NONLINEAR OPTICAL PROPERTIES OF QUANTUM WELL STRUCTURES IN THE PRESENCE OF INTENSE INFRARED LASERS

by

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Abstract

We theoretically and experimentally investigate the non-linear optical properties of undoped and n-doped quantum wells (QW's) in the presence of intense infrared lasers. These lasers are polarized along the QW growth directions and near resonance with the transitions between the conduction subbands. In n-doped QW's by inclusion of electron-electron scattering and dynamic screening we study for the first time: (i) the effects of the frequencies and intensities of the infrared fields on the dephasing rates associated with the intersubband transitions, (ii) the generation of various nonlinear gain processes, and (iii) coherent and optical processes such as coherent population trapping, dark line and laser induced transparency effects. In undoped QW's by presenting a consistent theory we show how one can use infrared coupling of the conduction subbands of a QW to coherently manipulate absorption and emission spectra of excitons. We discuss these effects in terms of multi-photon coupling of the excitons or electrons, quantum interferences, and by presenting a new model to treat laser-matter interaction. Following these developments we put some of our own results to test by observing the emission dynamics of excitons in the presence of an intense CO$_2$ laser. We report the first observations of infrared enhancement of the effective non-radiative decay rates of excitons and the effects of multi-level transition processes in the emission of QW's. This thesis also includes an extensive analysis of nonlinear effects in atomic systems. While the primary motivation for this analysis was to understand and design nonlinear optics of QW's, in the process some original contributions were made to the study of nonlinear optics of atomic systems.
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Chapter 1

Introduction

Since the pioneering work of Esaki and Tsu in 1970 [1], the study of two-dimensional semiconductors or quantum wells (QW's) has developed rapidly, both from a physics point of view and in practical applications. A significant feature of QW's is that the carriers are confined along the growth direction, \( z \). Therefore, although they can move freely in the plane of the QW, their energy states along the \( z \)-axis are quantized. This gives QW's a distinct feature: by varying their growth parameters, the spacing between these quantized levels or subbands can be adjusted to fall within the infrared frequency range. This frequency range has many scientific and technologic applications. Another important feature of QW's is that for optical fields polarized along the QW growth direction, the dipole moments associated with the transitions between the subbands can be very large [2].

These interesting features and the fact that QW's can be doped in various ways have already paved the way for many applications, such as infrared detectors [3], modulators, switches [4], lasers [5] etc. However, in addition to these features, QW's are unique in that their intrinsic quantum mechanical properties can be manipulated. For example, by using proper growth parameters one can design a QW structure such that its subbands have either well-defined or ill-defined parities. In other words we can engineer the electron wave-functions and transition dipole moments. Combinations of these exotic features have made QW's appealing hosts for studying nonlinear optics, and promising candidates for future photonic and optical devices.

However, despite the possibility of revolutionary applications and the intensive work
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that has been done, the study of nonlinear optics of QW's is still in its infancy. There are two major reasons for this: First, QW systems are complicated. Various effects such as carrier-carrier scattering, band structures, impurities and imperfections, etc, make investigation of these systems difficult. In addition, the interaction of these systems with intense laser fields causes more complexity. Because of this, as we will show in this thesis, some basic and rather trivial effects in the optics of QW's have not yet been studied. The second reason is that in many cases the investigation and design of nonlinear optics in QW's requires bridging to the optics of atomic systems. As will be shown in this thesis, the search for new coherent effects in atomic systems is not only rewarding in itself, but it also provides some basic building blocks for new developments in QW optics. This is particularly evident in the cases where the optical coherent processes involve mixing of several levels of a system by one or more intense near resonance lasers. These processes generate exotic nonlinear effects in atomic systems, such as laser induced transparency [6], index of refraction enhancement [7], lasing without inversion [8], etc.

Near resonance interactions with lasers are also the backbone of nonlinear optical processes in QW systems, and the coherent manipulation of their optical properties. For example, by inducing quantum interferences they can lead to the generation of currents with preferred directions in the QW continua [9], gain without inversion [10], etc. Also by dressing two conduction subbands with an intense infrared laser, the linear interband absorption of an undoped QW can be modified (interband laser induced transparency) [11]- [15]. In n-doped QW's such a dressing process was used to study saturation effects [16]- [17]. In addition to these, intense lasers were used to generate optically-pumped and Raman gain processes in a wide range of frequencies [18] and to create light-modulators [19]. Even at this level of work, however, one can find some major shortcomings. For example, in the cases where the interband laser-induced transparency was studied, either the Coulomb interaction between electrons and holes was ignored [12]- [15] or the quantum mechanical properties and realistic coupling of excitons
were not considered [11]. Also in all cases where nonlinear effects of n-doped QW’s were studied, the important effects of the laser fields on the dephasing rates associated with transitions in the conduction bands were overlooked [16, 17, 19].

1.1 Thesis objectives

Our objectives in this thesis are to theoretically study nonlinear coherent effects of QW and atomic systems, and to put some of our own theoretical results to test experimentally. In addition to presenting proper treatments for some of the effects which have already been discussed, a major part of our theoretical development is devoted to a comprehensive study of new nonlinear optics in QW’s. This includes quantum interference effects, coherent manipulation of the optical properties of QW’s and electron distributions, and prediction of new gain processes. Here we not only introduce new options for conventional application of QW’s, such as lasers, infrared detectors, modulators, etc., but also shed light into some new topical subjects. The experimental results also provide some insight into the dynamics of excitons in infrared-coupled QW’s and reveal the effects of mixing of internal states of excitons in their emission spectra. The highlights of these developments are as follows:

1.1.1 Theoretical developments

A. Atomic systems:

The first section of our theoretical development is devoted to atomic systems. The results of these investigations have been published in three papers [20]-[22]. Here we specifically discuss the following novel effects:

1. Generation of two-frequency laser induced transparency. Here we present a new type of laser induced transparency (LIT) in atomic systems which makes a medium transparent at two tunable frequencies. This development may have some impact on the utilizations of LIT, such as lasing without inversion [chapter 4].
2. Manipulation of the emission spectrum of an atom by inducing multi-photon quantum interferences. One dramatic effect predicted is that by creating constructive quantum interferences one can narrow the emission spectrum of an atom. Here the intensity of the emission line-center increases while that of its tails decreases. We also show how these interferences can generate a two-frequency dark line effect in the emission spectra of an atom [chapter 4].

3. The most common picture used to describe resonant laser-matter interaction is the dressed-state picture. This picture, however, has no application at low laser intensities where we expect to see interesting and fundamental phenomena. Here we present a new alternative picture which is compatible with the dressed-state picture at high field intensities and successfully describes the nearly resonant laser-matter interaction at low laser intensities [chapters 3 and 4].

4. We use this alternative picture to show how an atomic transition can generate a very narrow gain spectrum, and why it switches to a narrow absorption spectrum as the atom-field coupling conditions change. The gain occurs in the absence of any kind of inversion [chapter 4].

B. Quantum well systems:

Grasping the basic underlying physics of these developments we use them to study new optical coupling processes and quantum interferences in QW's. This is done using one or two infrared lasers to coherently couple two or more conduction subbands. The results of this part have been published in eight papers [23]-[30]. Specifically, in this thesis we discuss the following:

1. Coherent modification of the interband excitonic emission and absorption spectra of QW's. Here we study the infrared coupling of excitons when the coupled conduction subbands are probed from the valence band. We also discuss the selection rules for the excitonic transitions caused by infrared fields, the exciton multi-photon
coupling processes, and the effects of the Coulomb interaction between holes in the infrared-coupling of excitons [chapters 5 and 6].

2. Nonlinear optical processes and coherent effects when infrared coupling of conduction subbands occurs in n-doped QW's. Here we address the following new developments:

(a) Various types of laser induced transparencies and multi-photon coupling effects in the conduction intersubband transitions [chapter 7].

(b) Effects of the electron-electron scattering processes in the conduction intersubband transitions. Specifically, here we show how these processes allow the infrared fields to affect the polarization dephasing rates associated with these transitions. This also includes predictions of steady state non-Fermi distributions in the conduction subbands. These are major steps towards a more realistic treatment of infrared coupled n-doped QW's than those used in the previous studies [chapter 8].

(c) Prediction of new types of gain processes in n-doped QW's. These include hyper-Raman, extended hyper-Raman and stimulated Rayleigh processes [chapters 8 and 9].

(d) Prediction of coherent population trapping and some other quantum interferences in QW's [chapter 9].

1.1.2 Experimental investigations

In our experimental part we address the evolution of the exciton emission spectra in the presence of the coherent coupling of two conduction subbands. The results of this part reveal the following [chapter 11]:

1. Dynamics of excitons in an infrared-coupled QW and infrared enhancement of the effective non-radiative decay rates of excitons.
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2. Multi-level coupling effects and their roles in the coherent and incoherent dynamics of the infrared-coupled QW emission spectra.

1.2 Thesis outline

This thesis is organized in twelve chapters. Except for chapters 3 and 5 which include some introductory material, all work presented is original. In Fig.1.1 we show a road map for reading various chapters of this thesis and how they are connected together. Briefly, in chapter 2 we present the core calculations addressing the linear responses of multi-level systems driven by one or two laser fields. These calculations are used for the atomic and QW systems considered in this thesis. More extensive calculations are carried out for the cases of QW's in the following chapters. In chapter 3 we apply the formalism developed in chapter 2 to some simple atomic systems. Although this chapter contains some new developments, it mostly serves as an introductory chapter. Our principal developments for the atomic systems are presented in chapter 4.

In chapter 5 we discuss the basic physics of excitons interacting coherently with infrared fields. Here we first present a customized theory of the interband excitonic transitions in the absence of any infrared field and then discuss the infrared transitions between exciton states. Following this we study how the interband excitonic absorption and emission spectra of QW's evolve in the presence of strong infrared coupling of the exciton states. In chapter 6 we use an extension of the formalism of chapter 5 to discuss some previously unexplained phenomena observed in an experiment conducted by Fröhlich et. al. [9]. This experiment examined the effects of an intense CO$_2$ laser on the interband absorption spectra of a QW.

Chapters 7, 8 and 9 contain our developments for the optical properties of n-doped QW's coupled by intense laser fields. In chapter 7 we study various types of laser induced transparencies and multi-photon coupling processes in the conduction bands. In chapter 8 we discuss the dynamic effects of electron-electron scattering on the intersubband
transitions, generation of non-Fermi distributions and various types of gain processes. In chapter 9, these are extended to more complex systems where they show extended Raman and Rayleigh gain processes, coherent population trapping of electrons, and some other quantum interference effects. In chapter 10 we outline the basic setup and optics of the experiment briefly. The results and analysis of the experiments are presented in chapter 11. The concluding remarks and some suggestions for future works are presented in chapter 12.
Chapter 2

Interaction of intense lasers with multi-level systems

2.1 Introduction

Resonant or nearly resonant interactions of lasers with solids and gases are the sources of much of nonlinear optics. Such interactions occur when the laser frequencies are comparable to the matter's transition frequencies. In this chapter we present a comprehensive study of these interactions in systems where one or two intense laser fields couple two or three states. This includes investigation of the linear responses of these systems to weak probe fields using linear response theory and the quantum regression theorem. These analyses are applied to atomic and QW systems in the following chapters to study a wide range of new optical nonlinear effects.

2.2 Coupling configurations, General description

The optical properties of an atomic or QW system can be drastically changed when it interacts with a single laser. The simplest case occurs when the laser is nearly resonant with a single transition of the system (Fig.2.1). In atoms, as studied by Mollow [32] and Grynberg and Cohen-Tannoudji [33] this can lead to saturation phenomena, partial gain and absorption spectra, quantum interference effects and stimulated Rayleigh scattering. As we discuss in chapter 8 and Ref. [24], in n-doped QW's, however, the analysis of this system is complicated. This is because here electron-electron scattering makes the system's damping rates dependent on the frequency and intensity of the coupling laser.

In the case of the system in Fig.2.1 the probe and coupling fields are nearly resonant with the same transition. A large number of interesting nonlinear effects, however, can
be explored in systems where either the lower or upper transition level of the probe field is one of the coupled levels. Using the common notations in atomic systems, here when the lower or upper coupled level is detected we have a $\Xi$ (Fig.2.2a) or a $\Lambda$ (Fig.2.2b) configuration, respectively. Also the system has a $V$ configuration when we detect a higher uncoupled state from the lower coupled transition level (Fig.2.2c). These configurations which have been studied extensively in the past [34], [35] can lead to laser-induced transparency (LIT) in which the system exhibits transparency at a frequency where it naturally has a high absorption, lasing without inversion [38], quenching or destruction of the spontaneous emission spectra [39], or generation of dark line effect [41]. Also these configurations lead to many interesting nonlinear effects in QW systems. As we discuss in the following chapters and reported in Refs. [25, 26, 30] these include laser-induced transparency in the excitonic and electronic transitions of QW’s and altering their emission spectra.

Another aspect of nonlinear optics which has technological and fundamental impact is the coherent manipulation of the optical properties of solids and atoms. Here we want to exploit the intrinsic properties of these materials, such as their damping rates, energy configurations, etc., to induce desired optical properties. To a limited extent this can be
Figure 2.2: Schematic diagrams of three-level systems driven by a single laser field. (a), (b) and (c) represent, respectively, Ξ, Λ and V configurations. Here $|0>$ refers to the ground state. Other specifications are the same as those in Fig.2.1.
achieved by employing the $V$, $\Xi$, and $\Lambda$ systems (Fig.2.2). One can expand this, however, to reach dynamic and rich cases by considering optically driven multi-level systems. Here, as shown schematically in Fig.2.3, two lasers couple three upper levels and the probe field is tuned from the ground level to any of these coupled levels. We designate these systems as $\Xi_-$, $K$, or $\Pi$ configurations, respectively, when the lower, middle or higher energy coupled level is detected. As we discuss in chapters 3 and 4 and reported in Refs. [20]-[22], in atomic systems the coherent manipulation provided by these systems can lead to LIT and dark line effects with adjustable zero absorption and dark frequencies. It also can narrow the line-width of the spontaneous emission spectrum while increasing its line-center intensity. As shown in this thesis and reported in Ref. [23, 25], in QW systems using these configurations one can coherently manipulate the intersubband and interband transitions.

The other two systems which are studied in the following chapters are $\Xi_d$ and $\Lambda_d$ systems (Fig.2.4). Here the three levels of the $\Xi$ and $\Lambda$ systems (Fig.2.2) are driven by two strong laser fields. Although the optical properties of these systems in atomic systems have been studied extensively in the past [35]-[36], we will show that these systems can still lead to new phenomena. These include generation of an ultra-narrow gain profile in the absence of any type of inversion [21] and its rapid switching into a narrow absorption profile. Quantum well systems with these configurations can cause generation of tunable gain with or without inversion in the intersubband transitions, coherent population trapping, etc. We have reported these results in Refs. [27, 28].

2.3 Equations of motion of systems with $\Pi$, $K$ and $\Xi_-$ configurations

In this section we develop a general formalism to study $\Pi$, $K$ and $\Xi_-$ systems. To start with, let us consider a system consisting of four levels, one ground ($|0>$) and three higher levels ($|i>$ $i=1,2,$ and 3). This system is driven by two laser fields with frequencies $\omega_1$ and $\omega_2$ nearly resonant with the transition between $|1>$ and $|2>$ (1-2) and that between
Figure 2.3: Schematic diagrams of the four-level systems driven by two laser fields. (a), (b) and (c) are called, respectively, Ξ, K and Π systems. |0⟩ refer to the ground state and |1⟩, |2⟩ and |3⟩ to the upper levels. $\omega_1$ and $\omega_2$ represent the frequencies of the coupling lasers near resonance with their corresponding transitions.
Figure 2.4: Schematic diagrams of (a) $\Xi_d$ and (b) $\Lambda_d$ system. All other specifications are the same as those in Fig.2.3.
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|2> and |3> (2-3), respectively. The coupling configuration of this system depends on which of the coupled levels is detected from the ground level. Based on our notations in a Ξ– (Fig.2.3a), K (Fig.2.3b), or Π configuration (Fig.2.3c), this level is |1>, |2> or |3>, respectively.

Within the electric-dipole and resonance approximations the interaction of two classical optical fields with the system is given by

\[ H_{\text{int}} = -d_{12}(t)E_1(t) - d_{23}(t)E_2(t). \]  

(2.1)

\(E_1(t)\) and \(E_2(t)\) refer to the optical fields where each can be written as follows

\[ E_i(t) = E_i^+ + E_i^- . \]  

(2.2)

Here \(E_i^+\) and \(E_i^-\) are the positive and negative frequency parts of the fields. In terms of the field quantization, these terms correspond to absorption and emission of photons, respectively. Under the resonant approximation we may only consider positive frequency parts of the fields \((E_i^+(t) = E_i e^{-i\omega_it})\). This is called rotating-wave approximation. Also we drop the superscript (+) for simplicity. \(d_{ij}\) in Eq. (2.1) is the transition dipole moment between the \(i\)th and \(j\)th levels given by

\[ |d_{ij}| = \mu_{ij}a_i^\dagger a_j + \mu_{ij}^* a_j^\dagger a_i. \]  

(2.3)

Here \(\mu_{ij} = <i|d|j>\) in which \(|i>\) and \(|j>\) correspond to the \(i\)th and \(j\)th levels. \(a_i (a_i^\dagger)\) is the annihilation (creation) operator for an electron in state \(|i>\). In the interaction picture, therefore, we have:

\[ H'_{\text{int}} = H_1 + H_2, \]  

(2.4)

where

\[ H_1 = h g_{12}\{e^{i\Delta_1t}a_2^\dagger a_1 + e^{-i\Delta_1t}a_1^\dagger a_2\}, \]  

(2.5)

and

\[ H_2 = h g_{23}\{e^{i\Delta_2t}a_3^\dagger a_2 + e^{-i\Delta_2t}a_2^\dagger a_3\}. \]  

(2.6)
Here $H_1$ is the interaction term corresponding to the 1-2 transition and $H_2$ is that of the 2-3 transition. $g_{12} = -\mu_{12}E_1/\hbar$ and $g_{23} = -\mu_{23}E_2/\hbar$ are the Rabi frequencies. The detunings of the fields are

$$\Delta_1 = \omega'_2 - \omega'_1 - \omega_1,$$

and

$$\Delta_2 = \omega'_3 - \omega'_2 - \omega_2,$$

where $\hbar\omega'_i$ is the energy of $|i\rangle$.

