j e^{-i\omega_{\pm} t} \]  

where \( A_j \) is the amplitude of oscillation and \( \omega_{\pm} \) are the normal mode frequencies. Using (2.5), equation (2.3) can be expressed as

\[ \omega_{\pm}^2 M x = K x \]  

Now we calculate the eigenvalues and eigenvectors of (2.6). The eigenvectors are normal modes.

\[ \begin{cases} 
    \begin{align*}
    x_1(t) &= A_1 \cos \omega_- t \\
    x_2(t) &= A_1 \cos \omega_- t
    \end{align*}
\end{cases} \quad \begin{cases} 
    \begin{align*}
    x_1(t) &= A_2 \cos \omega_+ t \\
    x_2(t) &= -A_2 \cos \omega_+ t
    \end{align*}
\end{cases} \]  

Note that the equations of motion should be real. Here, \( \omega_- \) and \( \omega_+ \) are the frequencies of each normal mode. Notice that in one of the normal modes, the two oscillators are oscillating in phase, whereas in the other normal mode, oscillations are completely out of phase. In the exciton-photon coupling in QW microcavity, these two normal modes are known as the upper polariton branch (UPB) and the lower polariton branch (LPB). The frequencies of oscillation \( \omega_+ \) and \( \omega_- \) can be found by calculating the eigenvalues of \( K \).

\[ \omega_{\pm} = \sqrt{\frac{1}{2} \left( \Omega^2 + \omega_c^2 + \omega_0^2 \pm \sqrt{\Omega^4 + (\omega_c^2 - \omega_0^2)^2} \right)} \]  

Any solution to (2.3) can be expressed as a linear combination of these two normal modes.

Fig.2.2 describes an anti-crossing behavior of the two harmonic oscillators. The energy separation between the normal modes depends on the frequency difference \( \delta = \omega_+ - \omega_- \) between the cavity mode and the exciton mode. We will call \( \delta \) as the cavity detuning because \( \omega_c \) is varied while \( \omega_e \) is kept constant. In the figure, \( \delta \) is negative when exciton mode frequency \( \omega_e \) is larger than the cavity mode frequency \( \omega_c \). Thus, \( \delta \) is positive when cavity mode has a higher frequency. An exciton-polariton is a half-light half-mater quasiparticle, and thus the relative portion of exciton mode depends on the cavity detuning. (2.8) can be written as

\[ \omega_{\pm} = \sqrt{\frac{1}{2} \left( 2\Omega^2 + \delta^2 + 2\delta \omega_e + 2\omega_0^2 \pm \sqrt{4\Omega^4 + (\delta^2 + 2\delta \omega_e)^2} \right)} \]
for \( \delta > 0 \), and
\[
\omega_{\pm} = \sqrt{\frac{1}{2} \left( 2\Omega^2 + \delta^2 - 2\delta \omega_e + 2\omega_e^2 \pm \sqrt{4\Omega^4 + (\delta^2 - 2\delta \omega_e)^2} \right)}
\]  
(2.10)
for \( \delta < 0 \). In the case of \( \omega_e = \omega_c \) (\( \delta = 0 \)), the splitting, which we define as \( \Delta = \omega_+ - \omega_- \), is at its minimum. The two modes are in resonance, and the coupling between them is the strongest at this point. The minimum energy separation is adapted by the Ref. [1], which is \( \Delta = 6.4 \text{meV} \).

When \( k_c = k_e \), i.e. when \( \omega_c = \omega_e \), we can use substitutions \( \omega^2 = (k_e + k)/m \) and \( \Omega^2 = k/m \) to reduce (2.8) as
\[
\omega_{\pm} = \sqrt{\omega^2 \pm \Omega^2}
\]  
(2.11)
We can relate \( \Delta = \omega_+ - \omega_- \) with \( \Omega \) as \( \Delta \approx \Omega^2/\omega = 6.4 \text{meV} \). Here, \( \omega \) is the average photon frequency of the optical pulse, and this is adapted by Ref. [1] as \( h\omega = 1.489 \text{eV} \). This leads to \( \omega/2\pi = 360.1 \text{THz} \), which gives the coupling constant \( \Omega/2\pi = 23.6 \text{THz} \).

Figure 2.2: **Avoided crossing.** Anti-crossing of normal mode coupling. Detuning is the frequency difference \( \omega_c - \omega_e \). When the detuning is zero, splitting is minimum.

We now add a term which accounts for the optical pulse as a driving force. Electric field of the optical pulse has a function
\[
E(t) = E_{\text{opt}} e^{-\frac{(t-t_{\text{opt}})^2}{\tau_{\text{opt}}^2}} \sin \omega_c t.
\]  
(2.12)
Here, \( t_{\text{opt}} \) is the peak time, and \( \tau_{\text{opt}} = 0.1 \text{ps} \) and \( E_{\text{opt}} = 1 \text{V} / \text{m} \) are the duration (FWHM) and the amplitude of the optical pulse, which are adapted from Ref [1]. The factor \( a = 4\ln 2 \) in the Gaussian function comes from normalizing its amplitude.

To account for the losses in the system caused by the exciton decoherence and the photon leaking from the cavity, we must add damping forces \( m_i \gamma \dot{x}_i \) to our ODE. We consider a microcavity designed with a high quality factor which leads to the same value of decay rate \( \gamma = 1.519 \text{Hz} \) (adapted from Ref. [1]) for both exciton mode and the photon mode. When the rate of coupling between the exciton and the cavity field is much larger than this decay rate, the elementary excitations are polaritons, and the system is in a strong coupling regime. Adding (2.12) and the damping force, (2.1) and (2.2) become
\[
\ddot{x}_1 = -\omega_e^2 x_1 + \Omega^2 x_2 - \gamma \dot{x}_1 + E_{\text{opt}} e^{-\frac{(t-t_{\text{opt}})^2}{\tau_{\text{opt}}^2}} \sin \omega_c t
\]  
(2.13)
\[ \ddot{x}_2 = -\omega_c^2 x_2 + \Omega^2 x_1 - \gamma \dot{x}_2. \] (2.14)

2.3 THz field effect

THz pulse removes most of the exciton component of exciton-polariton by exciting the 1s exciton to higher energy states. In this work, instead of directly driving the system with the THz excitation, motion of the exciton mode is attenuated at a specific time by incorporating the THz excitation in the damping force. The damping force is determined by the time dependent damping factor \( \gamma_1(t) \), which consists of a Gaussian function as an approximation to a single cycle THz pulse function. This damping force diminishes the energy of the exciton mode at a time \( t_{\tau_{THz}} \). The effect of THz pulse depends on its arrival time. Since THz field only interacts with the exciton mode and has no effect on the cavity photon mode, when the exciton component of polariton is strong, the effect is the largest, strongly changing the subsequent exciton motion. On the other hand, the effect is the smallest when the cavity field component of polariton is strong. Therefore, motion of the oscillators depends on the time delay of THz pulse with respect to the optical pulse. The electric field of THz pulse has a function

\[ E(t) = E_{THz} e^{-\frac{-a(t-t_{\tau_{THz}})^2}{\tau_{THz}^2}} \cos(2\pi v(t-t_{\tau_{THz}})) \] (2.15)

where \( t_{\tau_{THz}} \) is the peak time, and \( \tau_{THz} = 1 \)ps and \( v = 1 \)THz are duration and frequency of the THz pulse adapted from Ref.[1]. Most of excitonic energy transition happens at the peak of THz pulse because of extreme nonlinear characteristics of THz field [3]. Thus, one can approximate (2.15) as a Gaussian function. In fact, this work uses a very short THz pulse duration of \( \tau_{THz} = 50 \)fs, and (2.15) becomes a Gaussian. We can introduce a strong damping effect by incorporating this approximation to the damping factor of exciton mode. Therefore, \( \gamma \) in (2.13) is replaced by

\[ \gamma_1(t) = E_{THz} e^{-\frac{-a(t-t_{\tau_{THz}})^2}{\tau_{THz}^2}} + \gamma \] (2.16)

where \( \gamma = 1.519 \)Hz. (2.13) then becomes

\[ \ddot{x}_1 = -\omega_c^2 x_1 + \Omega^2 x_2 - \left[ E_{THz} e^{-\frac{-a(t-t_{\tau_{THz}})^2}{\tau_{THz}^2}} + \gamma_2 \right] \dot{x}_1 + E_{opt} e^{-\frac{-a(t-t_{opt})^2}{\tau_{opt}^2}} \sin \omega_c t. \] (2.17)