Substituting $H'_\text{int}$ in the following equation

$$\frac{\partial \rho'}{\partial t} = -i\frac{\hbar}{\hbar}[H'_\text{int}, \rho'] + \frac{\partial \rho'}{\partial t}\big|_{\text{incoh}}, \quad (2.9)$$

one can obtain the equations of motion of the system. Here $\rho'$ is the density matrix in the interaction picture and $\frac{\partial \rho'}{\partial t}\big|_{\text{incoh}}$ represents the incoherent or damping part of the system. This term has various forms, depending on the system characteristics and its coupling configuration. We will discuss this in the case of QW systems in the following chapters in detail. After substitution of Eq. (2.4) in (2.9) and doing some algebra the equations of motion are found as follows:

$$\frac{\partial \rho'_{00}}{\partial t} = \frac{\partial \rho'_{00}}{\partial t}\big|_{\text{incoh}} + P_0 \quad (2.10)$$

$$\frac{\partial \rho'_{11}}{\partial t} = -ig_{12}(e^{-i\Delta_1 t} \rho'_{12} - e^{i\Delta_1 t} \rho'_{21}) + \frac{\partial \rho'_{11}}{\partial t}\big|_{\text{incoh}} + P_1 \quad (2.11)$$

$$\frac{\partial \rho'_{22}}{\partial t} = -ig_{23}(e^{-i\Delta_2 t} \rho'_{32} - e^{i\Delta_2 t} \rho'_{23}) - ig_{12}(e^{-i\Delta_1 t} \rho'_{21} - e^{i\Delta_1 t} \rho'_{12}) + \frac{\partial \rho'_{22}}{\partial t}\big|_{\text{incoh}} + P_2 \quad (2.12)$$

$$\frac{\partial \rho'_{33}}{\partial t} = -ig_{23}(e^{i\Delta_2 t} \rho'_{32} - e^{-i\Delta_2 t} \rho'_{32}) + \frac{\partial \rho'_{33}}{\partial t}\big|_{\text{incoh}} + P_3. \quad (2.13)$$

The equations of motion of the off-diagonal elements of the density matrix which determine the motion of the coherences are:

$$\frac{\partial \rho'_{12}}{\partial t} = ig_{23}e^{i\Delta_2 t} \rho'_{13} - ig_{12}e^{-i\Delta_1 t}(\rho'_{22} - \rho'_{11}) - \gamma_{12} \rho'_{12} \quad (2.14)$$
\[
\frac{\partial \rho'_{13}}{\partial t} = i g_{23} e^{i \Delta_2 t} \rho'_{12} - i g_{12} e^{-i \Delta_1 t} \rho'_{23} - \gamma_{13}(k) \rho_{13} 
\]

(2.15)

\[
\frac{\partial \rho'_{23}}{\partial t} = -i g_{23} e^{-i \Delta_2 t} (\rho'_{33} - \rho'_{22}) - i g_{12} e^{i \Delta_1 t} \rho'_{13} - \gamma_{23} \rho'_{23} 
\]

(2.16)

\[
\frac{\partial \rho'_{01}}{\partial t} = i g_{12} e^{i \Delta_1 t} \rho'_{02} - \gamma_{12} \rho'_{01} 
\]

(2.17)

\[
\frac{\partial \rho'_{02}}{\partial t} = i g_{23} e^{i \Delta_2 t} \rho'_{03} + i g_{12} e^{-i \Delta_1 t} \rho'_{01} - \gamma_{02} \rho'_{02} 
\]

(2.18)

\[
\frac{\partial \rho'_{03}}{\partial t} = i g_{23} e^{-i \Delta_2 t} \rho'_{02} - \gamma_{03} \rho'_{03}. 
\]

(2.19)

Here \( \rho_i \) refers the net scattering-in rate to \( |i> \) due to relaxation of the electrons in the upper levels, and \( \gamma_{ij} \) represents the dephasing rate of \( \rho'_{ij} \). These rates are added to Eqs. (2.10)-(2.19) in a phenomenological way. After making the following substitutions:

\[
\rho_{00} = \rho'_{00} \quad \rho_{32} = \rho'_{32} e^{-i \Delta_2 t}, 
\]

(2.20)

\[
\rho_{11} = \rho'_{11} \quad \rho_{21} = \rho'_{21} e^{-i \Delta_1 t}, 
\]

(2.21)

\[
\rho_{22} = \rho'_{22} \quad \rho_{31} = \rho'_{31} e^{-i \delta_{31} t}, 
\]

(2.22)

\[
\rho_{33} = \rho'_{33} \quad \rho_{30} = \rho'_{30} e^{-i \delta_{30} t}, 
\]

(2.23)

\[
\rho_{10} = \rho'_{10} e^{-i (\omega_2' - \omega_1') t} \quad \rho_{20} = \rho'_{20} e^{-i \delta_{20} t}, 
\]

(2.24)

where

\[
\delta_{20} = \omega_2' - \omega_0' - \omega_1, \quad \delta_{30} = \omega_3' - \omega_0' - \omega_1 - \omega_2 \quad \delta_{42} = \omega_4' - \omega_2' - \omega_1 - \omega_2, 
\]

(2.25)

Eqs. (2.10)-(2.19) take simple forms suitable to solve easily. For example, in the case of an atomic system with a \( \Xi_+ \) configuration the linear equations of motion for the diagonal elements of the density matrix are:

\[
\frac{d \rho_{00}}{dt} = \Gamma_1 \rho_{11} + \Gamma_3' \rho_{33} 
\]

(2.26)

\[
\frac{d \rho_{11}}{dt} = -i g_{12} (\rho_{21} - \rho_{12}) - \Gamma_1 \rho_{11} + \Gamma_2 \rho_{22} 
\]

(2.27)
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\[ \frac{d\rho_{22}}{dt} = -i g_{23}(\rho_{32} - \rho_{23}) - i g_{12}(\rho_{12} - \rho_{21}) - \Gamma_2 \rho_{22} + \Gamma_3 \rho_{33}. \]  
\[ \frac{d\rho_{33}}{dt} = -i g_{23}(\rho_{23} - \rho_{32}) - (\Gamma_3 + \Gamma'_3) \rho_{33}. \]  

(2.28)

(2.29)

The off-diagonal elements for such a system are:

\[ \frac{d\rho_{12}}{dt} = [i \Delta_1 - \gamma_{12}] \rho_{12} - i g_{12}(\rho_{22} - \rho_{11}) + i g_{23} \rho_{13} \]  
\[ \frac{d\rho_{13}}{dt} = [i \gamma_{31} - \gamma_{13}] \rho_{13} - i g_{32} \rho_{12} - i g_{21} \rho_{23} \]  
\[ \frac{d\rho_{23}}{dt} = [i \Delta_2 - \gamma_{23}] \rho_{23} - i g_{23}(\rho_{33} - \rho_{22}) - i g_{12} \rho_{13} \]  
\[ \frac{d\rho_{01}}{dt} = [-i(\omega'_2 - \omega'_1) - \gamma_{01}] \rho_{01} + i g_{12} \rho_{01} \]  
\[ \frac{d\rho_{02}}{dt} = [i \delta_{20} - \gamma_{02}] \rho_{02} + i g_{23} \rho_{03} + i g_{12} \rho_{01} \]  
\[ \frac{d\rho_{03}}{dt} = [i \delta_{30} - \gamma_{03}] \rho_{03} + i g_{23} \rho_{02}. \]  

(2.30)

(2.31)

(2.32)

(2.33)

(2.34)

(2.35)

Eqs. (2.26)-(2.35) can be written in a linear matrix form as follows:

\[ \frac{d\Phi}{dt} = L\Phi + K. \]  

(2.36)

Here the elements of \( \Phi \), a 16-element vector, are:

\[ \Phi_1 = \rho_{22} \quad \Phi_2 = \rho_{23} \quad \Phi_3 = \rho_{13} \]  
(2.37)

\[ \Phi_4 = \rho_{32} \quad \Phi_5 = \rho_{32} \quad \Phi_6 = \rho_{21} \]  
(2.38)

\[ \Phi_7 = \rho_{11} \quad \Phi_8 = \rho_{12} \quad \Phi_9 = \rho_{00} \]  
(2.39)

\[ \Phi_{10} = \rho_{01} \quad \Phi_{11} = \rho_{03} \quad \Phi_{12} = \rho_{10} \]  
(2.40)

\[ \Phi_{13} = \rho_{02} \quad \Phi_{14} = \rho_{30} \quad \Phi_{15} = \rho_{20}; \]  
(2.41)

\[ \Phi_{16} = \rho_{33} \]  
(2.42)

The 16 \times 16 matrix \( L \) and the 16-element vector \( K \) contain the coefficients of the Eqs. (2.26)-(2.35).
2.4 Linear absorption and emission spectra

In this section we show how the general expressions for the emission and absorption spectra of the $\Xi_-$, $K$ and $\Pi$ systems can be calculated using the linear response theory. Based on this theory the emission spectra of these systems can be obtained from \[32, 42\]

\[
S_{\Xi_-}(\omega_r, t') = \text{Re} \Gamma_{1\Xi}^{-}(z, t')|_{z=i\omega_r} \tag{2.43}
\]

\[
S_{K}(\omega_r, t') = \text{Re} \Gamma_{1K}(z, t')|_{z=i\omega_r}, \tag{2.44}
\]

\[
S_{\Pi}(\omega_r, t') = \text{Re} \Gamma_{1\Pi}(z, t')|_{z=i\omega_r}. \tag{2.45}
\]

Here $z$ is the Laplace conjugate of the time of observation $t$, and $t'$ refers to the initial time ($t > t'$). $\omega_r$ refers to the emission frequencies and $\Gamma_{1\Xi}^{-}(z, t')$, $\Gamma_{1K}(z, t')$ and $\Gamma_{1\Pi}(z, t')$ are given as follows:

\[
\Gamma_{1\Xi}^{-}(z, t') = \langle \hat{p}_{\Xi_-}(z, t')p_{\Xi_-}^+(t') \rangle = \int_{0}^{\infty} \langle \hat{p}_{\Xi_-}(t' + \tau)p_{\Xi_-}^+(t') \rangle e^{-z\tau} d\tau, \tag{2.46}
\]

\[
\Gamma_{1K}(z, t') = \langle \hat{p}_K(z, t')p_{K}^+(t') \rangle = \int_{0}^{\infty} \langle \hat{p}_K(t' + \tau)p_{K}^+(t') \rangle e^{-z\tau} d\tau, \tag{2.47}
\]

\[
\Gamma_{1\Pi}(z, t') = \langle \hat{p}_{\Pi}(z, t')p_{\Pi}^+(t') \rangle = \int_{0}^{\infty} \langle \hat{p}_{\Pi}(t' + \tau)p_{\Pi}^+(t') \rangle e^{-z\tau} d\tau. \tag{2.48}
\]

Here $\tau = t - t' > 0$, $p^+ = [p^-]^*$ in each of these systems, and

\[
p_{\Xi_-}^+ = \mu_{01}a_1^+a_1, \quad p_{K}^+ = \mu_{02}a_2^+a_2, \quad p_{\Pi}^+ = \mu_{03}a_3^+a_3. \tag{2.49}
\]

$\mu_{01}$, $\mu_{02}$, and $\mu_{03}$ in Eq. (2.49) are the dipole moments associated with the 0-1, 0-2, and 0-3 transitions. Note that when parities of the states are well-defined in a system only one or two of these transitions can occur. In asymmetric QW's, however, since parities are not well-defined all of these transitions can co-exist. This will be discussed in the following chapters in detail. Note that in order to keep linear response theory valid, one should assume that the probe field (in the case of absorption) and the pump field (in the case of emission) are weak. As will be clarified in the following chapters, this is scaled by
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the number of carriers generated in the excited states and by the various damping rates involved in the optical processes.

To calculate the linear absorption spectrum of each of these systems, based on the linear response theory we calculate [32]

\[
A(z, t') = \text{Re} [r_{2}(z, t') - r_{1}(z, t')]_{z=\omega_p},
\]

(2.50)

\[
A^{K}(z, t') = \text{Re} [\Gamma^{K}^{2}(z, t') - \Gamma^{K}^{1}(z, t')]_{z=\omega_p},
\]

(2.51)

\[
A^{\Pi}(z, t') = \text{Re} [\Gamma^{\Pi}^{2}(z, t') - \Gamma^{\Pi}^{1}(z, t')]_{z=\omega_p},
\]

(2.52)

for the \(\Xi\), \(K\), and \(\Pi\) systems, respectively. Here \(\omega_p\) is the frequency of the probe field and \(\Gamma^{\Xi}^{2}\), \(\Gamma^{K}^{2}\), and \(\Gamma^{\Pi}^{2}\) are given by:

\[
\Gamma^{\Xi}^{2}(z, t') = \langle p_{\Xi}^{\pm}(t') \rho_{\Xi}^{\pm}(z, t') \rangle,
\]

(2.53)

\[
\Gamma^{K}^{2}(z, t') = \langle p_{K}^{\pm}(t') \rho_{K}^{\pm}(z, t') \rangle,
\]

(2.54)

\[
\Gamma^{\Pi}^{2}(z, t') = \langle p_{\Pi}^{\pm}(t') \rho_{\Pi}^{\pm}(z, t') \rangle.
\]

(2.55)

To calculate \(\Gamma_{1}\)'s, and \(\Gamma_{2}\)'s we first use the density operator to obtain the following averages

\[
\langle p_{\Xi}^{-}(t) \rangle = \text{Tr} [\rho(t) p_{\Xi}^{-}] = \mu_{01} \Phi_{10}(t),
\]

(2.56)

\[
\langle p_{K}^{-}(t) \rangle = \text{Tr} [\rho(t) p_{K}^{-}] = \mu_{02} \Phi_{13}(t) e^{i\omega_1 t},
\]

(2.57)

\[
\langle p_{\Pi}^{-}(t) \rangle = \text{Tr} [\rho(t) p_{\Pi}^{-}] = \mu_{03} \Phi_{11}(t) e^{i(\omega_1 + \omega_2) t}.
\]

(2.58)

We then apply the quantum regression theorem to calculate the two-time correlations \((\Gamma_{1}\)'s and \(\Gamma_{2}\)'s). This theorem describes the motion of a system operator, \(M\), in terms of a set of system operators, \(\{M_{\mu}\}\). Here \(M\) could be one member of the set \(\{M_{\mu}\}\) or a linear combination of them [43]. Based on this theorem, if the time evolution of the average of this operator can be written as:

\[
\langle M(t) \rangle = \sum_{\mu} O_{\mu}(t, t') \langle M_{\mu}(t') \rangle + \lambda_{\mu}.
\]

(2.59)
and if the numerical coefficients $O_{\mu}(t, t')$ are known for a single set of $t$ and $t'$ ($t > t'$), then the mean of a two-time operator can be obtained by:

$$< M(t)N(t') > = \sum_{\mu} O_{\mu}(t, t') < M_{\mu}(t')N(t') > + \lambda_{\mu} < N(t') >.$$  \hspace{1cm} (2.60)

Here $N$ can be any system operator and $\lambda_{\mu}$ is a number.

To apply this theorem we consider the Laplace transform of Eqs. (2.56)-(2.58) with respect to $\tau = t - t' > 0$:

$$< P_{\Xi_{-}}(z, t') >= i\mu_{01}\Phi_{10}(z, t'),$$  \hspace{1cm} (2.61)

$$< \hat{p}_{K}(z, t) >= i\mu_{02}\Phi_{13}(z_{1}, t'),$$  \hspace{1cm} (2.62)

$$< \hat{p}_{\Pi}(z, t') >= i\mu_{03}\Phi_{11}(z_{2}, t'),$$  \hspace{1cm} (2.63)

where $z_{1} = z - i\omega_{1}$ and $z_{2} = z - i(\omega_{1} + \omega_{2})$.

Expanding $\Phi_{10}(z, t')$, $\Phi_{13}(z_{1}, t')$ and $\Phi_{11}(z_{2}, t')$ in terms of the set of averages defined by Eqs. (2.37)-(2.42) results in a form for Eqs. (2.61)-(2.63) suitable for application of the quantum regression theorem. To see this let us find the Laplace transform of Eq. (2.36):

$$\Phi(z, t') = R(z)\Phi(t') + \frac{1}{z}R(z)K,$$  \hspace{1cm} (2.64)

where

$$R(z) = (zI - L)^{-1}.$$  \hspace{1cm} (2.65)

Here $I$ refers to the identity matrix. Eqs. (2.64) and (2.65) are written for the $\Xi_{-}$ system (Eq. (2.61)). The same functional forms are also valid for the $K$ and $\Pi$ systems (Eqs. (2.62)-(2.63)), if we substitute $z$ by $z_{1}$ and $z_{2}$, respectively. By substitution of Eq. (2.64) in Eq. (2.61) and corresponding ones for the $K$ and $\Pi$ systems in Eqs. (2.62) and (2.63), we have

$$< \hat{p}_{\Xi_{-}}(z, t') >= \mu_{01}\sum_{j=1}^{16}\{R_{10,j}(z)\Phi_{j}(t') + \frac{1}{z}R_{10,j}(z)K_{j}\}.$$  \hspace{1cm} (2.66)
\begin{align}
\langle \hat{p}_K(z, t') \rangle &= \mu_{02} \sum_{j=1}^{16} \left\{ R_{13,j}(z_1) \Phi_j(t') + \frac{1}{z_1} R_{13,j}(z_1) K_j \right\} \\
\langle \hat{p}_n(z, t') \rangle &= \mu_{03} \sum_{j=1}^{16} \left\{ R_{11,j}(z_2) \Phi_j(t') + \frac{1}{z_2} R_{11,j}(z_2) K_j \right\}.
\end{align}

These equations have the same functional forms as that of Eq. (2.59). Here \( \Phi_j(t') \)'s act as \( \langle M_\mu(t') \rangle \), and \( R_{i,j}'s \) and \( K_j's \) which are the elements of the matrix \( \mathbf{R} \) and the column \( \mathbf{K} \), correspond to \( O_\mu(t, t') \) and \( \lambda_\mu \), respectively. To implement the quantum regression theorem we should replace \( \Phi_j's \) and \( K_j's \) in the right hand sides of Eqs. (2.66)-(2.68) with specific two-time correlation functions, similar to the step between Eq. (2.59) and (2.60). For example, to obtain \( \langle \hat{p}_\Xi(z, t') \hat{p}_\Xi(t') \rangle \), based on this theorem, we should make the following substitutions for the \( \Phi_j's \) in Eq. (2.66):

\begin{equation}
\Phi_9(t') = \langle |2 \rangle \langle 2 | \rangle \rightarrow \langle |2 \rangle \langle 2 | p_{\Xi}^+(t') \rangle = \mu_{01} p_{10} = \mu_{01} \Phi_{12}(t')
\end{equation}

\begin{equation}
\Phi_{10}(t') = \langle |1 \rangle \langle 0 | \rangle \rightarrow \langle |1 \rangle \langle 0 | p_{\Xi}^+(t') \rangle = \mu_{01} p_{11} = \mu_{01} \Phi_7(t')
\end{equation}

\begin{equation}
\Phi_{11}(t') = e^{-(\omega_1 + \omega_2)t'} \langle |3 \rangle \langle 0 | \rangle \rightarrow e^{-(\omega_1 + \omega_2)t'} \langle |3 \rangle \langle 0 | p_{\Xi}^+(t') \rangle = \mu_{01} \Phi_3(t'),
\end{equation}

\begin{equation}
\Phi_{13}(t') = e^{-i\omega_1 t'} \langle |2 \rangle \langle 0 | \rangle \rightarrow e^{-i\omega_1 t'} \langle |2 \rangle \langle 0 | p_{\Xi}^+(t') \rangle = \mu_{01} p_{12} = \mu_{01} \Phi_5(t').
\end{equation}

The rest of the \( \Phi_j's \) should be replaced by zero. Here \( p_{\Xi}^+(t') \) plays the role of \( N(t') \) in Eq. (2.60). Also \( K_j \) should be replaced with \( K_j \langle p_{\Xi}^+(t') \rangle \), as required by the quantum regression theorem.

To calculate \( \langle \hat{p}_K(z, t') p_K^+(t') \rangle \) we make the following substitutions in Eq.(2.67):

\begin{equation}
\Phi_9(t') \rightarrow \mu_{02} e^{-i\omega_1 t'} \Phi_{15}(t'),
\end{equation}

\begin{equation}
\Phi_{10}(t') \rightarrow \mu_{02} e^{-i\omega_1 t'} \Phi_6(t'),
\end{equation}

\begin{equation}
\Phi_{11}(t') \rightarrow \mu_{02} e^{-i\omega_1 t'} \Phi_2(t'),
\end{equation}

\begin{equation}
\Phi_{13}(t') \rightarrow \mu_{02} e^{-i\omega_1 t'} \Phi_1(t').
\end{equation}
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For the calculation of $\langle \hat{n}_n(z,t') \hat{n}_n^\dagger(t') \rangle$ the substitutions in Eq.(2.68) are found as follows

$$\Phi_9(t') \rightarrow \mu_{03} e^{-i(\omega_1+\omega_2)t'} \Phi_{14}(t'),$$

$$\Phi_{10}(t') \rightarrow \mu_{03} e^{-i(\omega_1+\omega_2)t'} \Phi_5(t'),$$

$$\Phi_{11}(t') \rightarrow \mu_{03} e^{-i(\omega_1+\omega_2)t'} \Phi_{16}(t'),$$

$$\Phi_{13}(t') \rightarrow \mu_{03} e^{-i(\omega_1+\omega_2)t'} \Phi_4(t').$$

For these two correlations $K_j$'s should be substituted by $K_j = \langle p^+_K(t') \rangle$ and $K_j = \langle p^-_n(t') \rangle$, respectively.