The simulation code is written in MATLAB R2019a. The solutions to the equations (2.17) and (2.14) are solved numerically using the built-in function ode45 in MATLAB with initial conditions of \( x_1(0) = x_2(0) = \dot{x}_1(0) = \dot{x}_2(0) = 0 \). Computation is performed controlling three parameters: THz field amplitude \( (E_{THz}) \), peak time of THz pulse \( (t_{\tau_{THz}}) \), and the FWHM of THz pulse \( (\tau_{THz}) \). Data sets are stored on a cloud storage.

2.4 Simulation of polariton dynamics

Time-evolution of the polariton oscillation is simulated by computing the difference in the power spectrum \( \Delta I = I_{THz} - I_0 \) with \( (I_{THz}) \) and without \( (I_0) \) the THz pulse as a function of time delay \( (\Delta t = t_{\tau_{THz}} - t_{opt}) \) between the peak times of optical \( (t_{opt}) \) and THz pulse \( (t_{\tau_{THz}}) \). Power
spectrum is obtained by Fourier transforming the solutions to the equation of motion in time domain to frequency domain. The Fourier transform is computed using the built-in function, \texttt{fft}, in MATLAB. The peak time of optical pulse is kept constant at $t_{\text{opt}} = 3\text{ps}$, and the delay between the optical and the THz pulse is varied by changing $t_{\text{THz}}$ in the range of $t_{\text{THz}} = 2\text{ps}$ to $t_{\text{THz}} = 7\text{ps}$ with 0.025ps interval.
Chapter 3

Results and Discussion

3.1 Solution in time domain

To compute the exciton-polariton spectra, the equations of motion for both exciton and photon modes are solved numerically, and then Fourier transformed into frequency domain. The polariton oscillation is represented by the exciton-polariton spectra as a function of time delay between the THz and optical pulses. Fig.3.1 shows the computed solutions for the equations of motion for the exciton mode (blue curve) overlapped on the solution for the photon mode (red curve) at $\Delta t = 0.63\text{ps}$ (Fig.3.1(a)) and $\Delta t = 0.95\text{ps}$ (Fig.3.1(b)). The optical pulse excites the system at $t_{\text{opt}} = 3\text{ps}$, and the THz pulse arrives to the system at $t_{\text{THz}}$, which is indicated as a dashed green line. THz field amplitude of $E_{\text{THz}} = 15V_m$ and duration of $\tau_{\text{THz}} = 0.05\text{ps}$ were used to compute the solutions. Depending on the values of $E_{\text{THz}}$ and $\tau_{\text{THz}}$, phase shift between the gray curve and the oscillation of each mode (blue and red curves) becomes apparent. In order to minimize the phase shift, the values of $E_{\text{THz}}$ and $\tau_{\text{THz}}$ were optimized. The minima of exciton mode oscillation coincide with the maxima of photon mode oscillation and vice-versa.

Figure 3.1: Exciton and cavity photon modes in time domain. Gray curves are the solutions without the THz reset pulse. Optical pulse is applied at $t_{\text{opt}} = 3\text{ps}$, exciting the exciton mode. The green dashed lines at 3.63ps in (a), and 3.95ps in (b) indicate the peak times of THz pulse.
Coherent energy exchange is clearly observed through the beating effect of the oscillations. Motions are attenuated due to the energy loss of the system and the THz field effect. At $t_{THz} = 3.63\text{ps}$, the exciton component dominates the polariton oscillation, and thus subsequent exciton dynamics is strongly modified (Fig.3.1(a)). The very short duration of THz pulse $\tau_{THz} = 0.05\text{ps}$ leads to an abrupt change in subsequent exciton mode motion. At $t_{THz} = 3.95\text{ps}$, cavity field component dominates the polariton oscillation, so the subsequent dynamics of the system remain unchanged (Fig.3.1(b)).