Since we assume the optical field amplitude variations are quite slow compared to the characteristic dephasing rates in the systems, we will proceed with the calculations in the steady state conditions, i.e. $t' \rightarrow \infty$ ($t \rightarrow \infty$). Under these conditions Eq. (2.36) takes the following form:

$$\Phi_i(\infty) = -\sum_j (L^{-1})_{ij} K_j.$$  \hspace{1cm} (2.81)

Imposing the steady state conditions the following general expressions for the emission spectra of the $\Xi_-, K, \Pi$ systems are obtained:

$$S_{\Xi_-}(\omega_r) = \mu_{01}^2 \text{Re} \{ R_{10,9}(z) \Phi_{12}(\infty) + R_{10,10}(z) \Phi_7(\infty) + R_{10,11}(z) \Phi_3(\infty) + R_{10,13}(z) \Phi_8(\infty) \}_{z=i\omega_r},$$

$$S_K(\omega_r) = \mu_{02}^2 \text{Re} \{ R_{13,9}(z_1) \Phi_{15}(\infty) + R_{13,10}(z_1) \Phi_6(\infty) + R_{13,11}(z_1) \Phi_2(\infty) + R_{13,13}(z_1) \Phi_1(\infty) \}_{z=i\omega_r},$$

$$S_{\Pi}(\omega_r) = \mu_{03}^2 \text{Re} \{ R_{11,9}(z_2) \Phi_{14}(\infty) + R_{11,10}(z_2) \Phi_5(\infty) + R_{11,11}(z_2) \Phi_{16}(\infty) + R_{11,13}(z_2) \Phi_4(\infty) \}_{z=i\omega_r}.$$
Note that in Eqs. (2.82)-(2.84) we dropped the terms containing $K_j$. This is because these terms refer to the coherent Rayleigh emission. In absence of a coupling field near resonance with the transition of the probe field this process does not occur.

To calculate the absorption spectra one should find $\Gamma_2$'s. This is done by taking steps similar to those in the case of the $\Gamma_1$'s. To apply the quantum regression theorem, here the operators $N(t')$ should be inserted in the left sides of $< M_{\mu}(t') >$. We found that in order to calculate $\Gamma_2^{\pm}$ all $\Phi_j$ in Eq. (2.66) should be replaced by zero except for:

$$
\Phi_5(t') \rightarrow \mu_{01} \Phi_{14}(t'), \\
\Phi_6(t') \rightarrow \mu_{01} \Phi_{15}(t'), \\
\Phi_7(t') \rightarrow \mu_{01} \Phi_{12}(t'), \\
\Phi_{10}(t') \rightarrow \mu_{01} \Phi_{9}(t'). 
$$

For the case of $\Gamma_2^K$ the non-zero substitutions for Eq. (2.67) are

$$
\Phi_4(t') \rightarrow \mu_{02} e^{(-i\omega_1 t') \Phi_{14}(t')}, \\
\Phi_8(t') \rightarrow \mu_{02} e^{(-i\omega_1 t') \Phi_{12}(t')}, \\
\Phi_{13}(t') \rightarrow \mu_{02} e^{(-i\omega_1 t') \Phi_{9}(t')}.
$$

and for $\Gamma_2^\Pi$ the substitutions for Eq.(2.68) are found as follows

$$
\Phi_2(t') \rightarrow \mu_{03} e^{-i(\omega_1+\omega_2) t'} \Phi_{15}(t'), \\
\Phi_3(t') \rightarrow \mu_{03} e^{-i(\omega_1+\omega_2) t'} \Phi_{12}(t'), \\
\Phi_{11}(t') \rightarrow \mu_{03} e^{-i(\omega_1+\omega_2) t'} \Phi_{9}(t'), \\
\Phi_{16}(t') \rightarrow \mu_{03} e^{-i(\omega_1+\omega_2) t'} \Phi_{14}(t').
$$

Here, similar to the previous cases, $K_j$'s should be replaced with the corresponding terms already found in the case of the emission spectra.
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The results for the absorption spectra of these systems are found as follows:

\[ A^\Xi_-(\omega_p) = \mu_{01}^2 \text{Re}\{R_{10,5}(z)\Phi_{14}(\infty) + R_{10,6}(z)\Phi_{15}(\infty) \]  
\[ + R_{10,7}(z)\Phi_{12}(\infty) + R_{10,10}(z)\Phi_9(\infty)\} \big|_{z=i\omega_p} - S^\Xi_-(\omega_p), \]  
(2.96)

\[ A^K(\omega_p) = \mu_{02}^2 \text{Re}\{R_{13,4}(z_1)\Phi_{14}(\infty) + R_{13,8}(z_1)\Phi_{12}(\infty) \]  
\[ + R_{13,13}(z_1)\Phi_9(\infty)\} \big|_{z=i\omega_p} - S_K(\omega_p), \]  
(2.97)

\[ A^\Pi(\omega_p) = \mu_{03}^2 \text{Re}\{R_{11,2}(z_2)\Phi_{15}(\infty) + R_{11,3}(z_2)\Phi_{12}(\infty) \]  
\[ + R_{11,11}(z_2)\Phi_9(\infty) + R_{11,16}(z_2)\Phi_{14}(\infty)\} \big|_{z=i\omega_p} - S^\Pi(\omega_p). \]  
(2.98)

In chapters 3 and 4 by applying the specific features of atomic systems to Eqs. (2.82)-(2.84) and Eqs. (2.96)-(2.98) we study various coherent effects of atoms. For the case of QW's, however, as will be shown in the following chapters this require further extensive calculations to properly include the incoherent effects of the carrier-carrier scattering.

2.5 Emission and absorption spectra of systems with \( \Xi_d \) and \( \Lambda_d \) configurations

Up to now we considered systems where the ground levels \(|0>\) were left unchanged by the intense fields. For this reason the nonlinear effects associated with these systems were not accompanied by the real excitation of electrons. A large number of interesting nonlinear effects, however, occur in the presence of real excitations, when the ground level is affected by an intense field (Fig.2.4). The developments of the \( \Xi_d \) and \( \Lambda_d \) systems are very similar to that presented in the previous sections. As an example, for an atomic system with a \( \Xi_d \) configuration the equations of motion are:
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\[
\frac{d\rho_{00}}{dt} = -i\gamma_{21}(\rho_{10} - \rho_{01}) + \Gamma_1\rho_{11} \quad (2.99)
\]
\[
\frac{d\rho_{11}}{dt} = -i\gamma_{12}(\rho_{21} - \rho_{12}) - i\gamma_{01}(\rho_{10} - \rho_{01}) - \Gamma_1\rho_{11} + \Gamma_2\rho_{22} \quad (2.100)
\]
\[
\frac{d\rho_{22}}{dt} = -i\gamma_{12}(\rho_{12} - \rho_{11}) - \Gamma_2\rho_{22} \quad (2.101)
\]
\[
\frac{d\rho_{10}}{dt} = -(i\Delta_1 + \gamma_{10})\rho_{10} - i\gamma_{12}\rho_{20} - i\gamma_{01}(\rho_{00} - \rho_{11}) \quad (2.102)
\]
\[
\frac{d\rho_{20}}{dt} = -(i\Delta_2 + \gamma_{20})\rho_{20} - i\gamma_{12}\rho_{10} + i\gamma_{01}\rho_{21} \quad (2.103)
\]
\[
\frac{d\rho_{12}}{dt} = (i\Delta_2 - \gamma_{12})\rho_{12} - i\gamma_{12}(\rho_{22} - \rho_{11}) - i\gamma_{01}\rho_{02} \quad (2.104)
\]

Eqs. (2.99)-(2.104) can be written in a matrix form as Eq. (2.36). Here, however, \( L \) is a 9 × 9 matrix and \( K \) a 9-element vector.

The results for \( \Gamma_1^{\Xi_d} \) and \( \Gamma_2^{\Xi_d} \) associated with the 0-1 transition are found as follows

\[
\Gamma_1^{\Xi_d}(z, t') = \mu_0^2 \left\{ R_{8,3}(z')\Phi_2(\infty) + R_{8,7}(z')\Phi_6(\infty) - R_{8,8}(z)\Phi_1(\infty) \right\}, \quad (2.105)
\]
\[
\Gamma_2^{\Xi_d}(z, t') = \mu_0^2 \left\{ R_{8,7}(z')\Phi_6(\infty) + R_{8,8}(z')\Phi_7(\infty) + R_{8,4}(z)\Phi_5(\infty) \right\}, \quad (2.106)
\]

where \( z' = z - i\omega_1 \). Having these two correlations, similar to the previous configuration, one can use the following equations to find the emission and absorption spectra of a \( \Xi_d \) system

\[
S_{\Xi_d}(\omega, t) = \text{Re}\left[\Gamma_{1}^{\Xi_d}(z, t)\right]_{z=i\omega}, \quad (2.107)
\]
\[
A_{\Xi_d}(\omega, t) = \text{Re}\left[\Gamma_{2}^{\Xi_d}(z, t) - \Gamma_{1}^{\Xi_d}(z, t)\right]_{z=i\omega}. \quad (2.108)
\]

One can easily apply this formalism to the \( \Lambda_d \) system after making proper changes.
Chapter 3

Coherent nonlinear effects in atomic systems

3.1 Introduction

Laser induced transparency (LIT) and dark line effects occur when the upper level of a transition is coupled to a metastable state forming a $\Lambda$ (2.2b) (or $\Xi$ (2.2a)) configuration. Here, depending on the coupling field intensity, the absorption or emission associated with this transition decreases or vanishes at a specific frequency, respectively. These effects are the foundations of several other interesting phenomena in atoms, such as lasing without inversion [46] and generation of transparent media with large indices of refraction [47].

In this chapter we briefly review LIT and dark line effects and underline a new picture to describe these phenomena. Based on this picture near resonance interaction of lasers with a system can be considered as a structuring process, similar to laser-induced continuum structure [46]. In contrast to the commonly used dressed state picture, this alternative picture can describe the dynamics of the absorption (LIT) and emission (dark line effect) spectra of the system when the intensity of the coupling field is low. In this chapter we present a preliminary introduction to these two pictures in simple systems. The details and utilizations of the alternative picture are presented in the following chapters. To review LIT and dark line effects in the $\Xi$ and $\Lambda$ systems in this chapter we apply the formalism developed in chapter 2 as the fields with frequencies $\omega_2$ in Figs.2.3a and 2.3b are turned off. Under these conditions our formalism becomes similar to that in Ref. [51].
3.2 Review of Laser-induced transparency in an atomic system with a Λ configuration

Elimination of the absorption (LIT) and emission spectra (dark line effect) of atoms with Λ configurations (Fig.2.2b) have been studied extensively in the past [34, 51]. LIT is the mechanism behind some other interesting nonlinear optical processes in atoms and quantum wells. For example, it can lead to a medium with sharp changes in its index of refraction. This has potential applications in photonic materials [37]. It is also the mechanism behind fast semiconductor switches [11, 19]. In this section we briefly review LIT. For convenience here we normalize the detuning of the field, Δ₁, and its Rabi frequency, g₁₂, to the width of the upper transition level, Γ₂ (Δ₁ = Δ₁/Γ₂, g₁₂ = g₁₂/Γ₂). To study the emission spectra of such a system a pump field can be considered to weakly populate the upper transition level (|2⟩). Under similar coupling conditions, however, these spectra have the same dynamics as those of absorption. For this reason here we avoid presenting the results of the dark line effect.

The results under the resonant coupling condition (Δ₁ = 0) are shown in Fig.3.1 for various field intensities. In the absence of the optical field (g₁₂=0) we have a Lorentzian spectrum (solid line). For g₁₂ = ½ the spectrum develops a dip or “transparency hole” at the line-center frequency (dashed line). When g₁₂ = 5 the transparency hole becomes wider (dotted line). Note that the peak amplitudes associated with LIT are equal to that of the uncoupled peak but are half as wide.

In the case of detuned coupling (Δ₁ = 1) as Fig.3.2 shows the effect is generation of an asymmetric LIT. As the field intensity increases the spectrum becomes more symmetric (dotted line), and at the high-field limit (g₁₂ >> 1) it becomes symmetric. The “transparency hole” here is generated at −Δ₁.

A typical example of a Λ system can be found in the transitions of ⁸⁷Rb atoms (Fig.3.3). Here when the 5P₁/₂ upper state is coupled to 5S₁/₂ with hyper fine (total angular momentum of the atom) F=2, the detection of the former from 5S₁/₂ with F=1
Figure 3.1: Absorption spectrum corresponding to the 0-2 transition in a resonant $\Lambda$ system. The solid line corresponds to $\hat{g}_{12} = 0$, dashed line to $\hat{g}_{12} = 1/2$, and dotted line to $\hat{g}_{12} = 5$.

Figure 3.2: Absorption spectrum corresponding to the 0-2 transition in a detuned $\Lambda$ system with $\hat{\Delta} = 1$. The solid line corresponds to $\hat{g}_{12} = 0$, dashed line to $\hat{g}_{12} = 1/2$, and dotted line to $\hat{g}_{12} = 5.5$. 
represents a well-defined LIT. Note that the natural line-width of $5P_{1/2}$ is 6 MHz and that of $5S_{1/2}$ with hyper fine $F=2$ is 0.01 MHz [40]. An experiment based on this configuration has been reported in detail in Ref. [49].

3.3 Structuring description of LIT and dark line effects in a $\Lambda$ system

In the preceding section we reviewed LIT dynamics at the medium and high field intensities. At these intensities one can apply a dressed state picture to explain how the absorption spectrum becomes doubled [52]. As discussed by Cohen-Tannoudji et al [50], based on this picture the atom-field states $|2, n\omega >$ and $|1, (n + 1)\omega >$ ($n$ is the photon number of the field with frequency $\omega$) are degenerate in the absence of atom-field interaction. When this interaction is included the degeneracy is removed and two mixed states are generated (see Fig.3.4). Under the resonant coupling condition these states are

$$|d >_+ = \frac{1}{2} [|1, (n + 1)\omega > + |2, n\omega >], \quad (3.1)$$

$$|d >_- = \frac{1}{2} [|1, (n + 1)\omega > - |2, n\omega >]. \quad (3.2)$$

The energies of these states from the line-center of $|2 >$ are $\pm g_{12}$. The dressed state picture described above is valid as long as the field intensity is such that $\tilde{g}_{12} > 1$. In other words, one does not include damping of the system. At the
Figure 3.4: Schematic diagram of the dressed system picture of a Λ system. \(|d>_\pm\) refer the dressed state doublet.

low-field limit \((\hat{g}_{12} \ll 1)\), however, this picture is not valid. This is because in contrast to the high-field limit \((\hat{g}_{12} \gg 1)\), here the widths of the coupled states play a role in the dynamics of the system. Therefore, to treat the system dynamics at the low-field limit we have to use a different treatment. To do this we borrow the laser induced continuum structure (LICS) concept [48]. LICS is the radiative analogy of the autoionized states [53]. Here a bound state of an atom is optically embedded into the continuum using a laser field. Since for a flat continuum the interaction is symmetric, the result of detection of the continuum from the ground state is a symmetric hole driven into the continuum [52]. However, if the interaction becomes asymmetric due to the finite width of the continuum, for example, the structure in the continuum would become asymmetric. These structures are often called Fano profiles [52, 53].

To describe LIT and dark line effects as structuring processes, similar to that in LICS, we consider the upper transition level in the Λ system as an “effective continuum”. This “continuum” is structured by the optical embedding of the metastable state into its line center vicinity (Fig.3.5). To illustrate this we study how LIT evolves at low field intensities in a small region around the uncoupled absorption peak line-center. As Fig.3.6 shows, under the resonant coupling condition the effect of the coupling process is to drive a Lorentzian “transparency hole” at the peak of the uncoupled absorption spectrum. The
Figure 3.5: Schematic diagram of the structuring process in a $\Lambda$ system. The shaded area represents the "effective continuum" in this system.

more intense the coupling field, the deeper the hole. This process is the same as that seen in LICS under the symmetric coupling conditions [52]. In fact, in the case of resonant coupling in the $\Lambda$ system, the embedded metastable state also sees the upper transition level symmetrically.

To see the dynamics of the system at the low-field limit under detuned coupling condition, we show in Fig.3.7 the detailed evolution of the asymmetric (left) peak in Fig.3.2 at its early development. Here the asymmetric structure is narrow and small both in depth and height at low field intensities. As the field intensity increases the structure becomes larger until it nearly reaches zero at $-\Delta$. At this stage its amplitude also becomes the same as that of the uncoupled peak. These dynamics are very similar to the Fano profiles in LICS under asymmetric coupling conditions. In fact in the case of Fig.3.7 the embedded metastable state sees the "effective continuum" asymmetrically. The ability of the structuring picture to explain other physical phenomena are discussed in the following chapters.
Chapter 3. Coherent nonlinear effects in atomic systems

Figure 3.6: Absorption spectrum of the 0-2 transition in a resonant Λ system at the early stages of the coupling process. The solid line corresponds to $\tilde{g}_{12} = 0$, the dashed line to $\tilde{g}_{12} = 0.017$, the dotted line to $\tilde{g}_{12} = 0.033$, and the dashed-dotted line to $\tilde{g}_{12} = 0.066$.

Figure 3.7: Absorption spectrum of the 0-2 transition in a detuned Λ system ($\tilde{\Delta}_1 = 1$) at the early stages of the coupling process. The solid line corresponds to $\tilde{g}_{12} = 0$, the dashed line to $\tilde{g}_{12} = 0.033$, the dotted line to $\tilde{g}_{12} = 0.067$, and the dashed-dotted line to $\tilde{g}_{12} = 0.133$. 
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3.4 Quantum interference effects

Since Young's double-slit experiment, quantum interferences have been found responsible for many exotic phenomena in physics. In recent years, in particular, they have opened fruitful new areas of research in nonlinear optics. It has been shown, for example, that they can decrease the spontaneous lifetime of an atom [39] or can be used to coherently control the intersubband transitions in QW's [9]. Designing or controlling the optical properties of gases and solids can revolutionize the realizations of nonlinear optics and their applications. In this introductory subsection we briefly discuss quantum interferences in a simple system. Later, in the following chapters, they are extended to more complicated cases.

The structuring picture presented in the preceding section has the advantage that it can naturally reveal the mechanism behind LIT and dark line effects in a Λ system. In fact in the case of LICS the Fano profiles are considered to be a direct indication of quantum interferences [52, 53]. Here they are caused by the transition between the ground state to the continuum (direct path) and by that between the ground and the bound state through the continuum (indirect or Raman path). Destructive interference between these two paths leads to the "Fano window" where the ground to continuum transition vanishes [52, 53]. Similarly, we can describe LIT and dark line effects in terms of quantum interferences. For a Λ system, for example, the interfering paths are a direct one between |0⟩ and |2⟩, and a Raman one between |0⟩ and |1⟩ through |2⟩ (see Fig.3.8).

Note that this picture only illustrates the types of interfering paths. When we detect |2⟩ from |0⟩, we in fact monitor |F⟩, the eigen-state of the full Hamiltonian. |F⟩ is located at the vicinity of the |2⟩ line-center at low field intensities. Based on the analogy between the structuring picture discussed here and LICS [48], this state is a linear superposition of the "effective continuum"(|2⟩) and a dressed state. This dressed state is the result of mixing of |1⟩ with some part of the "effective continuum". The
transition from $|0\rangle$ to $|F\rangle$ (or $|2\rangle$, if we do not change our notation), therefore contains a transition moment between $|0\rangle$ and $|2\rangle$ (direct) and a two-photon moment between $|0\rangle$ and $|1\rangle$ (indirect).
Chapter 4

Multi-photon coherent effects in atomic systems

4.1 Introduction

The coherent effects discussed in the preceding chapter were caused by probing an atomic level optically coupled to another level (a metastable state). The characteristic features of these effects were attributed to the one-photon coupling of these levels and to the quantum interferences between the direct and two-photon paths. In this chapter we show how one can explore new and interesting phenomena by introducing multi-photon coupling processes and extended quantum interferences. This includes dynamic narrowing of the spontaneous emission spectrum of an atomic transition by inducing a constructive interfering effect, LIT and dark line effects with tunable frequencies, control of the coupling mechanisms, etc. Here we study systems with $\Xi_-, K, \text{ and } \Pi$ configurations (Fig.2.3). As will be found out in the following chapters, the results of this chapter [20]- [22], serve as foundations for the development and understanding of the new nonlinear effects in QW's.

4.2 Dynamic dark line and laser-induced transparency effects in systems with $\Pi$ configurations

We start our investigation considering LIT and dark line effects in an atomic system with a $\Pi$ configuration. This is done by applying the formalism developed in chapter 2 (Eqs. (2.84) and (2.98)). Here we find the dynamics of the absorption spectrum similar to those of the corresponding emission spectra under the same coupling conditions. Therefore, we do not present the absorption spectrum results. Here we also normalize the Rabi
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Figure 4.1: Resonance fluorescence spectra (in arbitrary units) corresponding to the 3-0 transition of a resonant II system with (a) $\hat{g} = 1$ and (b) $\hat{g} = 3$. The thin solid line corresponds to $\hat{g} = 0$. $\omega_r$ and $\omega_{03}$ are the frequencies of the emitted light and that corresponding to the energy separation between the 0-3 transition, respectively.

Figure 4.1: Resonance fluorescence spectra (in arbitrary units) corresponding to the 3-0 transition of a resonant II system with (a) $\hat{g} = 1$ and (b) $\hat{g} = 3$. The thin solid line corresponds to $\hat{g} = 0$. $\omega_r$ and $\omega_{03}$ are the frequencies of the emitted light and that corresponding to the energy separation between the 0-3 transition, respectively.

frequencies and detunings of the fields to the width of $|3 > (\Gamma_3)$, i.e. $\hat{g}_{ij} = \frac{q_{ji}}{\Gamma_3}$ and $\tilde{\Delta}_i = \frac{d_i}{\Gamma_3}$. Note also that $|1 >$ and $|2 >$ are metastable states. The results of the calculation of Eq.(2.84) for various field intensities under the conditions of $\hat{g}_{12} = \hat{g}_{23} = \hat{g}$ and $\tilde{\Delta}_1 = \tilde{\Delta}_2 = 0$ are shown in Fig.4.1. For $\hat{g} = 0$, we have a Lorentzian emission spectrum (thin solid line). However, when $\hat{g}=1$ the emission spectrum becomes dark at two frequencies (dark lines). The centers of these lines are located at +1 and -1 (Fig.4.1a).

A further increase in the field intensities such that $\hat{g} = 3$ causes the dark lines to become broader with frequencies shifted to +3 and -3 of the scaled frequency(Fig.4.1b). Note that in practice the condition of having $\hat{g}_{12} = \hat{g}_{23}$ may be reached by applying two laser fields near resonance with the 1-2 and 2-3 transitions. Also here the emission spectra are obtained by weakly populating the upper transition level using a time-independent incoherent pump rate.

On the other hand, for any value of $\hat{g}$ one can identify generation of three peaks of
emission, one central and two sideband peaks all with the same amplitudes as that of the uncoupled peak. Any increase in the field intensities pushes the sideband peaks away from each other but does not change the central peak frequency. This is accompanied by changing these peaks from having subnatural line-shapes to Lorentzian ones. At the high-field limit all three peaks become Lorentzian. At this limit the width of the central peak, $\Gamma_c$, is $\frac{\Gamma_4}{2}$ and those of the sideband peaks, $\Gamma_s$, are equal to $\frac{\Gamma_4}{4}$. Note here that if the field resonant with the 1-2 transition is turned off ($\hat{g}_{12} = 0$) the central peak disappears, the widths of the sideband peaks increase by a factor two, and their corresponding frequencies change, i.e. the system becomes a $\Lambda$ system (Fig.3.1).

Now let us consider the effects of detuning the coupling fields from their corresponding transitions. We start with the case in which both fields are detuned equally, $\hat{\Delta}_1 = \hat{\Delta}_2 = -1$. As Fig.4.2a shows for $\hat{g} = 1$ the spectrum contains three emission peaks one at +1 of the scaled frequency, and two others at each side of this peak. The peak on the right, however, is very narrow compared to other two peaks. Once we increase the field intensity such that $\hat{g} = 3$ the peak at +1 remains unchanged in frequency but the other two peaks are shifted away from each other. This is accompanied by increasing the width of the peak at the right and narrowing the one on the left side (Fig.4.2b). When the high-field limit ($\hat{g} >> 1$) is reached both sideband peaks become similar. At this limit the effect of detuning is seen to be just a shift of the resonant coupling spectrum (Fig.4.1) by +1.

If we now detune the fields equally but with opposite signs, $\hat{\Delta}_1 = -\hat{\Delta}_2 = +1$, the emission spectrum changes differently. As Fig.4.2c shows for $\hat{g} = 1$ an emission peak is generated at the central emission frequency of the uncoupled peak and two sideband peaks at the sides. For $\hat{g}=3$ the central peak remains unshifted but the sideband peaks are shifted away from each other (Fig.4.2d). Further increase in the field intensities causes the sideband peaks to become similar to each other both in frequency and line-shape. At the high-field limit the spectrum becomes identical to that of resonant coupling. In
Figure 4.2: Resonance fluorescence spectra associated with the 3-0 transition in a detuned \( \Pi \) system. (a) and (b) are the spectra corresponding to \( \Delta_1 = \Delta_2 = -1 \) with \( \hat{g} = 1 \) and \( \hat{g} = 3 \), respectively. (c) and (d) are those with \( \Delta_1 = -\Delta_2 = +1 \) for field intensities the same as those of (a) and (b), respectively. All other specifications are the same as those in Fig.4.1.
other words, the detunings lose their effects. Note that this is a characteristic feature of \( \Delta_1 = -\Delta_2 \) and can happen for any value of the detunings which satisfy this condition.

Up to now we have considered \( \tilde{g}_{12} = \tilde{g}_{23} \). Let us now consider the case in which the two fields remain resonant but they may have different intensities (\( \tilde{g}_{12} \neq \tilde{g}_{23} \)). To show this effect we put \( \tilde{g}_{12} = 1 \) and change \( \tilde{g}_{23} \) such that \( \frac{\tilde{g}_{12}}{\tilde{g}_{23}} \leq 1 \). For \( \frac{\tilde{g}_{12}}{\tilde{g}_{23}} = 1 \) we have a triplet such as that seen in Fig. 4.1a (solid line). However, as Fig. 4.3a shows, when \( \frac{\tilde{g}_{12}}{\tilde{g}_{23}} = \frac{1}{3} \) the central peak is narrowed while the sideband peaks become wider and move away from the uncoupled peak line-center. Ultimately, at the limit of \( \frac{\tilde{g}_{12}}{\tilde{g}_{23}} \ll 1 \), the central peak disappears and the remaining part is just an illustration of the optical coupling of \( |2> \) with \( |3> \). This phenomenon shows a transition from the dark line effect in a system with a \( \Pi \) configuration to that with a \( \Lambda \) configuration.

The inverse case of a similar system is shown in Fig. 4.3b. Here we consider the case where \( \tilde{g}_{12} \) is increased as compared to \( \tilde{g}_{23} \). For \( \frac{\tilde{g}_{12}}{\tilde{g}_{23}} = 3 \) the central peak becomes broader and the sideband peaks narrower. In the limit of \( \tilde{g}_{12} \gg \tilde{g}_{23} \) the sideband peaks have vanished and the central peak becomes identical to that of the uncoupled emission peak of the 3-0 transition. In other words, the strong field resonant with 1-2 transition converts the \( \Pi \) system spectrum to that of natural emission of \( |3> \) to \( |0> \). This means that the field resonant with the 1-2 transition can decouple the two-level system from the strongly driven metastable states. The reason of these phenomena will be discussed in Sect. 4.4.

### 4.3 Two-field structuring processes in a \( \Pi \) system

The results in Sect. 4.2 were concerned with the field intensities such that their corresponding Rabi frequencies were comparable or larger than the largest damping rate of the system. Many characteristic features of LIT and dark line effects, however, are expected to be found in the low-field limit (\( \tilde{g} \ll 1 \)). In this section we describe these features and explain them in terms of the structuring processes. Thus it is an extension of the picture presented in Sect. 3.2. For the reasons which will be clarified in the following, it
Figure 4.3: Evolution of the resonance fluorescence associated with the 3-0 transition in a resonant $\Pi$ system for various ratios of $\frac{g_{11}}{g_{23}}$. (a) represents the emission spectrum when $\frac{g_{11}}{g_{23}} = \frac{1}{3}$ and (b) represents that with $\frac{g_{11}}{g_{23}} = 3$. All other specifications are the same as in Fig. 4.1.
is illustrative to note that a Π system is in fact a Λ system where the metastable state is replaced by a dressed system system consisting of two metastable states.

Let us start with the evolution of the 0-3 transition emission spectrum at the early stage of its development. The results for $\tilde{g}_{23} = \tilde{g}_{12} = \tilde{g}$ and $\tilde{A}_1 = \tilde{A}_2 = 0$ at the vicinity of the uncoupled emission peak frequency are shown in Fig.4.4a. When $\tilde{g}=0.01$ the effect is seen to be the driving of two Lorentzian dips or “emission holes” into the uncoupled emission peak of the 0-3 transition spectrum (solid line). The frequency centers of these dips are separated from that of uncoupled peak emission by $\tilde{g}$. Increasing $\tilde{g}$ to 0.03 pushes the dips to 0.03 and increases their depths and widths by three times (dashed line). In Fig.4.4b and 4.4c we show the cases where the intensities of the fields are different, $\tilde{g}_{23}=0.03$, $\tilde{g}_{12}=0.02$, and $\tilde{g}_{23}=0.02$, $\tilde{g}_{12}=0.03$, respectively. These two figures clearly show that the central frequencies of the emission holes and dark lines are determined by $\tilde{g}_{12}$. However, the strength of coupling of $|3>$ with the dressed two-level system is proportional to $\tilde{g}_{23}$. In Fig.4.4d we show development of an “emission hole” in a Λ system in the low-field limit by turning off the second field ($\tilde{g}_{12}=0$). Here the Lorentzian hole is generated at the center of the uncoupled emission peak, similar to Fig.3.6.

Fig.4.4 suggests that the two optical fields have two distinct roles in the coupling process of a Π system. The field resonant with 2-3 transition is responsible for the strength of the change in the 3-0 transition emission spectrum. In fact, based on our model, this has the role of an “embedding field”.

On the other hand, the field resonant with the 1-2 transition acts as a ”dressing field” which determines the type of the system embedded into $|3>$. It does not have any direct role in the strength of the coupling of $|3>$ with the dressed system. However, since the width of $|3>$ is finite, the coupling process depends on the frequencies where the structuring process is occurring. To see this more clearly and find out the dynamics of the whole emission spectra beyond the low-field limit, we show in Fig.4.5 four different stages of the coupling process of the Π system. Fig.4.5a shows the case where $\tilde{g}_{23}=0.1$ and
Chapter 4. Multi-photon coherent effects in atomic systems

Figure 4.4: Early stages of development of the resonance fluorescence spectra (in arbitrary units) associated with the 3-0 transition in a resonant Π system for various values $\hat{g}_{23}$ and $\hat{g}_{12}$. (a) represents $\hat{g}_{23} = \hat{g}_{12}=0.01$ (solid line) and $\hat{g}_{23} = \hat{g}_{12}=0.03$ (dashed line); (b) and (c) represent the cases where $\hat{g}_{23} = 0.03$ and $\hat{g}_{12}=0.02$; and $\hat{g}_{23} = 0.02$, and $\hat{g}_{12}=0.03$, respectively; and (d) is for the case where $\hat{g}_{12} = 0$ and $\hat{g}_{23}=0.01$ (solid line) or $\hat{g}_{23}=0.03$ (dashed line).
\( \hat{g}_{12} = 0.3 \). Here one can see that the emission holes are actually converted to two dark lines. The frequencies of these lines are determined by \( \hat{g}_{12} \). In other words, the emission spectrum now consists of three extremely subnatural peaks. If we keep \( \hat{g}_{23} \) constant but increase \( \hat{g}_{12} \) to 1.2 we in fact change the embedded system. Here, as Fig.4.5b shows, the tails of the uncoupled emission spectrum are structured. The reason that the effect of the embedding field is now weaker here is that the embedded states are now very off-resonant from the 2-3 transition frequency. In Figs.4.5c and 4.5b we look at the whole emission spectra keeping \( \hat{g}_{12} = 0.6 \) but changing \( \hat{g}_{23} \). Again as Fig.4.5c shows for \( \hat{g}_{23} = 0.1 \) the emission spectrum has two dark lines at +0.6 and -0.6 of the scaled frequency. If we increase \( \hat{g}_{23} \) to 1 central frequencies of the dark lines do not change but they become wider. In other words, the more intense the embedding field, the stronger is the change in \( |3> \).

Putting together these features and the fact that the emission holes are Lorentzian at the low-field limit with depths and widths proportional to \( \hat{g}_{23} \) we come up with the following picture: Since \( |2> \) and \( |1> \) are metastable states and \( |3> \) is a broad one, for any field intensities, the coupling between these two metastable states is much more effective than that between these two states and \( |3> \). Therefore, the two metastable states, while still weakly coupled to \( |3> \), are developed into a well-defined dressed system. Because of this, \( |2> \) and \( |1> \) are each substituted by a doublet of dressed states similar to that shown in Fig.3.4. In the resonant coupling case each doublet contains two satellites, \( |d>_+ \) and \( |d>_- \) (Eqs. (3.1) and (3.2)), separated from each other by \( 2\hat{g}_{12} \). This system is then embedded by the field resonant with the 2-3 transition into \( |3> \) (see Fig.4.6). In this way the two mixed states (\( |d>_+ \) and \( |d>_- \)) optically structure \( |3> \) at the frequencies determined by the Rabi frequency of the dressing field (\( \hat{g}_{12} \)). Such an optical embedding contaminates \( |3> \) at \(-\hat{g}_{12} \) and \(+\hat{g}_{12} \) from its line-center. As a result at these frequencies the emission or absorption spectrum becomes undetectable for transitions from the ground state. In other words, it creates emission or transparency holes. When
Figure 4.5: Early stages of development of the resonance fluorescence spectra (in arbitrary units) associated with the 3-0 transition in a resonant Π system beyond the low-field limit for various values $\hat{g}_{23}$ and $\hat{g}_{12}$. (a) represents $\hat{g}_{23} = 0.1$ and $\hat{g}_{12} = 0.3$; (b) and (c) represent the cases where $\hat{g}_{23} = 0.1$ and $\hat{g}_{12} = 1.2$; and $\hat{g}_{23} = 0.1$, and $\hat{g}_{12} = 0.6$, respectively; and (d) is for the case where $\hat{g}_{23} = 1$ and $\hat{g}_{12} = 0.6$. 

\[ S(\omega) \]

\[ (\omega - \omega_{03})/\Gamma_3 \]
Figure 4.6: Schematic diagram of the coupling process of a system with a resonant $\Pi$ configuration. The shaded area represents the broad state $|3\rangle$ and the unlabelled double-headed arrows show the field resonant with the 2-3 transition with frequency $\omega_2$. We turn off the dressing field only $|2\rangle$ is embedded into the line-center of $|3\rangle$. However, since here the embedded state is a pure state, we do not see any change in the frequency of the generated dip as we increase the field intensity (Fig.4.5.d).

Let us now discuss the evolution of the 3-0 emission spectrum in the low-field limit when the fields are equally detuned from their corresponding transitions but with opposite signs, $\hat{\Delta}_1 = -\hat{\Delta}_2 = +1$. Fig.4.7 shows the evolution of the emission spectra as a function of the coupling field intensities. For $\hat{g}=0.05$ (dashed line) we see generation of a weak structure at the tail of the uncoupled emission spectrum at -1 of the scaled frequency. Once we increase $\hat{g}$ to 0.2 this structure becomes larger and red shifted while another
structure (dip) is generated at the vicinity of uncoupled line-center of $|3>$ (dotted line). A further increase in the field intensities causes further enhancement of the structure at the left and the dip on the right.

This situation can also be explained in terms of the structuring picture presented for the resonant coupling in this section. Here, since the fields are off-resonant, the initial one-photon dressing between $|1>$ and $|2>$ requires higher field intensities. Therefore, before they are effectively dressed by $\hat{g}_{12}$, the bare state $|2>$ is optically embedded into $|3>$ by the embedding field. Such a one-photon structuring process causes generation of the weak structure at the tail of $|3>$ at -1 of the scaled frequency. However, once the dressing field intensity is increased the coupling between $|1>$ and $|2>$ becomes strong enough to convert the bare state $|2>$ into a detuned dressed doublet, similar to that
seen in Fig.3.2. This doublet has a weak peak around +1 above the $|2\rangle$ line-center and a strong peak slightly below this line-center. The optical embedding of these two peaks into $|3\rangle$ respectively generates a weak dip close to the uncoupled emission line-center of the 3-0 transition and a strong structure slightly lower than -1 of the scaled frequency. Increasing the field intensity shifts the peaks in the doublet away from each other and so do the frequencies in which they structure $|3\rangle$.

When we detune the fields such that $\Delta = \Delta' = -1$, a similar structuring phenomenon is repeated. Here as Fig.4.8 shows for $\tilde{g}=0.2$ we see generation of a peak at +1 of the scaled frequency and a much weaker structure at a frequency slightly above +2 (dashed line). For $\tilde{g}=0.4$ the structure at +1 remains at the same frequency but it becomes broader. On the other hand the structure on the right is blue shifted and becomes larger and stronger. This is accompanied by a similar frequency shift in the essential peak to the left. Once we approach the high-field limit the spectrum develops into a triplet, one peak at center with scaled peak frequency at +1, and two sideband peaks (Figs.4.2a and 4.2b).

To test the results of this section experimentally one has to choose an atomic system which satisfies the conditions discussed in this paper: coupling of two metastable states to a higher transition level. Since, except for some changes in the case of detuned coupling, the results of this section remain essentially the same if we consider the energies of the metastable states larger than that of the upper transition level ($\Xi_-$ system), one may find such a system in the transitions of Rb atoms. As mentioned in the preceding section transitions in Rb atoms have already been used to study LIT in systems with either ladder or $\Lambda$ configurations [34, 51]. In Ref. [51], for example, the system was constructed by considering transitions from 5s to 5p and from 5p to 5d with 780 and 776 nm wavelengths, respectively. Since the width of 5p is one order of magnitude larger than that of 5d, when these states were coupled by a laser field, a well-resolved LIT was observed in the transition between 5s and 5p. To test the results of this paper one may
Figure 4.8: Early stages of development of the resonance fluorescence associated with the 3-0 transition in a detuned $\Pi$ system with $\Delta_1 = \Delta_2 = -1$ for various values of $\hat{g}_{23} = \hat{g}_{12} = \hat{g}$. The dashed line corresponds to $\hat{g} = 0.2$, and dotted line to $\hat{g} = 0.4$. All other specifications are the same as those in Fig.4.1.
add to this system coupling between 7p and 5d states using a second laser field with 4615 nm wavelength [54]. Here since 7p has nearly the same width as that of 5d (one tenth of that of 5p), the requirement of this paper is nearly satisfied. Therefore we expect to see the basic features presented in this section in the absorption spectrum associated with the 5s to 5p transition. Note that if |2 > and |1 > are not narrow compared to |3 >, we expect to see an initial broadening in the dynamics of the system. However, as we increase the field intensities a triplet spectrum should start to be resolved. This will be discussed in the following sections.

4.4 One- and two-photon coupling mechanisms

In the previous section we discussed LIT and dark line effects in terms of structuring processes. This picture was especially useful for describing the system dynamics at low field intensities, and for giving an insight into the involved quantum interferences. At medium and high field intensities, however, one can focus on the dynamics of the spectral peaks and explain their associated coupling mechanisms. Knowing these mechanisms allows one to present a more in-depth picture of quantum interference effects and to investigate coherent control of the optical transitions.

To explain the dynamics of emission peaks and their natures at high field intensities in a II system, we use a dressed-state picture. For this system we identify two distinct coupling mechanisms. One of these mechanisms is responsible for mixing of |1 >, |2 > and |3 >, through one-photon coupling, and the other for mixing |1 > with |3 > through |2 >, two-photon or Raman coupling. In the case of Raman coupling the state |2 > acts only as an intermediate state. The signs of these coupling mechanisms in the resonance fluorescence of the 3-0 transition were seen in Sect.4.2 as the development of triplet spectra. The central peaks here are associated with the two-photon coupling mechanism. The state related to this coupling process can be an anti-symmetric superposition of |3 > and |1 >. Therefore it could become immune from decay into the intermediate state [48].
This can be clearly seen in a dressed state picture. Using the development of Manka et al. [35] for dressed states of three strongly coupled levels, we find the state associated with two-photon coupling under the resonant coupling condition as follows:

\[ |r> = -(\sin{\beta})|1> + (\cos{\beta})|3>, \quad (4.1) \]

with

\[ H_{\text{int}}|r> = 0. \quad (4.2) \]

Here \( \beta \) is defined by the following relation:

\[ \tan{\beta} = \frac{g_{12}}{g_{23}}. \quad (4.3) \]

Eq. (4.2) shows that under resonant coupling conditions the peak corresponding to the two-photon coupling process should occur at the same energy as that of the uncoupled peak line-center, a fact seen in Fig.4.1.