3.2 Exciton-polariton spectra

Figure 3.2 shows the Fourier transform of the motion of the exciton mode with $\Delta t = 0.05\text{ps}$ (red line), $\Delta t = 1.3\text{ps}$ (black line), without the THz pulse (gray-shaded area), and the uncoupled exciton resonance (blue dashed line). The splitting shown here is called the normal mode splitting. The peaks separated by $\Delta = \omega_+ - \omega_- = 6.4\text{meV}$ are the normal modes explained in the theory section. Single peak in the middle is the uncoupled exciton mode with a frequency of oscillation $\omega = 360.1\text{THz}$. In the QW microcavity, one can measure the transmission of the optical pulse through the cavity using the method of spectroscopy. The two peaks that appear in the cavity transmission spectrum are known as the upper (UPB) and lower (LPB) branches of polaritons, and they correspond to the normal modes shown here in the classical case. In cavity QED, this splitting is called the vacuum Rabi splitting, which signifies the strong coupling of the cavity photon mode with the two-level atom placed within a resonant cavity. In the strong coupling regime of cavity QED, the transmission resonance of the cavity splits with a separation called the vacuum Rabi frequency.

![Figure 3.2: Normal mode splitting](image)

The LPB and UPB at different cavity detuning $\delta$ at $\Delta t = 1.0\text{ps}$ with $E_{THz} = 15V/m$ and $\tau_{THz} = 0.05\text{ps}$ are presented in Fig.3.3. The frequencies of the two peaks increase as detuning
increases from $-2\text{meV}$ to $+1\text{meV}$. LPB has an increased cavity-field component when the detuning is large negative, and has an increased exciton component when the detuning is large positive. The opposite is true for the UPB [24]. The intensities decrease as cavity mode frequency increases. As magnitude of detuning increases, the two modes become nonresonant and the energy separation increases as shown in Fig.2.2.

Figure 3.3: Cavity detuning dependent power spectrum. Computed $\Delta I$ of the exciton mode with cavity detuning from $\delta = -2\text{meV}$ to $1\text{meV}$.

### 3.3 Simulated polariton oscillation

Fig.3.4 shows the simulated polariton oscillation as a function of time delay between the two pulses. Here, zero delay means that peaks of the optical and THz pulses arrive to the system simultaneously. Delay is negative when the THz pulse arrives earlier than the optical pulse. Since the effect of the THz pulse is associated with the damping of the exciton mode, the motion of the system remains unchanged when $\Delta t < 0$, and therefore $\Delta I = 0$. In the QW in microcavity, the two resonances correspond to LPB and UPB as shown in Fig.3.4 (a). The oscillation shows an interference pattern between the LPB and UPB. This is caused by the interference of the optical and THz excitations with a frequency inversely proportional to the time delay. Fig.3.4 (b) shows a slice through the power spectrum in Fig.3.4 (a), measured at the UPB. The energy separation $\Delta = 6.4\text{meV}$ in Fig.3.2 corresponds to the oscillation period of 0.646ps, which is shown by the gray-shaded area in Fig.3.4 (b). The dependence of oscillation period on the cavity detuning was explored, but the classical model could not demonstrate the expected relationship presented in the Fig.5 of Ref.[1]. Computed polariton oscillation for $\delta = -2\text{meV}$ is shown in appendix.
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Figure 3.4: **Computed polariton oscillation.** (a) Computed differential power spectrum (ΔI) at zero detuning (δ = 0) is plotted as a function of time delay between the optical and THz pulse. (b) Slice through the upper polariton branch at ω_+ = 360.9THz.

Fig.3.5 shows the result of Ref.[1] where polariton oscillation in QW microcavity was computed using the theory of Maxwell–semiconductor Bloch equations extended to THz fields. The difference in reflectivity ΔR = R_{THz} − R_0 with (R_{THz}) and without (R_0) the THz pulse is computed as a function of the time delay between the optical and THz pulses.

Figure 3.5: **Computed polariton oscillation.** Computed differential reflectivity ΔR of the microcavity at zero detuning is plotted a a function of energy and time delay. Figure taken from Ref. [1].