On the other hand, the one-photon coupling process is associated with the sideband peaks. The states related to this process are linear superpositions of all three coupled states. Again in a dressed state picture and under the resonant coupling condition we can have the following forms for these states [35]:

\[ |s> = \frac{1}{\sqrt{2}}[(\cos{\beta})|3> + |2> + (\sin{\beta})|1>], \quad (4.4) \]

\[ |t> = \frac{1}{\sqrt{2}}[(\cos{\beta})|3> - |2> + (\sin{\beta})|1>]. \quad (4.5) \]

where

\[ H_{\text{int}}|s> = \hbar G|s>, \quad (4.6) \]

\[ H_{\text{int}}|t> = -\hbar G|t>, \quad (4.7) \]

and \( G = \sqrt{g_{23}^2 + g_{12}^2} \). This shows that the peaks corresponding to the one-photon coupling process are located at \(-G\) and \(+G\) from the uncoupled peak line-center. This fact can
be seen in Fig. 4.1 as we reach the high-field limit. The same coupling processes can also be found in the δ₁⁻ system.

Eqs. (4.1)-(4.7) can also be used to explain the phenomena seen in Fig. 4.3. In fact Fig. 4.3a represents the destruction of the coherences generated by the coupling of |1> with the rest of the Π system when the ratio \( \tilde{g}_{12} \) is decreased. To see this note that this ratio which determines \( \beta \), is responsible for different dressed state mixtures. If we increase \( \tilde{g}_{23} \) as compared to \( \tilde{g}_{12} \), the states |s> and |t> become independent of |1> and therefore illustrate a simple coupling between |2> and |3> (A configuration). The same is true for the inverse case shown in Fig. 4.3b. Here an increase in \( \tilde{g}_{12} \) destroys the coherences involving |3>. There it virtually decouples the two-level system from the dressed system.

One can relate the frequency of the central peaks in the Fig. 4.2 to the efficient channels of the two-photon coupling processes generated by the detuned fields. One of these channels, in the case of Figs. 4.2a and 4.2b, starts by the field with frequency \( \omega_1 \) and \( \hat{\Delta}_1 = -1 \) from the |1> line-center to +1 of scaled frequency above the |2> line center and then by the field with frequency \( \omega_2 \) and \( \hat{\Delta}_2 = -1 \) it continues to +2 above of the |2> line-center. The existence of a finite damping rate of |3>, however, generates other channels which in the average results in a central peak centered at +1 above the |3> line-center. Such a consideration can also explain why in the case of Figs. 4.2c and 4.2d where \( \hat{\Delta}_1 = -\hat{\Delta}_2 \), the central peak is not shifted against the uncoupled peak line-center.

4.5 Competition between one- and two-photon coupling processes, elements of coherent control of optical transitions

In this section we study the competition between one and two-photon coupling processes. This leads to explanations of the coherent effects discussed in the previous sections and shows how one can control optical transitions in atomic systems. This competition depends on the coupled states' widths in comparison to the coupling field intensities. In a
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Π system, for example, the strengths of one- and two-photon effects depend on the ratio of \( \frac{\beta}{\Gamma_2} \) and \( \frac{\gamma}{\Gamma_1} \), respectively. This means that if the former ratio reaches unity sooner, the one-photon coupling mechanism becomes dominant and vice versa. For the type of the Π system discussed in the previous sections, however, since both \( |1> \) and \( |2> \) were metastable, for any value of \( \hat{g} \) both mechanisms contribute equally. These competitive features also occur in the Ξ₋ system. Therefore one can present an argument similar to that of the Π system but should exchange \( \Gamma_1 \) with \( \Gamma_3 \). In the following we explore this competition in the Ξ₋ system by considering the cases where one of these mechanisms is dominant. The dynamics of such a system are obtained using Eq. (2.82).

4.5.1 Dynamically damped dark effect; dominance of the one-photon coupling process

In this subsection we consider the case where one-photon coupling is the dominant process. In a Ξ₋ system this can be achieved by choosing \( |2> \) to be a metastable state (\( \Gamma_2 << 1 \)) and \( |3> \) a relatively broad one. This can be experimentally tested in potassium atoms. Here we may consider a laser field to couple \( 4p \) with \( 6d \), and another field to couple \( 6d \) with \( 4f \). These states with 26.6 ns, 1066 ns, and 62.9 ns lifetimes [54] act, respectively, as \( |1> \), \( |2> \), and \( |3> \) in the Ξ₋ system. As Fig.4.9a shows, when \( \hat{g} = 0.2 \) the emission spectrum of this system develops a "nearly dark" line (solid line). A further increase in the field intensities decreases the associated peak amplitudes and shifts them away from each other. This continues until a limit is reached where the central peak starts to appear. This is shown in Fig.4.9a for \( \hat{g} = 0.5 \) (dashed line). Note that here the depth of the dark line is determined by the width of the \( |3> \). The broader this state is, the deeper is the dark line.

One can explain the evolution seen in Fig.4.9a in terms of the dominant coupling process, one-photon coupling. Since \( |3> \) is relatively broad, the coupling between this level and \( |2> \) and \( |1> \) is relatively weak, therefore the coupling process occurs in
three stages. The first stage is nearly pure coupling between $|2>$ and $|1>$, similar to the kind of coupling that occurs in a $\Xi$ system. This causes a dark line effect in the emission spectrum of $|1>$. Here $|3>$ causes a slight broadening of the corresponding emission peaks (Fig.4.9b). As the field intensities increase, the broad state $|3>$ starts to contribute more (second stage). This generates a strong broadening of the individual peaks and a shift in their frequencies[13], yet it does not destroy the emission hole. The broadening and frequency shift continue until the third stage is reached, where the two-photon coupling effects appear. Here the spectrum, although extremely broad, has three peaks of emission (not shown).

An interesting feature to mention here is the way in which the system evolves from a situation where one- and two-photon coupling have the same strength to the case where one-photon coupling is dominant. To see this we consider $\Xi$ systems with three different values of $\tilde{\Gamma}_3$, $\tilde{\Gamma}_2=0.001$, and $\tilde{g}=3$. Under these conditions and when $|3>$ is a metastable state ($\tilde{\Gamma}_3 = 0.001$), as Fig.4.10 shows the emission spectrum becomes a triplet, similar...
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Figure 4.10: Emission spectra (in arbitrary units) corresponding to the 1-0 transition of a resonant Ξ system with $\Gamma_2=0.001$ and $g = 3$, and three values of the $|3>$ width ($\Gamma_3=0.001, 1, \text{and} 2$). The thin solid line corresponds to $g = 0$.

the Π system response (Fig.4.1). Increasing the width of $|3>$, however, causes the central peak to become broader and suppressed. The sideband peaks, however, mainly broaden and shift in frequency. In the limit $\Gamma_3 >> 1$ the central peak vanishes and the dynamically damped dark line effect occurs.

4.5.2 Linewidth Suppression of Resonance Fluorescence; Dominance of the Two-Photon Coupling Process

In this section we show how the dominance of two-photon coupling can also induce linewidth suppression. To see this in a Ξ system we should consider $|3>$ a metastable state and $|2>$ a broad one. This can be found in the transitions of Li atoms to some extent. Here if a laser couples 2p and 3d, and another laser 3d and 7f, the emission corresponding to the 2p to 2s transition should show line-width suppression. The corresponding wavelengths of these transitions are 610.5 nm, 1003.6 nm, and 671 nm, respectively. Also here 2p, 3d, and 7f with 27.3, 14.7, and 376 ns lifetimes act as $|1>$, $|2>$, and $|3>,$
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Figure 4.11: Line-width suppression in the emission spectrum (in arbitrary units) of a $\Xi_-$ system in Li atoms. The solid line corresponds to $\hat{g} = 0$ and the dashed line to $\hat{g} = 5$. The results of calculations for such a system are shown in Fig.4.11. A prominent feature of this spectrum is the generation of a central peak with amplitude larger than that of the uncoupled emission peak, but narrower in width.

If one considers a system in which $|2>$ is much broader than $|1>$ ($\hat{\Gamma}_2=10$) and $|3>$ is a metastable state ($\hat{\Gamma}_3=0.001$), the main effect of the fields would only be a dynamic narrowing of the emission spectrum associated with the $|1>$ to $|2>$ transition (see Fig.4.12). Here there is no sign of the sideband peaks. Under the conditions of Fig.4.12 the minimum width of the emission spectrum is half of that of the uncoupled peak, $W_c = \Gamma_2/2$.

It is interesting to see how the emission spectrum alters as the coupling process is changed from a situation where one- and two-photon coupling have nearly the same strengths as compared to dominant two-photon coupling. This is shown in Fig.4.13 for the case of a $\Xi_-$ system, where we assume that $|3>$ is a metastable state, $\hat{g} = 3$, and $\hat{\Gamma}_2$ is a variable parameter. In the absence of any field ($\hat{g} = 0$) we have a Lorentzian line (thin
Figure 4.12: Emission spectra (in arbitrary units) corresponding to the 1-0 transition of a resonant $\Xi_-$ system with $\hat{\Gamma}_2=10$, $\hat{\Gamma}_3=0.001$ and variable Rabi frequency ($\hat{g}$).

When $\hat{g} = 3$ and $\hat{\Gamma}_2 << 1$ we have a triplet spectrum, similar to the corresponding case in Fig.4.10. As $\hat{\Gamma}_2$ increases the sideband peaks are suppressed in amplitude and the central peak increases in amplitude without changing its width. Ultimately when $\hat{\Gamma}_2 >> 1$ the situation becomes similar to that of Fig.4.12, in that the sidebands vanish and the central peak reaches its maximum amplitude (not shown). In this limit any increase in the field intensities does not induce any change in the emission spectrum. In other words, the system's dynamics become saturated.

Up to now we have considered the cases where the coupling fields were resonant with their corresponding transitions. Now let us consider the case where these fields are detuned. To show this we first consider the case where $\hat{\Delta}_2 = -2$ and $\hat{\Delta}_1 = 0$. As Fig.4.14a shows, when $\hat{g} = 3$ the emission spectrum is red-shifted (long dashed line). There is also some residual emission at higher frequencies. When $\hat{g} = 15$, however, the residual emission disappears and the result of the coupling process is generation of an emission peak at $\omega_r - \omega_{01} = -\Gamma_1$ (short dashed line). As in the resonant case this peak has higher amplitude than that of the uncoupled peak (solid line), but is narrower. The
Figure 4.13: Emission spectra (in arbitrary units) corresponding to the 1-0 transition of a resonant $\Xi_-$ system with $\tilde{\Gamma}_3=0.001$, $\tilde{g}=3$ and three values of the $|2>$ width ($\tilde{\Gamma}_2=0.001$, 1, and 2). Other notations are the same as those in Fig.4.10.
Figure 4.14: Resonance fluorescence spectra (in arbitrary units) corresponding to the 1-0 transition of a detuned Ξ\(_{-}\) system. In (a) \( \Delta_2 = -2 \) and \( \Delta_1 = 0 \) with field intensities corresponding to \( \hat{g} = 3 \) (dashed line) and \( \hat{g} = 15 \) (dotted line). In (b) \( \Delta_2 = \Delta_1 = -2 \) for field intensities with \( \hat{g} = 7 \) (dashed line) and \( \hat{g} = 20 \) (dotted line). The solid lines correspond to \( \hat{g} = 0 \).

The case where both fields are detuned such that \( \Delta_1 = \Delta_2 = -2 \) is shown in Fig.4.14b. Here one can see that the overall features are nearly the same as those in Fig.4.14a, but the emission peak is generated at \( \omega_r - \omega_{01} = -2\Gamma_1 \). When we consider the case where \( \Delta_1 = -\Delta_2 \), the emission spectrum is narrowed as in the resonant case, independent of the magnitude of the detunings, with no frequency shift.

Note that the previous studies of line narrowing by optical fields have been done in different systems. In Ref.7, for example, a driven two-level atom is put at the center of a cavity, and by increasing the field intensity the corresponding peaks of the Mollow spectrum become narrower. In Fig.4.12, however, the narrowing process occurs virtually as squeezing the natural Lorentzian line of the uncoupled emission associated with the 1-0 transition. This is somewhat similar to the situation in a three-level atom where
the two upper levels decay to the ground state through a common vacuum. It has been shown that under particular conditions the emission spectrum of one of the upper levels can be narrowed and frequency shifted due to quantum interferences [55].

4.6 Multi-photon Quantum interference picture

In the previous sections we showed that when the dominant coupling mechanism of a Ξ_- or Π system was either one- or two-photon coupling, the emission spectrum could develop a dynamically-damped dark line effect, become suppressed in line-width, or generate two transparency holes or dark lines with tunable frequencies. These features, when accompanied with our capability to tune the field frequencies, showed that one could manipulate the emission and absorption spectra of atoms by controlling the involved coherent processes. In this section we explain these features in terms of multi-photon quantum interferences. To do this let us first consider the case where both one- and two-photon coupling contribute equally. In a Ξ_- system this occurs when the widths of $|2>$ and $|3>$ are the same. Of particular interest, however, is the case when these two states are both metastable. As shown in Fig.4.1 this causes generation of two dark lines at $\pm g$. This means that, in contrast to the Ξ or Λ system where the dark line was generated at a fixed frequency, here they are generated at two frequencies and shifted away from each other as $g$ increases. Inspection of the Ξ_- system dynamics suggests that, in contrast to the Λ or Ξ system, there exists an extra interfering path here. This path is a three-photon Raman transition between $|0>$ and $|3>$ through $|1>$ and $|2>$, 0-1-2-3 (see Fig.4.15). The dark lines seen in Fig.4.1 are associated with quantum interferences between this path and the 0-1 direct and the 0-1-2 two-photon path. Under the condition where the the two- and three-photon paths have the same contributions in a Π system they leave the line-center emission unchanged but destroy emission at $\pm g_{12}$ (Fig.4.1).

The dynamically damped dark line effect and line-width suppression can also be explained in terms of these interference paths. In the case of the dynamically damped
Figure 4.15: Schematic diagram of the quantum interferences involved in a $\Xi_-$ system. (1) is the 0-1 direct, (2) the 0-1-2 two-photon Raman, and (3) the 0-1-2-3 three-photon Raman path.

dark line effect, the suppression of the two-photon coupling process reduces the effect of the three-photon Raman path allowing an interference between the 0-1 direct and 0-1-2 Raman paths. This explains why the basic features of this kind of dark line effect are similar to some extent to those seen in the $\Xi$ system. In contrast to the $\Xi_-$ system, however, increasing the width of $|2\rangle$ in the $\Xi$ system destroys quantum interferences. Because of this the emission spectrum of this system becomes broadened and then splits into a doublet as the field intensities increase. The fact that in the $\Xi_-$ system (Fig.4.9) a dark line can exist regardless of the field intensities suggests that $|3\rangle$ does not affect $|2\rangle$ but broadens $|1\rangle$. This means that $|3\rangle$ participates in the coupling process through a residual two-photon coupling with $|1\rangle$.

The dynamic line-width suppression is another sign of the multi-photon quantum interference processes in atomic systems. In fact Fig.4.13 illustrates the way in which the emission spectrum of the 1-0 transition evolves as the two-photon Raman path (0-1-2) vanishes in a $\Xi_-$ system. This occurs parallel to the reduction of the one-photon coupling contribution. Under the conditions of Figs.4.11 and 4.12, the quantum interferences
mainly occur between the 0-1 direct and the 0-1-2-3 Raman path. This suggests that the increase of the emission spectrum amplitude at the line-center of 1-0 transition can be attributed to the constructive interference between these two paths.

The results of Fig.4.14 show that constructive quantum interference can occur at frequencies besides that of the line-center if we consider $\Delta_1$ and $\Delta_2 \neq 0$. This is because the field frequencies actually affect the frequencies of the interfering paths. For example for the case of Fig.4.14a since $\Delta_1 = -2$ the effective quantum interference occurs below the uncoupled emission line-center with a maximum at $\Delta_1/2$. In the case of Fig.4.14b, however, the interference is centered at -2, since both fields are equally detuned by -2.

In summary, for a $\Xi_-$ system we showed that:

(i) When both $|2>$ and $|3>$ are considered to be metastable states, in addition to the 0-1 and 0-1-2 paths, the 0-1-2-3 Raman path exists. These three paths can destructively interfere with each other generating two tunable dark lines. Here the line-center of the emission spectrum of the 2-1 transition remains unaffected by the quantum interferences.

(ii) When level $|3>$ is much wider than $|2>$, the three-photon Raman path is suppressed. The quantum interferences then would be between the 0-1 and 0-1-2 paths. This destructively removes the emission at the line-center of the 0-1 transition generating a dynamically damped dark line effect.

(iii) If $|2>$ is considered to be a broad state compared to $|3>$, the 0-1-2 path is suppressed. In that case the constructive interference between the 0-1 direct and 0-1-2-3 three-photon Raman path increases the emission amplitude at the line-center. This is accompanied by dynamic narrowing of the natural emission spectrum of the 1-0 transition.

4.7 LIT and dark line effects in a $\Pi$ system

In the previous sections we discussed LIT and dark line effects in $\Pi$ and $\Xi_-$ systems. In terms of coupling mechanisms and quantum interferences these two systems showed some
similarities, i.e. they were both affected by the one- and two-photon coupling processes and involved with three types of interfering paths. Their differences came from the fact that they described realistic systems with different damping rates configurations. As we find out in this section, however, the case of the K system (Fig.2.3b) is different both in terms of coupling mechanisms and quantum interferences.

To discuss LIT in the K system we apply the formalism developed in Sect. 2.2 (Eq. (2.97)) to a realistic system consisting of $^{85}$Rb atoms (see Fig.4.16). Here $|1\rangle$, $|2\rangle$ and $|3\rangle$ are $5S_{1/2}$ with $F=3$, $5P_{3/2}$ with $F=3$, and $5D_{5/2}$ with $F=4$, respectively. Also the ground state is $5S_{1/2}$ with $F=2$. The results for the resonant coupling ($\Delta_1 = \Delta_2 = 0$) are shown in Fig.4.17. We see here that the LIT in K system is apparently similar to that seen in the $\Lambda$ (or $\Xi$) system. Inspection of the graphs, however, shows that the associated peaks here are separated from each other by $2\sqrt{2}g$, rather than $2g$ predicted for the $\Lambda$ system. Note that the absence of any central peak, in contrast to the LIT in the $\Pi$ (or $\Xi_-$) system, suggests that the dynamics of the K system are fully governed by the one-photon coupling of all three coupled states. This is compatible with the concepts developed in Sect. 4.3.
A distinctive difference between K and A system dynamics, however, appears when we consider the case when the fields are detuned. To see this we show in Fig.4.18 the results of calculations for a system which is the same as that of Fig.4.17 but with $\hat{\Delta}_1 = \hat{\Delta}_2 = +2$. Here we see that the spectrum of the K system develops two transparency holes with central frequencies at +2 and -2 of the scaled frequency. As the field intensities increase these frequencies remain unchanged but the holes become wider. In Fig.4.18a the central peak is much wider than the sideband peaks. For higher field intensities, as Fig.4.18b shows, the sideband peaks become wider and shifted away from each other, and the central peak becomes narrower. Ultimately when we reach the high-field limit the central peak vanishes and the situation becomes similar to that seen in the resonant coupling case (Fig.4.17). Note that the asymmetries seen in the central and sideband peaks are caused by the inequality of the widths of $5S_{1/2}$ ($|1>$) and $5D_{5/2}$ ($|3>$) in rubidium atoms.
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Figure 4.18: Absorption spectra (in arbitrary units) corresponding to the 0-2 transition of a detuned K system with $\hat{A}_1 = \hat{A}_2 = +2$ and (a) $\hat{g} = 1$, and (b) $\hat{g} = 3$. All other specifications are the same as those in Fig.4.17.

4.8 Two-field structuring processes and quantum interferences in a K system

In section 4.3 we showed how one can describe the LIT effect in terms of a structuring process in a $\Pi$ or $\Xi_-$ system. In this section we use this picture to explain LIT in the K system. Here the structuring process occurs due to direct optical embedding of the two metastable states ($|1 >$ and $|3 >$) into the line-center vicinity of $|2 >$. Fig.4.19 shows such a structuring process at the early stages of the coupling process when (a) $\hat{A}_1 = \hat{A}_2 = 0$, and (b) $\hat{A}_1 = \hat{A}_2 = 0.2$. Note that for the detuned case when $\hat{g} = 0.02$ the fields generate two narrow transparency holes at ±0.2 (Fig.4.19b). At higher field intensities ($\hat{g} = 0.1$, for example) these frequencies remain unchanged but the holes become deeper and broader. At this stage the spectrum consists of a very subnatural triplet spectrum.

In terms of the structuring process, under resonant coupling condition both metastable states are embedded into the same frequency within $|2 >$. Therefore, they generate two superimposed uncorrelated dips in the absorption spectrum (Fig.4.19a). This explains why no triplet spectrum is generated for any field intensities. When the fields become
Figure 4.19: Early stages of development of the absorption spectra (in arbitrary units) associated with the 0-2 transition of the K system for various values of $g$ and (a) a resonant, and (b) detuned, $\Delta_1 = \Delta_2 = 0.2$, case. Solid lines correspond to $g = 0$, dashed lines to $g = 0.02$ and dotted lines to $g = 0.1$

detuned, however, each field structures $|2\rangle$ at a specific frequency. Under the conditions of $\Delta_1 = \Delta_2 = 0.2$, $|3\rangle$ is embedded at 0.2 above and $|1\rangle$ at 0.2 below the line-center of $|2\rangle$. This process is schematically shown in Fig.4.20. If one considers $\Delta_1 = -\Delta_2$, for any field frequencies and intensities the transparency holes are driven into $\Delta_1$.

Based on this concept the central peaks in Figs.4.18 and 4.19b are just the remaining parts of $|2\rangle$ which are not contaminated by the metastable states yet. This is consistent with the quantum interference processes involved in the K system and the fact that in this system there is no two-photon coupling mechanism. The interference processes responsible for generation of the transparency holes in a K system are schematically shown in Fig.4.21. Here we see that only one direct and two two-photon Raman paths are involved and no three-photon path exists. The Raman paths are responsible for transition between $|0\rangle$ and $|1\rangle$ and that between $|0\rangle$ and $|3\rangle$ through $|2\rangle$. Here since these two Raman paths are uncorrelated, the interference processes occur independently between each of these paths and the 2-0 direct path. This explains the evolution of the LIT in a
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K system and why each transparency hole’s dynamics were the same as those in the \( \Lambda \) or \( \Xi \) system.

4.9 Ultra-narrow gain generation in atomic systems

In the previous sections we found that quantum interferences were responsible for generation of one or two sharp dips in the emission (dark lines) or absorption spectrum (transparency holes) of an atomic transition. Recent studies have shown that under specific conditions in a \( V \) system (Fig.2.2c), quantum interferences can also generate an ultranarrow spectral line in the resonant fluorescence. This occurs when the energy spacing between \(|1>\) and \(|2>\) is so small that the laser with frequency \( \omega_1 \) drives both levels. A similar spectral line has also been predicted in the resonance fluorescence of the 0-2 transition in a \( \Lambda_g \) system (Fig.2.4b). This occurs when \(|1>\) is a metastable state and the intensity of the laser field with frequency \( \omega_2 \) is much less than that of other field with frequency \( \omega_1 \). This effect has been attributed to an electron-shelving effect [44, 45]. Due to the transitions of electrons into the metastable state, the resonance fluorescence corresponding to the 0-2 transition is randomly interrupted. The average of this process over a long time generates a narrow profile superimposed on the resonance fluorescence.
of the driven 2-0 transition.

In this section we predict a new type of ultranarrow spectral line generation in the 0-2 transition of a $\Lambda_d$ system. We show that under specific conditions, this transition develops a narrow gain profile. This gain, which occurs in the absence of any inversion, switches into an ultranarrow absorption profile as we increase the field resonant with the 0-2 transition. We explain this effect as a stimulated Rayleigh scattering process using the structuring concept developed for the K system.

4.9.1 Switching between ultranarrow gain and absorption spectra

To study generation of the ultra-narrow gain-absorption process we consider the 0-2 absorption spectrum in a $\Lambda_d$ system. The schematic diagram of such a system in $^{87}$Rb atoms is shown in Fig.4.22. In the absence of the optical fields the absorption spectrum of the 0-2 transition has a Lorentzian line-shape. However, as Fig.4.23 shows when the field with frequency $\omega_2$ is turned on with $\Delta_2 = 0.1$ and $\tilde{g}_{12} = 0.1$, we have an asymmetric transparency hole centered at $\Delta_2$ below the 0-2 transition line-center (solid line). When
we keep $\hat{g}_{12}$ unchanged but turn on the second field with $\hat{g}_{02}=0.1$, the transparency hole becomes deformed and shifted towards the line-center of the uncoupled peak (dashed line). A further increase in the value of $\hat{g}_{02}$ causes further shift in the frequency of the hole while the whole absorption spectrum is decreasing in amplitude. For $\hat{g}_{02}=0.3$, we see generation of a narrow gain region.

To see the overall absorption spectrum under these conditions we show in Fig.4.24a the full spectrum corresponding to the dashed-dotted line in Fig.4.23. Here the absorption spectrum is deformed and the central frequency which had maximum absorption amplitude in the absence of any optical field now has gain. This phenomenon, however, is sensitive to $\hat{g}_{02}$. To see this we show in Figs.4.24b-4.24d three stages of its development for three values of $\hat{g}_{02}$. In Fig.4.24b we can see that by increasing $\hat{g}_{02}$ to 0.6 the gain disappears and the absorption spectrum now has a small structure at its line center. Here we can distinguish two sets of asymmetric sideband peaks. For $\hat{g}_{02}=0.9$ the structure flips into a sharp spectral absorption peak (Fig.4.24c) while the overall spectrum becomes more symmetric. Fig.4.24d presents the case where $\hat{g}_{02} \gg \hat{g}_{12}$. Here the spectrum essentially consists the linear response of a strongly driven two-level system ($|0>$ and $|2>$) superimposed by a very sharp line.

Note that the sharp feature seen at the line-center of the 0-2 transition has been
Figure 4.23: Absorption spectrum of the 0-2 transition in the $\Lambda_d$ system for $\hat{\Delta}_1=0$ and $\hat{\Delta}_2=+0.1$, $\hat{g}_{12}=0.1$, and various $\hat{g}_{02}$. The solid line corresponds to $\hat{g}_{02}=0$, the dashed line to $\hat{g}_{02}=0.1$, the dotted line to $\hat{g}_{02}=0.2$, the dashed-dotted line to $\hat{g}_{02}=0.3$, and the long dashed line to $\hat{g}_{02}=0.4$. 
attributed to the electron-shelving effect [44, 45]. Also the two sets of sideband peaks can be attributed to the transitions of the probe field between dressed states caused by coupling of the three levels (Eqs. (4.1)-(4.7)). When $\hat{g}_{02} \gg \hat{g}_{12},$ however, similar to the case of \Pi or \Sigma- system, coherence destruction occurs. This decouples the metastable state ($|1>),$ and diminishes the inner sideband and the sharp feature.

**4.9.2 Ultra-narrow gain generation description based on LIT in K system**

To understand the nature of the ultra-narrow gain process and its rapid switching, we use the structuring picture of the LIT effect in the K system. In this system, similar to the $\Lambda_{g}$ system, the upper transition level is coupled to two metastable states. However, in the K system since the ground state is not coupled to the upper transition level, there
is no effect due to the electron excitation. To shorten the following discussion we avoid repeating notations of the K and \( \Lambda_d \) system when they are the same.

Under the same conditions as those of Fig. 4.23 and when \( \gamma_{02} = \gamma_{23} = 0 \), the absorption spectrum of the K system (Fig. 4.25a) is identical to that of the \( \Lambda_d \) system (Fig. 4.23 solid line). Here the structuring process occurs by the field with frequency \( \omega_2 \). When \( \gamma_{02} = \gamma_{23} = 0.1 \), however, while the absorption spectrum of the \( \Lambda_d \) system shows a convoluted transparency hole, that of the K system shows generation of two distinct nearly similar transparency holes (Fig. 4.25b). In other words, the absorption spectrum associated with the 0-2 transition becomes a triplet. The central peak of this spectrum, however, is very sensitive to the value of \( \gamma_{23} \). As Fig. 4.25c shows for \( \gamma_{02} = \gamma_{23} = 0.3 \), the condition in which the spectrum of the \( \Lambda_d \) system shows the narrow gain profile (Fig. 4.24a), this peak becomes narrow and shifted towards lower frequencies (Fig. 4.25c). For \( \gamma_{02} = \gamma_{23} = 0.6 \) the narrow peak becomes strongly suppressed (Fig. 4.24d) while the spectrum of the \( \Lambda_d \) system shows a small structure at its line-center (Fig. 4.24b). A further increase in \( \gamma_{23} \) diminishes the structure in the K system response. The spectrum of the \( \Lambda_d \) system, however, develops a narrow absorption line. Under this condition the effect of the detuning is nearly lost in both systems.

These results can be used to explain the narrow gain profile generation and its switching process. To do this note that essentially the dynamics seen in the K and \( \Lambda_d \) systems are nearly similar when \( \gamma_{02} << 1 \). This includes generation of the narrow peak in \( |2> \) in the K system (Fig. 4.25b). In fact under the conditions when the gain process occurs, this peak appears at \( \Delta_2 \) below the \( |2> \) line-center. Therefore, it is separated from \( |0> \) by \( \omega_1 - \Delta_2 \). On the other hand, based on our discussion in Sect. 4.4, in a \( \Lambda_d \) system when \( |1> \) and \( |0> \) both are metastable, there is a strong two-photon coupling process between these two states. Therefore even when \( \gamma_{02} \) and \( \gamma_{12} \) are small the two-photon coupling states are well established. To find out where these states are located
Figure 4.25: Absorption spectrum of a K system for $\hat{g}_{12}=0.1$, $\hat{\Delta}_1=0$ and $\hat{\Delta}_2=+0.1$, and various driving field intensities. (a) corresponds to $\hat{g}_{02}=0$, (b) to $\hat{g}_{02}=0.1$, (c) to $\hat{g}_{02}=0.3$ and (d) to $\hat{g}_{02}=0.6$. 
we should look for the most efficient channels for the two-photon coupling process. Under the condition of ultra-narrow gain generation these channels start from $|1>$ to the narrow structure in $|2>$ by the field with frequency $\omega_2$ and then from there to $\Delta_2$ below $|0>$ by the field with frequency $\omega_1$ (see Fig. 4.26). As a result, an extremely narrow two-photon state $|r>$ is generated at $\Delta_2$ below $|0>$. Under these conditions the driving field with frequency $\omega_1$ is in fact resonant with the transition between the narrow structure in $|2>$ (shown by a horizontal arrow) and $|r>$. Therefore, the probe photons can be coherently scattered at this transition frequency, i.e. stimulated Rayleigh scattering occurs. This generates gain at the same frequency as that of the 0-2 transition. Note that since the two-photon dressed state and the central structure in $|2>$ are both very narrow the gain profile should be narrow. Also as $g_{02}$ increases, due to the suppression of the central structure in $|2>$ such a scattering process should diminish rapidly. This destroys the ultra-narrow gain profile. For higher values of $g_{02}$ the effect of detuning decreases and the electron-shelving effect starts to become a dominant process. This causes generation of a Mollow spectrum superimposed by a narrow profile (Fig.4.25d).
Figure 4.26: Energy diagram of the $\Lambda_9$ system under the conditions where the narrow gain profile is generated. $|r\rangle$ is the two-photon dressed state, the dashed line marked by a horizontal arrow is the structured middle narrow line in $|2\rangle$, and the thick bands are the broad sidebands at the sides of the narrow line (Figs. 4.25b-4.25d). Solid upward and dashed downward arrows represent the stimulated Rayleigh scattering process.
Chapter 5

Intraband excitonic dressing of quantum well structures

5.1 Introduction

The formalism developed in chapter 2 provides a sound basis for the theoretical development of optical coherent effects in QW's and atomic systems. In addition to predicting new results, the applications of this formalism to atomic systems presented in chapters 3 and 4 can be utilized to understand and predict new nonlinear optics in QW's. Bearing this in mind in the remaining parts of this thesis we present the results of our investigations for QW systems.

The optical properties of QW's are distinctively different from those of spatially homogeneous or bulk structures. One basic difference is the enhancement of the Coulomb interaction between electrons and holes in undoped QW's. This causes persistent structures in their absorption, photoluminescence (PL), and reflection spectra. As mentioned in the introduction, the other important feature of QW's is the possibility of having very large dipole moments for transitions between the conduction subbands. This occurs when the optical fields are polarized along the QW growth direction. Combination of these two features provides a basis from which we can explore and study new nonlinear optics of semiconductors. These optics play crucial roles in the development and design of optical devices and the enrichment of semiconductor physics.

Motivated by these features, we present in this chapter rigorous treatments for the linear interband absorption and emission (PL) spectra of an undoped QW when it interacts with a strong infrared field. This field is polarized along the QW growth direction and is nearly resonant with the transition between the first and second conduction subbands.
Figure 5.1: In-plane schematic diagrams of infrared coupling of two conduction subbands of an undoped QW. The thick two-sided arrows refer to the intense infrared field and the thin two-sided arrows to the absorption or emission processes. (a) refers to Ξ and (b) to Λ system.

(Fig.5.1). When the detection of this system from the valence band involves the first conduction subband, this system has a Ξ configuration, Fig.5.1a. On the other hand if it involves the second conduction subband the system has a Λ configuration (Fig.5.1b). These are similar to those in Figs.2.2a and 2.2b, respectively. Note that because of the parity considerations here the Ξ system can occur in both symmetric and asymmetric QW's. The Λ system, however, can only occur in asymmetric QW's where the interband transitions can have different valence and conduction subband indices. Except for our recently-reported developments (Ref. [30]-[31]), to our knowledge there exists no other theory to deal with the interband PL spectra of such infrared-coupled QW's. The interband absorption spectrum, however, has been treated by several authors. Their methods, however, have several shortcomings:

1. In the majority of these investigations the electron-hole Coulomb interactions were ignored [12]-[15]. Therefore, they did not illustrate realistic systems.
2. Although in one of these studies [11] the electron-hole Coulomb interaction was included, some of the important features of the QW excitations were ignored, however. This includes the quantum mechanical aspects of the infrared excitations in undoped QW's.

3. In none of these attempts was the possibility of multi-transition excitations considered. These excitations occur when the infrared field is nearly resonant with two or more transitions in the conduction band. Many QW systems which were considered in the past to study this subject experimentally were actually affected by these excitations [11, 56].

In this chapter we address (1) and (2) and in the next one the case of multi-excitation events is discussed. Here we first briefly review some of the basic electrical and optical properties of QW's. We then address, to our knowledge for the first time, the selection rules of the infrared-induced excitonic transitions in undoped QW's. This chapter also includes our detailed investigation of the interband absorption and PL spectra of the QW in the presence of strong infrared fields. In chapter 11 we will return to this subject by putting to test some of the results of this chapter experimentally.

5.2 Review of the electronic and optical properties of quantum wells

In III-V QW structures, such as GaAs/Al\textsubscript{2}Ga\textsubscript{1−2}As, carriers in the valence and conduction bands are confined in the same layer (Fig. 5.2a). This gives rise to discrete levels (subbands) both in the valence and conduction band along the growth direction within the same layer. The energy spacings between these subbands depend on the QW width, material structure, etc. Confinement of carriers in QW's causes drastic changes in their optical properties. To recall these, let us review some of the important features of these structures.
Figure 5.2: Schematic diagram of a III-V quantum well along the growth direction (a), and its dispersion in the basal plane (b).

5.2.1 Conduction band energy states

Conduction subbands of QW's are usually well-spaced. Within the envelope-function approximation these subbands can be described by:

$$\phi_{ik}(r) = e^{ik \cdot \rho} C_i(z) U_i.$$  \hspace{1cm} (5.1)

Here $\rho$ and $z$ are the coordinates of an electron in the $i$th subband with the in-plane wave-vector $k$, and $C_i(z)$ is its envelope function. Within the effective mass approximation this function can approximately be obtained from the Schrödinger equation [57]:

$$\left[-\frac{\hbar^2}{2m^*} \frac{d^2}{dz^2} + V_c(z)\right]C_i(z) = E_{i,k}^c C_i(z).$$ \hspace{1cm} (5.2)

Here $m^*$ is the effective mass of the electron and $E_{i,k}^c$ is its energy. $V_c(z)$ refers to the conduction band potential. For the Fig.5.2 structure $V_c(z)$ is a symmetric square well. $U_i$ in Eq. (5.1) is the Bloch function of the electron. We associate the ket $|S>$ to $U_i$, which explicitly shows the s-type nature of this function.
5.2.2 Valence band energy states

In contrast to the conduction band, the valence subbands have much more complicated states. Here since the upper states are p-type, the spin-orbit interaction removes the six-fold degeneracy of the uppermost states. In bulk structure this leaves a four-fold degenerate state at the band-edge and a two-fold degenerate one (spin-orbit split subbands) at lower energy. In QW's, however, because of the carrier confinement, the four-fold degenerate subbands split into to two two-fold degenerate subbands. Here neither orbital angular momentum ($L$) nor spin ($S$) are conserved, but the total angular momentum ($J = L + S$) is conserved. The spin-orbit split states are associated with $J_z = \pm 1/2$ ($z$ components of $J=1/2$). The uppermost subbands, however, are associated with $J_z = \pm 3/2$ and $J_z = \pm 1/2$ ($z$ components of $J=3/2$). Here the subbands with $J_z = \pm 3/2$ have less curvature compared to those with $J_z = \pm 1/2$. Therefore the former are called heavy hole (HH) and the latter light hole (LH) subbands. The valence subbands are not parabolic. This is because except for $k=0$ where they have either $J_z = \pm 3/2$ or $J_z = \pm 1/2$ components, states with different $J_z$'s are mixed. Therefore attributing the valence subbands to either HH or LH subbands is rather problematic. Here we designate each valence subband by considering its nature at $k=0$. For example, HH1 refers to the first valence subband which at $k=0$ has a single angular momentum component ($J_z = \pm 3/2$). At $k \neq 0$, however, this subband is mixed with LH ($J_z = \pm 1/2$). The same is correct for LH1, HH2, LH2, and the other valence subbands. Note that the spin-orbit split subbands are relatively far from the band-edge. Therefore we do not consider them in our analysis.

It is convenient to work in a basis where spin-orbit interaction is diagonal. Using the notation $|J = \frac{3}{2}, J_z \rangle$, this basis is [58]:

$$
\begin{align*}
|\frac{3}{2}, \frac{3}{2} \rangle &= -\frac{1}{\sqrt{2}}|(X + iY) \uparrow\rangle, \\
|\frac{3}{2}, -\frac{1}{2} \rangle &= \frac{1}{\sqrt{6}}|(X - iY) \uparrow\rangle + \sqrt{\frac{2}{3}}|Z \downarrow\rangle,
\end{align*}
$$

(5.3) (5.4)
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\[ |\frac{3}{2}, \frac{1}{2} \rangle = -\frac{1}{\sqrt{6}} |(X + iY) \downarrow \rangle + \sqrt{\frac{2}{3}} |Z \uparrow \rangle, \]  
\[ |\frac{3}{2}, -\frac{3}{2} \rangle = \frac{1}{\sqrt{2}} |(X - iY) \downarrow \rangle. \]

Here arrows refer to the spin states and \(|X \rangle, |Y \rangle\) and \(|Z \rangle\) to the Bloch functions of the uppermost states of the valence band at \(k=0\). These functions have the symmetries of the atomic \(p_x, p_y\) and \(p_z\) orbitals. In the following we designate each \(|J, J_z \rangle\) with \(U_\nu\) where \(\nu\)'s stand for \(J_z\)'s.