Here, HEP and LEP stand for the higher and lower energy peaks, respectively, which correspond to the two peaks in Fig.3.4 (a). In this work, optical reflection spectrum of the cavity is computed by Fourier transforming the reflected and incident optical fields, and calculating the reflectance R, which is defined as $R = |E_{0r}/E_{0i}|^2$. $E_{0r}$ and $E_{0i}$ are the amplitudes of reflected field and incident field, respectively. This result in comparison with Fig.3.4 (a) shows that using a classical model of coupled harmonic oscillators with optical and THz excitations can indeed demonstrate the dynamics of the exciton-photon coupling in semiconductor QWs, which is inherently quantum mechanical.
Chapter 4

Conclusion

The exciton-polariton dynamics in a semiconductor microcavity is demonstrated computationally using a classical model of coupled harmonic oscillators representing the exciton mode and cavity photon mode. As an analog to the optical pump pulse applied to the QW microcavity, the system is driven by the electric field of the optical pulse which excites the exciton mode. As a result, normal mode coupling associated with a coherent energy exchange between the exciton mode and the cavity photon mode is shown. A single-cycle THz pulse is applied to the exciton mode after some time delay and attenuates the motion of the exciton mode. The effect of the THz pulse is applied to the system by employing the time-dependent damping of the exciton mode.

Computation is performed controlling the peak time, amplitude, and duration of THz pulse. The peak time of the optical pulse is kept constant, and thus the delay between the two pulses depends on the peak time of the THz pulse. Method of time-dependent damping causes a phase shift between the oscillators’ motion with and without the THz pulse, and the the magnitude of phase shift depends on the amplitude and duration of the THz pulse. The amplitude and duration that lead to a reasonably small phase shift are found and used to compute the polariton oscillation.

Computed power spectrum as a function of time delay between the optical and THz pulse successfully describes the polariton oscillation in a QW microcavity, which is inherently quantum mechanical. The beats apparent in the solutions to the equations of motion show the coherent coupling of the photon mode and the exciton mode, and the computed normal-mode splitting is a signature of the strongly coupled system. This thesis shows that the hybridization of the cavity photon mode and the exciton mode, i.e. the light-matter interaction in a semiconductor optical microcavity, can be demonstrated using classical coupled harmonic oscillators driven by optical and THz pulses. It is then shown that computed exciton-polariton oscillation is the coherent energy exchange between the excition and photon modes. The effects of cavity detuning on the exciton-polariton spectra is also explored.

Solutions to the equations of motion were also solved using time-dependent mass of the exciton mode, although its results are not presented in this thesis. The reason is because solutions in the time-domain did not show any beats in oscillators’ motion after arrival of the THz pulse.
Future work may provide the exact analytical solutions to the equations of motion. Time
dependent frequency of the exciton mode can also be explored to solve for the motions. Cou-
pled harmonic oscillators or simple harmonic oscillator have been widely used to study various
branches of physics. Mandal explores the squeezing states of the quantum driven harmonic
oscillator coupled to coherent light using scaled wronskian [25], and Hertzog et.al. explains
the quantum description of strong light-matter hybridization in a cavity [9]. Exciton-polariton
dynamics can also be studied using coupled quantum harmonic oscillators. The system and
its solutions in this work are entirely classical, and classical mechanics cannot fully explain
the quantum aspects of polariton dynamics since vacuum fluctuations are not treated in the
classical model.
Bibliography


A.1 Cavity detuning dependent polariton oscillation

The polariton oscillation with cavity detuning $\delta = -2\text{meV}$ is shown in Fig.A.1. The energy separation is $\Delta = 6.9\text{meV}$ which corresponds to the oscillation period of 0.55ps, which is shown by the gray-shaded area in Fig.A.1 (b). Unlike the result of zero detuning shown in Chapter 3.3, UPB has a higher intensity than the LPB for both with and without the THz pulse. The cavity detuning dependence of the polariton oscillation period was investigated. The result did not follow the trend shown in Fig.5(b) of Ref.[1]. The cause of inability to demonstrate the cavity detuning dependence is thought to be the model used in this work being entirely classical whereas QW exciton-polariton has quantum features. This thesis shows that a classical model of coupled harmonic oscillators is capable of demonstrating the exciton-polariton dynamics, but the model’s classical features may cause limitations on its capability. However, there may be other causes which are not clear at this moment.

Figure A.1: Polariton oscillation with $\delta = -2\text{meV}$. (a) Computed differential power spectrum ($\Delta I$) at $\delta = -2\text{meV}$ is plotted as a function of time delay between the optical and THz pulse. (b) Slice through the upper polariton branch at $\omega_+ = 360.76\text{THz}$. 