Using \(U_\nu\)'s one can expand the valence band wave functions according to:

\[ \psi_{jk}(r) = e^{ik \cdot r} \sum_\nu v^\nu_{jk}(z) U_\nu. \]  

Here \(j\) refers to the valence subband index (HH1, LH1, etc.) and \(v^\nu_{jk}\) to the envelope function of the valence band states. These functions can be obtained from the following [58]:

\[ \sum_\nu [H_{\nu\nu'}(k, -i\partial/\partial z) + V_v(z)\delta_{\nu\nu'}]v^\nu_{jk}(z_h) = E^\nu_{jk} v^\nu_{jk}(z). \]

Here \(H_{\nu\nu'}\) refers to the valence band Hamiltonian in bulk structures (Lottinger-Kohn Hamiltonian) in which we replaced \(k_z\) with \(-i\partial/\partial z\). \(V_v(z)\) is the valence band confinement potential which like that of the conduction band is a symmetric well in Fig.5.2. \(E^\nu_{jk}\) represents the energy associated with a state in the \(j\)th valence subband with wave-vector \(k\). Note that under \(z \to -z\) operation \(v^\nu_{jk}(z)'s\) with \(j = HH1\) and LH1 are even when \(\nu = 3/2\) and \(-1/2\), respectively, and odd when \(\nu = -3/2\) and \(1/2\). The fact that the valence band (or hole) wave functions are written in the form of Eq.(5.7) shows that a state in this band cannot be described by a single \(\nu\).

5.2.3 Excitonic transitions in quantum wells

In an undoped QW when the interband transitions occur at frequencies close to that of the band-edge, the photo-excited electrons and holes have strong Coulomb interaction. For frequencies slightly less than that of the band-edge, this interaction gives rise to bound
states of the electron-hole pairs, or to bound excitons. The sharp features seen in the interband absorption and emission spectra of undoped QW’s are signs of these excitons. The Coulomb interaction plays an effective role even when the interband excitations generate free electron-hole pairs or continua of excitons. This effect, which is seen as the enhancement of the oscillator strengths associated with these transitions, is called Coulomb enhancement effect.

In general the exciton states can be determined from the following:

\[ H\Psi^\sigma = E^\sigma \Psi^\sigma. \]  

(5.9)

Here \( H \) is the total Hamiltonian of the QW in the absence of any optical field (Eq. (5.31)), and \( \Psi^\sigma \) and \( E^\sigma \) are, respectively, the eigenstates and eigenvalues of the QW or those of excitons. \( \sigma \) is the index which fully specifies the state of an exciton. This will be clarified later. In general one can express \( \Psi^\sigma \) in terms of the electron and hole states as follows:

\[ \Psi^\sigma(r_e, r_h) = \sum_{i,j} \sum_k G^\sigma_{ij}(k) \phi_{i,k}(r_e) \psi_{j,-k}(r_h). \]  

(5.10)

Here \( G^\sigma_{ij}(k) \) refers to the envelope function of an exciton with zero center of mass wave-vector, \( K=0 \). Note that \( K \) is the sum of electrons and holes wave-vectors, \( K = k_e + k_h \). Since in the optical processes considered in this thesis one can ignore the photons’ wave-vectors, we have \( k_e = k \) and \( k_h = -k \). \( G^\sigma_{ij}(k) \) can be obtained from [59]:

\[ [E^e_{i,k} + E^h_{j,k}]G^\sigma_{ij}(k) - \sum_{j',q \neq 0} V_{jj'ii'}(k,q)G^\sigma_{ij'}(k+q) = E^\sigma_{ij}G^\sigma_{ij}(k), \]  

(5.11)

where \( E^h_{j,k} \) represents the energy of a bare hole in the \( j \)th subband with wave-vector \( k \). The Coulomb term in this equation is given by

\[ V_{jj'ii'}(k,q) = \frac{\epsilon^2}{2\epsilon q} \int dz_e d\bar{z}_h e^{-q\bar{z}_h-z_h} C^*_{ij}(z_e) C_{i'}(z_e) \Phi_{j',k-\bar{q}} \Phi_{j',\bar{k}}(z_h), \]  

(5.12)

where \( \epsilon \) here is the dielectric constant of the crystal.
The Coulomb term in Eq.(5.11) refers to the general case of the electron-hole interaction. Regarding this term the following comments are necessary:

1) In single QW's since the energy spacing between the conduction subbands are relatively large, the electron-hole interactions can not mix the electron subbands. This leads us to the case where we may only include $i = i'$. This means that in such structures each exciton state can be designated by a conduction subband index. In other words, $\sigma \equiv (\sigma', i)$. $\sigma'$ will be determined in the following.

2) Although for wide QW's the conduction subbands can still be well-separated from each other, the hole subbands may not. Because of this, holes can undergo intersubband mixing when they interact with electrons. Under this condition, in Eq. (5.11) we have to include $j \neq j'$, in addition to $j = j'$. This effect is usually referred to as the hole Coulomb mixing effect. For sufficiently narrow QW's one can ignore the hole Coulomb mixing effect [60].

To find out the quantum mechanical features of the excitonic transitions let us consider a polar coordinate system where the in-plane wave-vector $k$ is given by its magnitude $k$ and an angle $\theta$. Then using the angular momentum operator $(J_z)$ properties, we have [61]:

\[ \nu_{j\kappa}^\nu(z_h) = e^{-i\nu\theta} \nu_{j\kappa}^\nu(z_h). \tag{5.13} \]

Also for the exciton envelope function we have:

\[ G_{ij}^{\sigma'}(k) = e^{i\xi \theta} g_{ij}^{\sigma', \beta}(k). \tag{5.14} \]

In Eq. (5.14) $\xi = m + s$ represents the $z$-component of the total angular momentum of the exciton. Here $s$ and $m$ refer the spin and the orbital angular momentum of the exciton. Note that since $\xi$ is a conserved quantity, it can be used to designate an exciton state. In other words, we can put $\sigma' \equiv \xi$ or $\sigma \equiv (\xi, i)$. The envelope functions, $g_{ij}^{\xi, \beta}(k)$, have some similarities with the hydrogen atom wave-functions, $\phi_{nm}$ ($m$ here stands for the orbital angular momentum). Therefore, we can put $\beta \equiv (n, m)$, where $n$ and $m$ are
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The conventional quantum numbers used to label hydrogen atom states (1s, 2s, 2p±, etc.).

Based on the above discussions we can write the exciton states as follows:

\[ \Psi^\sigma = \Psi_i^\xi = \sum_j \Psi^\xi_{ij}, \]

where

\[ \Psi^\xi_{ij} = \sum_\nu F^\xi_\nu U^\nu_i, \]

and

\[ F^\xi_\nu = \int \frac{dk}{(2\pi)^{3/2}} g^{\xi_\nu}(k)e^{ik\vec{p}}C_i(z_e)U_iU^\nu_j(z_h). \]

Here \( \vec{p} = \vec{p}_e - \vec{p}_h \) in which \( \vec{p}_e \) and \( \vec{p}_h \) refer to the electron and hole lateral position vectors. As Eq. (5.16) shows each exciton state can contain four orbital angular momentum components. In other words, for each value of \( \nu \) there is one value for \( \beta \). In the next section we discuss how the selection rules of the interband transitions restrict excitation of these components.

5.3 Selections rules of the interband and intraband excitonic transitions

Most of the previous studies have been devoted to the properties of excitons associated with the interband excitations (interband excitonic transitions) [59]- [61]. In QW's, however, there is another type of excitonic transition. Here electrons are excited from one conduction subband to another while they are strongly correlated with the holes. In other words, they are excited from one exciton state to another (intraband excitonic transitions). The conduction band transitions can occur by an infrared field polarized along the growth direction, \( z \). Our goals in this section are to review the interband excitonic transitions and then investigate the selection rules and quantum mechanical properties of the intraband excitonic transitions.
5.3.1 Interband excitonic transitions

To review the selection rules of the interband excitonic transitions let us consider the following matrix element using Eqs. (5.15)-(5.17) [61]:

\[ < \Psi^\xi_{ij} | \tilde{e} \cdot \mathbf{p} | G > = \sum_j < \Psi^\xi_{ij} | \tilde{e} \cdot \mathbf{p} | G >. \]  

(5.18)

Here

\[ < \Psi^\xi_{ij} | \tilde{e} \cdot \mathbf{p} | G > = \frac{1}{2\pi^3} \sum_\nu < U_i | \tilde{e} \cdot \mathbf{p} | U_\nu > \int dk d\tilde{\rho} g^\xi_{ij}(k) e^{ik \cdot \tilde{\rho}} e^{i(\xi - \nu)\theta} I^\nu_{ij}(k), \]  

(5.19)

and |G> refers to the QW ground state. Also p and \( \tilde{e} \) are the single particle momentum operator and the field polarization, respectively. \( I^\nu_{ij}(k) \) is given by

\[ I^\nu_{ij}(k) = \int dz C_i(z) \nu^\nu_{jk}(z). \]  

(5.20)

To go further and find out the selection rules, we integrate over \( \theta \) (angular part of \( \mathbf{k} \)). To do this we decompose vector \( \tilde{\rho} \) in Eq. (5.19) into \( \rho_{sc} \) (scaler part) and \( \alpha \) (angular part). Using [61]:

\[ \int d\theta e^{ik \rho_{sc} \cos(\theta - \alpha)} e^{i(\xi - \nu)\theta} = e^{i(\xi - \nu)\alpha} \alpha_{\xi - \nu}(k \rho_{sc}) \]  

(5.21)

the selection rules can be found from the following:

\[ < \Psi^\xi_{ij} | \tilde{e} \cdot \mathbf{p} | G > = \frac{1}{2\pi^3} \sum_\nu < U_i | \tilde{e} \cdot \mathbf{p} | U_\nu > \int dk d\rho g^\xi_{ij}(k) \rho_{sc} d\alpha_{\xi - \nu} e^{i(\xi - \nu)\alpha} J_{\xi - \nu}(k \rho_{sc}) I^\nu_{ij}(k). \]  

(5.22)

Note that this equation shows how various features of the electron states (Eq. (5.1)), the hole states (Eq. (5.7)) and the Coulomb interaction between electron and holes play roles in the interband exciton transitions.

Eqs. (5.20)-(5.22) contain everything we need to find out the interband transition selection rules. To start let us first consider the bracket on the right side of Eq. (5.22). This bracket provides the selection rules introduced by the Bloch functions. Depending
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on the interband optical field polarization, it shows if a transition is possible, allowed or forbidden. This can be seen considering Eqs. (5.3)-(5.6) and the following non-zero elements [57],

\[
< S | p_x | X > = < S | p_y | Y > = < S | p_z | Z >
\] (5.23)

For the optical fields with polarizations in the plane of the QW, excitations of both HH and LH excitons are possible. However, optical fields polarized along \( z \) can only excite LH excitons. \( < U_i | \hat{\epsilon} \cdot \mathbf{p} | U_\nu > \) can also be used to predict the spins of the photo-excited electrons. For a circularly polarized light, for example, when holes with \( J_z = \pm 3/2 \) are generated, the photo-excited electrons have \( \pm 1/2 \) spins.

The second source of the selection rules is \( I_{ij}^\nu \) (Eq.(5.20)). Since for any transition this term has to be non-zero, in symmetric QW's both \( C_i(z) \) and \( \psi_{jk}^\nu(z) \) should be either even or odd. The evenness or oddness of \( \psi_{jk}^\nu(z) \), however, depends on two factors, the subband index \( j \) and spin index \( \nu \). As mentioned before, for HH1 and LH1, components of \( \psi_{jk}^\nu(z) \) with \( \nu = 3/2 \) and \( \nu = -1/2 \) are even. However, the same spin components of \( \psi_{jk}^\nu \) are odd when \( j = HH2 \) or \( LH2 \). Note that in asymmetric QW's these restrictions are relaxed to some extend.

The third source of the interband excitonic transition selection rules comes from the integration over \( \alpha \) in Eq.(5.22). Since we do not consider the electron spins this stipulates that an interband transition is possible only if \( \xi = \nu \). This means that when an exciton is generated, from the four spin components of the holes only one of them is excited. It also means that the total angular momentum of the exciton (\( \xi = m + s \)) should be equal to the spin of the excited hole. In other words, \( s = \nu \) and \( m = \xi - \nu = 0 \). This means that in Eq. (5.15) we can make the following substitution

\[
\Psi_{ij}^\xi \rightarrow \Psi_{ij}^{\xi,\beta}
\]

In the absence of the Coulomb hole mixing \( \xi = \nu \) requires excitation of only s-state excitons. In the presence of such a mixing, however, the forbidden states ( \( m = +1 \)
or -1 or $p_+$ or $p_-$ states, etc.) can also be excited. Here an exciton state could be a mixture of both s-state and the forbidden states. Because of the total angular momentum conservation, however, all mixed states should have the same $\xi$. This has been reflected in Eq. (5.15) (see Eq. (5.25), for an example). For the excitations which involve $j = HH_1$, $LH_1$, $HH_2$ and $LH_2$ hole subbands, for example, we consider the facts that: (i) each subband is a mixture of four spins, and (ii) for the interband transitions $\xi = \nu$ has to be satisfied. Therefore if a mixed exciton state has an s-state component associated with LH1, the $\xi$ of this component is -1/2. As a result all other components of this mixed state should also have the same $\xi$ (see Eq. (5.25)). This leads us to the following table for this particular case and other cases which may involve these four hole subbands.

<table>
<thead>
<tr>
<th></th>
<th>$HH_1$</th>
<th>$LH_1$</th>
<th>$HH_2$</th>
<th>$LH_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\nu = 3/2$</td>
<td>$-1/2$</td>
<td>$3/2$</td>
<td>$-3/2$</td>
<td>$1/2$</td>
</tr>
<tr>
<td>$\nu = -1/2$</td>
<td>$1/2$</td>
<td>$5/2$</td>
<td>$-1/2$</td>
<td>$3/2$</td>
</tr>
<tr>
<td>$\nu = -3/2$</td>
<td>$-5/2$</td>
<td>$-7/2$</td>
<td>$-3/2$</td>
<td></td>
</tr>
<tr>
<td>$\nu = 1/2$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 5.1. $\xi$ for the exciton states associated with HH1, LH1, HH2, and LH2.

Note that each column of this table shows the values of $\xi$'s for a specific spin ($\nu$) of a hole subband (HH1, LH1, HH2, or LH2) and different orbital angular momenta of excitons. Using this table we can determine which components of the angular momenta of an exciton can be mixed with those of other excitons (all having the same $\xi$'s). Note that in the above table we only consider even hole envelope functions. One can alternatively choose the odd functions.

To find out how these selection rules come together to make an interband excitonic transition possible, let us first continue our discussion with the case where we can ignore the hole Coulomb mixing and consider excitons associated with the first conduction subband $i = 1$. To specify an exciton state we should choose a value for $\xi$. Consisdering
Table 5.1 this can be done among any value which consistently reflects the orbital angular momenta of the exciton and \( \nu \)'s. As seen in Eq. (5.16), for a given \( \xi \) an exciton state contains four spin components, and therefore four orbital angular momenta. However, because of the \( \xi = \nu \) condition only one of the \( \nu \)'s can satisfy all of the selection rules. This means that the exciton states are all s-types represented by \( \Psi_{ij}^{\xi,ns} \), where \( n \) takes 1, 2, etc. Note that for HH1 excitation, only \( \xi = \nu = 3/2 \) contributes, and for LH1 exciton only \( \xi = \nu = -1/2 \). The LH (HH) component of HH1 (LH1) can not be excited.

In wide QW's the hole Coulomb mixing effect may be significant. To see this let us consider a QW such as GaAs/Ga\(_{0.75}\)Al\(_{0.25}\)As with a 20 nm well width. As shown in Fig. 5.3 this structure contains four conduction subbands and several valence subbands. The energy spacing between HH1 and LH1 is around 5 meV, and that between HH2 and LH1 is less than 3 meV. The energy spacings between the other hole subbands are relatively large. For example that between LH1 and HH3 is larger than 10 meV and that between LH1 and LH2 is more than 20 meV. In this structure the E1-E2 energy separation
is 30 meV. Therefore one can safely ignore the conduction intersubband mixing caused by electron-hole scattering, i.e. \( i = i' \) in Eq.(5.11). For holes, however, the Coulomb coupling between HH1, LH1 and HH2 should be considered. This requires that \( j \) and \( j' \) in Eq. (5.11) run over these subband indices.

Under these conditions, based on Table.5.1, the only components which can contribute to a HH exciton state with \( \xi = 3/2 \) are an s-type state associated with HH1 and a \( p_+ \)-type state associated with LH2. Therefore, we may have:

\[
\Psi_{1}^{3/2} = a\Psi_{1HH1}^{3/2,s} + b\Psi_{1LH2}^{3/2,p+}.
\]

Here \( a \) and \( b \) are the coefficients representing the contributions of these states to \( \Psi_{1}^{3/2} \). Note that the condition of \( \xi = \nu \) is still satisfied. In the first and second terms of Eq.(5.24) \( \nu = 3/2 \) requires some contribution of HH in HH1 and LH2 with \( m=0 \) and \( m = +1 \), respectively. Since LH2 is rather far from HH1 we may drop the second term in this equation.

The situation is rather different for the case of LH excitons, however. To see this consider the exciton state associated with \( \xi = -1/2 \). Using Table.5.1 we may have the following form for this state:

\[
\Psi_{1}^{-1/2} = a\Psi_{1LH1}^{-1/2,s} + b\Psi_{1HH1}^{-1/2,d} + c\Psi_{1HH2}^{-1/2,p+}.
\]

For the structure shown in Fig.5.3, \( a \), \( b \) and \( c \) have been found in Ref. [60] to be 0.82, 0.49, and 0.15, respectively. The energy of \( \Psi_{1}^{-1/2} \) is -1.27 meV from the \( \Psi_{1HH1}^{3/2,s} \) continuum edge [60]. Here \( \xi = \nu \) in LH1 requires excitation of the s-states, in HH1 excitation of \( d_- \) state, and in HH2 that of the \( p_+ \) state. Note that in all of these cases \( \nu = -1/2 \), as required by the interband selection rules.

### 5.3.2 Intraband excitonic transitions

To identify the selection rules of the intraband excitonic transitions, we consider two exciton states, \( \Psi_i^\xi \) and \( \Psi_{i'}^{\xi'} \), associated with electrons in the \( i \)th and \( i' \)th conduction
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Subbands, respectively. The matrix element which gives the basic features of this type of transition is:

\[
< \tilde{\Psi}_f | p_z | \tilde{\Psi}_i > = \sum_{jj'} < \tilde{\Psi}_{ij} | p_z | \tilde{\Psi}_{i'j'} > .
\]  

(5.26)

Here

\[
< \tilde{\Psi}_{ij} | p_z | \tilde{\Psi}_{i'j'} > = \frac{1}{(2\pi)^3} \int dkd\rho d\rho' g_{ij}^{\xi,\beta}(k) g_{ij}^{\xi',\beta'}(k') e^{i(\xi - \nu)\theta - i(\xi' - \nu')\theta'} e^{-i(k - k')\rho} \times
\]

\[
I_{jj'ii'}(k, k') \delta_{\nu\nu'},
\]

(5.27)

where

\[
I_{jj'ii'}(k, k') = \int dz p_z C_i(z) C_{ij}^*(z) v_{j,k}(z) v_{j',k'}(z).
\]

(5.28)

Using Eq.(5-21), similar to the case of the interband selection rules, we have

\[
< \tilde{\Psi}_{ij} | p_z | \tilde{\Psi}_{i'j'} > = \frac{1}{(2\pi)^3} \int dkd\alpha g_{ij}^{\xi,\beta}(k) g_{ij}^{\xi',\beta'}(k) e^{i(\xi - \nu - \nu')} \alpha I_{jj'ii'}(k) \delta_{\nu\nu'} .
\]

(5.29)

After some algebra the term which provides the selection rules of the intraband excitonic transitions is found as follows:

\[
\int dkd\alpha g_{ij}^{\xi,\beta}(k) g_{ij}^{\xi',\beta'}(k) e^{i(\xi - \nu - \nu')} \alpha I_{jj'ii'}(k) \delta_{\nu\nu'} .
\]

(5.30)

Recall that \( \alpha \) is the angular part of \( \rho \equiv (\rho_{sc}, \alpha) \). Considering Eq. (5.30) one can come up with the following rules:

1. \( \delta_{\nu\nu'} \) (which comes from the periodicity of the Bloch functions) requires equal spin indices for the initial and final states (\( \nu = \nu' \)). This means that the transitions are possible only between the HH or between LH exciton states.

2. Integration over \( \alpha \) is non-zero when the total angular momenta of the initial and final states are the same, \( \xi = \xi' \). Considering \( \nu = \nu' \) and \( \xi = \xi' \) we find that the infrared field causes transitions between exciton states with the same orbital angular momenta, \( m = m' \). This means that the transitions are allowed between s-states, or p-states, and so on.
3. Intraband excitonic transitions can occur between states with the same Hydrogenic principle quantum numbers, \( n = n' \) (diagonal transition). They also can occur when \( n \neq n' \) (non-diagonal transition), although weaker. We will return to this subject in chapter 11.

Fig.5.4 shows a schematic diagram of intraband excitonic transitions in narrow QW's where the hole Coulomb mixing effect can be ignored. Here a near resonance infrared field can cause a transition between 1s of E1-HH1 (or \( \Psi_{1,HH1}^{3/2,1s} \)) and 1s of E2-HH1 (\( \Psi_{2HH1}^{3/2,1s} \)) excitons. Also since the binding energies of the E1-HH1 exciton states are assumed to the be the same as those of the E2-HH1 exciton states with the same \( \beta \)'s, the field can simultaneously cause transition between 2s states and between the two continua, etc. Note that when the hole Coulomb mixing is included, the coupling becomes between the mixed states, such as \( \Psi_{1,2HH1}^{-1/2,\beta} \) and \( \Psi_{2HH1}^{-1/2,\beta} \) (see Eq. (5.25)). This will be discussed in Sect.5.6 in more detail.
5.4 Excitonic infrared dressing of quantum well’s with Ξ and Λ configurations

The interband and intraband excitonic transitions discussed in the previous sections were caused by weak optical fields. These fields did not change the intrinsic optical properties of the QW’s. In the remaining parts of this chapter we study how the linear interband excitonic absorption and emission spectra of a QW change in the presence of a strong infrared field polarized along \( z \) and nearly resonant with the intraband excitonic transitions. For the emission case, we also consider a weak interband pump field to generate electron-hole pairs. The frequency of this field is considered to be larger than that of the band-edge. For the absorption spectrum case we only consider a \( Ξ \) configuration. For the emission case, however, both \( Ξ \) and \( Λ \) configurations are discussed.

5.4.1 Equations of Motion

The conduction band of a QW with a large band offset and/or wide well width can support several subbands. Let us consider two of these subbands represented by \( |i,k> \) \( (i=1, \text{ and } 2) \). Also assume that for small wave-vectors the electrons in these subbands have the same effective masses and their dispersions in the plane of the QW are parabolic. Such a QW can also have several valence subbands, \( |j,k>v \).

In the absence of any optical field the Hamiltonian of such a QW can have the following form:

\[
H = H_0 + V. \tag{5.31}
\]

Here \( H_0 \) is the Hamiltonian of the single particle states in the valence and conduction bands, given by:

\[
H_0 = \sum_{i,k} E^v_{i,k} a^\dagger_{i,k} a_{i,k} + \sum_{j,k} E^c_{j,k} a^\dagger_{j,k} a_{j,k}. \tag{5.32}
\]

The operator \( a^\dagger_{i,k} \) creates an electron in a conduction subband which is specified by \( k \) and \( i \), and \( a_{i,k} \) annihilates such an electron. \( a^\dagger_{j,k} \) and \( a_{j,k} \) are the corresponding operators for
electrons in the valence band. These operators are electron operators. One can transfer them into the electron-hole picture by changing $a^\dagger_{jk}$ to $b_{-k}$. Where $b_{jk}$ represents the annihilation operator of a hole in the $j$th subband with the wave-vector $k$.

$V$ in Eq.(5.31) describes the Coulomb interaction between carriers. It generally contains electron-electron, hole-hole and hole-electron interactions. In this study, however, we are interested in the low interband excitation limit where the electron-electron and hole-hole scattering processes can be ignored. Therefore, $V$ takes the following form:

$$V = \sum_{j'k'q} V_{jj'i}(q) a^\dagger_{ik+q} a^\dagger_{jk-q} a_{j'k'} a_{ik}.$$ (5.33)

The Coulomb interaction term $V_{jj'i}(q)$ has the form of Eq.(5.12) after applying the parabolic band approximation. Since here we consider near resonance interaction of a QW with an infrared field, employment of this approximation does not cause significant error. Note also that in Eq. (5.33) the electron intersubband transitions caused by electron-hole scattering are ignored.

Now let us consider the interaction of this system with an infrared field polarized along $z$ and nearly resonant with the $|1, k >_c$ to $|2, k >_c$ transitions. Within the rotating-wave and dipole approximations the interaction Hamiltonian involving the classical infrared field $E(t) = E_0 e^{-i\omega t}$ is given by:

$$H_1 = -\sum_k \{ \mu_{1z} E(t) a^\dagger_{2,k} a_{1,k} + \mu_{2z}^* E^*(t) a^\dagger_{1,k} a_{2,k} \}.$$ (5.34)

Here $\mu_{1z}$ is the electric dipole matrix element for transitions between the conduction subbands which is assumed to be $k$-independent. To obtain the time evolution of this system we use the Heisenberg equation for the density matrix of the system:

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [H + H_1, \rho].$$ (5.35)

After doing some algebra, for each set of the three-subband system (one valence and two conduction subbands) at a given $k$ the equations of motion for each component of the density matrix of the infrared-coupled QW are obtained as follows:
\[
\frac{d}{dt} \rho_{j,j,k} = -2\text{Im}\left\{ \sum_{j'q \neq 0} [V_{jj'}(q)\rho_{jj'}^{k+q} + V_{jj'}(q)\rho_{jj'}^{k-q}] - \Gamma_j(\rho_{jj}^k - 1) + \Gamma_{1j}(1 - \rho_{jj}^k)\rho_{11}^k + \Gamma_{2j}(1 - \rho_{jj}^k)\rho_{22}^k + \lambda_j(k) \right\}
\]

\[
\frac{d}{dt} \rho_{11}^k = -2\text{Im}\sum_{j'q \neq 0} V_{jj'}(q)\rho_{jj'}^{k+q} - i\Omega(\rho_{12}^k - \rho_{21}^k) - \Gamma_1^r \rho_{11}^k - \Gamma_{1j}(1 - \rho_{jj}^k)\rho_{11}^k + \lambda_1(k)
\]

\[
\frac{d}{dt} \rho_{22}^k = -2\text{Im}\sum_{j'q \neq 0} V_{jj'}(q)\rho_{jj'}^{k+q} - i\Omega(\rho_{12}^k - \rho_{21}^k) - \Gamma_2^r \rho_{22}^k - \Gamma_{2j}(1 - \rho_{jj}^k)\rho_{22}^k
\]

\[
\frac{d}{dt} \rho_{1j}^k = \left[ i(E_{1k}^c + E_{j,k}^h) - \gamma_{1j}\right] \rho_{1j}^k + i \sum_{j'q \neq 0} V_{jj'}(q)\rho_{jj'}^{k+q} (\rho_{11}^k - \rho_{jj}^k) + i\Omega \rho_{2j}^k + i \sum_{j'q \neq 0} V_{jj'}(q)\rho_{jj'}^{k+q} \rho_{2j}^k
\]

\[
\frac{d}{dt} \rho_{2j}^k = \left[ i(E_{2k}^c + E_{j,k}^h - \hbar\omega) - \gamma_{2j}\right] \rho_{2j}^k + i \sum_{j'q \neq 0} V_{jj'}(q)\rho_{jj'}^{k+q} (\rho_{22}^k - \rho_{jj}^k) + i\Omega \rho_{1j}^k + i \sum_{j'q \neq 0} V_{jj'}(q)\rho_{jj'}^{k+q} \rho_{1j}^k
\]

\[
\frac{d}{dt} \rho_{12}^k = \left[ -i(E_{1k}^c - E_{2,k}^c - \hbar\omega) - \gamma_{12}\right] \rho_{12}^k + i\Omega(\rho_{22}^k - \rho_{11}^k) + \sum_{j'q \neq 0} \{ V_{jj'}(q)\rho_{jj'}^{k+q} \rho_{1j}^k - V_{jj'}(q)\rho_{jj'}^{k+q} \rho_{1j}^k \}
\]

Here \(\rho_{ii}^k = \rho_{ii}^{k*}\), \(\omega\) is the frequency of the infrared field and \(\Omega = -\mu_{12}E/h\) refers to the Rabi frequency. The terms such as \(\sum_{j'q \neq 0} V_{jj'}(q)\rho_{jj'}^{k+q} (\rho_{ii}^k - \rho_{jj}^k)\) are the Coulomb
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terms responsible for excitons. The other terms involving the Coulomb interaction are the exchange terms. Because of the low interband excitations we ignore them.

In Eq. (5.36) the pump rate \(\lambda_j(k)\) is introduced to represent the rate at which the \(j\)th valence subband is depopulated by an interband optical pump field. Also \(\lambda_1(k)\) shows the rate at which the first conduction subband is populated by this field and by the intersubband and intrasubband transitions of other electrons in states with higher energies. As mentioned before, these terms are only considered for the PL emission of the QW.

In order to consider damping in Eqs. (5.36)-(5.41), various phenomenological scattering rates, \(\Gamma_l\), and dephasing rates, \(\gamma_{lw}\), are introduced (\(l\) and \(l'\) take \(i\) and \(j\)). These rates are introduced for the sake of the generality of these equations. When we transfer these equations into the exciton bases, we attribute to them a new set of damping rates. Briefly, \(\Gamma^\text{nr}_2\) is the non-radiative scattering rate of electrons in \(|2, k >\). This arises mainly from the intersubband LO phonon-electron scattering processes. \(\Gamma^\text{r}_2\) is the decay rate of electrons in this subband due to the radiative process. We recall that such a transition is possible only in asymmetric QW’s. \(\Gamma^\text{nr}_1\) and \(\Gamma^\text{r}_1\) are the first conduction subband \((|1, k >\) non-radiative and radiative scattering rates respectively. Since we assume that \(\Gamma^\text{r}_j\) \((\text{and } \Gamma^\text{nr}_j)\) is non-zero only when the system is in a bound state (band-band emission is ignored), we put \(\delta_{ji}^{k} = 0\) in the corresponding radiative pumping and decay terms in Eqs. (5.36)-(5.38). \(\Gamma_j\) in Eq. (5.36), is the scattering rate of electrons (or holes) in the ground valence subband.

In order to proceed further we transfer each element of the density matrix into an excitonic basis. Before doing this note that Eqs. (5.39) and (5.40) contain two Coulomb terms with different conduction subband indices. Therefore they basically generate two different sets of excitons associated with \(i=1\) and \(i=2\). For these reasons to transform Eqs. (5.39) and (5.40) we consider \(\{G^\text{e,}\beta_{ij}\}\) and \(\{G^\text{e,}\beta_{2j}\}\) bases, respectively. A typical
transformation then has the following form:

\[ \rho_{i\xi}^k = \sum_{\beta} \rho_{i\beta}^{\xi,\beta} G_{ij}^{\xi,\beta}(k). \] (5.42)

Here for Eq. (5.39) we put \( i = 1 \) and for Eq. (5.40) \( i = 2 \). Note that the index \( \xi \) characterizes the type of the bases. These two equations accompanied by Eq.(5.11) play the key roles in understanding the effects of the hole Coulomb mixing in the excitonic infrared coupling of a QW. We will return to this subject in the following sections.

The transferred equations for the diagonal elements of the density matrix in the exciton basis are found as follows:

\[ \frac{d\rho_{11}^0}{dt} = -\frac{1}{\tau_{\alpha}^0} (\rho_{11}^0 - 1) + \frac{1}{\tau_1^1} \rho_{11}^1 + \frac{1}{\tau_2^2} \rho_{22}^2 - \lambda_\alpha, \] (5.43)

\[ \frac{d\rho_{11}^0}{dt} = -i\Omega(\rho_{12}^0 - \rho_{21}^0) - \frac{1}{\tau_1^1} \rho_{11}^1 + \lambda'_\alpha, \] (5.44)

\[ \frac{d\rho_{22}^2}{dt} = -i\Omega(\rho_{12}^2 - \rho_{21}^2) - \frac{1}{\tau_2^2} \rho_{22}^2 - \frac{1}{\tau_2^2} \rho_{22}^2. \] (5.45)

The off-diagonal elements also take the following forms:

\[ \frac{d\rho_{12}^0}{dt} = [-i\Delta - \frac{1}{T_{12}^0}] \rho_{12}^0 + i\Omega(\rho_{22}^0 - \rho_{11}^0), \] (5.46)

\[ \frac{d\rho_{1j}^0}{dt} = [iE_\alpha^1 - \frac{1}{T_{ij}^1}] \rho_{1j}^0 + i\Omega \rho_{1j}^0, \] (5.47)

\[ \frac{d\rho_{2j}^2}{dt} = [i(E_\alpha^2 + \hbar\omega) - \frac{1}{T_{2j}^2}] \rho_{2j}^2 + +i\Omega \rho_{1j}^0. \] (5.48)

For simplicity in the above equations we introduced \( \alpha = (\xi, \beta) \). \( \tau_{\alpha}^i \) and \( \tau_{\alpha'}^{i'} \) are the phenomenological radiative and non-radiative relaxation times of the \( i \)th conduction sub-band electron in the \( \alpha \)th exciton state, respectively. \( T_{ij}^0 \)'s are the polarization dephasing times associated with the excitonic transitions with the corresponding indices. They include contributions of the quasi-elastic exciton-acoustic phonon and inelastic exciton-LO-phonon scattering processes.
\( \lambda'_\alpha \) and \(-\lambda_\alpha\) are the generation rates of the electrons and holes corresponding to the \( \alpha \)th state of E1-HH1 exciton. Like in the case of \( \lambda_1(k) \), \( \lambda'_\alpha \) includes the contribution from the pump field and that of electrons which are relaxed down from the higher conduction subband via non-radiative decay of E2-HH1. In fact in terms of these dynamics we consider in steady state fashion two effective loops both common in consisting of: bound radiative E1-HH1 exciton \( \rightarrow \) bound E2-HH1 exciton \( \rightarrow \) free electron-hole pairs \( \rightarrow \) bound radiative E1-HH1 exciton. One of the loops has leakage due to the radiative decay of bound E1-HH1 excitons (single symmetric QW), and the other has an extra leakage due to the radiative decay of E2-HH1 excitons (single asymmetric QW). These loops are based on the assumption that the infrared field has quasi time-independent amplitude. Note that based on our experimental analysis in chapter 11, there is an additional leakage in each of these loops due to the non-radiative recombination of excitons. However, since this rate may change from one sample to another, and in order to emphasis the coherent effects at this stage, we avoid introduction of such a decay here.

Eqs. (5.43)-(5.48) can be reduced to a linear matrix equation as follows:

\[
\frac{d\Phi^\alpha}{dt} = L^\alpha \Phi^\alpha + K^\alpha, \tag{5.49}
\]

where the \( \Phi^\alpha_i \)'s are:

\[
\begin{align*}
\Phi_1^\alpha &= \rho_{11}^\alpha & \Phi_2^\alpha &= \rho_{12}^\alpha & \Phi_3^\alpha &= \rho_{j_2}^\alpha \\
\Phi_4^\alpha &= \rho_{21}^\alpha & \Phi_5^\alpha &= \rho_{2j}^\alpha & \Phi_6^\alpha &= \rho_{1j}^\alpha \\
\Phi_7^\alpha &= \rho_{jj}^\alpha & \Phi_8^\alpha &= \rho_{j_1}^\alpha & \Phi_9^\alpha &= \rho_{22}^\alpha.
\end{align*}
\tag{5.50-5.52}
\]

5.5 **Interband excitonic absorption spectra of a quantum well in the presence of strong intraband excitonic coupling**

In this section we study the evolution of the excitonic interband absorption spectra of the system discussed in the preceding section. To do this we consider a probe field to detect the system when two of the conduction subbands are coupled by a near resonance
infrared field. The probe field, \( E_p(t) \), is polarized in the quantum well plane. Based on our discussion in section 5.3.1 this has the consequence that both light- and heavy-hole excitons are excited. Here we only consider the transitions which involve the first conduction subband, \( i = 1 \). Under these conditions the interband interaction Hamiltonian of the system has the following form:

\[
H_2 = -\sum_\alpha \sum_j \{ \mu_{ij}^\alpha E_p(t)a_{ij\alpha}^\dagger b_{ij\alpha}^\dagger + \mu_{ij}^{*\alpha} E_p^*(t)b_{ij\alpha}a_{ij\alpha} \}. \tag{5.53}
\]

Here \( b_{ij\alpha} \) and \( a_{ij\alpha} \) are the hole and electron annihilation operators in the excitonic state \( \Psi_{ij}^\alpha \), respectively. \( \mu_{ij}^\alpha \)'s are the shared electric dipole moments for the transitions from the ground to \( \Psi_{ij}^\alpha \) given by [53]:

\[
\mu_{ij}^{\xi,\beta} \sim \int d\rho dk' g_{ij}^{\xi,\beta}(k') e^{i\mathbf{k'}\cdot \mathbf{r}} e^{i(\xi-\beta)\mathbf{k}\cdot \mathbf{r}} \int dz v_{jk'}^\nu(z) C_i(z) < U_i |\mathbf{r} | U_i > . \tag{5.54}
\]

Using the linear response theory discussed in chapter 2 the interband response function of the system can be obtained in terms of the unequal-time commutations of the system polarization, \( < [p_{ij\alpha}(t), p_{ij\alpha}(t')] > \), in which \( t > t' \). Here \( p_{ij\alpha}^- \) and \( p_{ij\alpha}^+ \) are the positive and negative frequency parts of the polarization associated with \( \Psi_{ij}^\alpha \). They have the following explicit forms

\[
p_{ij\alpha}^-(t) = \mu_{ij}^\alpha a_{ij\alpha}^\dagger b_{ij\alpha}, \tag{5.55}
\]

and \( p_{ij\alpha}^+(t) = [p_{ij\alpha}^-(t)]^* \).

The absorption spectrum of the weak interband field in the absence of the hole Coulomb mixing can be calculated using the real part of the following expression:

\[
\sum_\beta \sum_j < [\tilde{p}_{ij\xi\beta}(t'), \tilde{p}_{ij\xi\beta}(t', z)] > |_{z=i\omega_p}. \tag{5.56}
\]

Here \( \omega_p \) refers to the weak interband field frequency. Here the sum over \( \beta \) includes only the s-states (1s, 2s, ...). Since in the absence of the hole Coulomb coupling \( \xi \)'s are correlated with \( j \)'s, there is no sum over \( \xi \). In fact here the values of \( \beta \) and \( j \) impose certain values on \( \xi \). This condition is not satisfied when the hole Coulomb mixing is included. Here one has to sum over \( \xi \) independently.
In order to calculate Eq. (5.56) we should calculate $< [p_{ij\alpha}^+(t'), \tilde{p}_{ij\alpha}^-(t', z)] >$. Taking steps similar to those in chapter 2, the following result was obtained

$$< [p_{ij\alpha}(z), p_{ij\alpha}^+(\infty)] > = |\mu_{ij}|^2 \{ R_{88}^0(z) \Phi_2^\alpha(\infty) + R_{88}^0(z) [\Phi_1^\alpha(\infty) - \Phi_1^\alpha(\infty)] \}. \quad (5.57)$$

The absorptive and refractive spectra of the QW can now be obtained by the substitution of Eq. (5.57) in Eq. (5.56).

Note that the process of infrared coupling depends on the interband field (probe) photon energy. For example, in narrow QW's when we monitor the system at the vicinity of the 1s E1-HH1 exciton ($\Psi_{1HH1}^{3/2,1s}$), the holes are mostly generated in the first valence band, but the electrons are shared between the two conduction subbands. Therefore the coupling process can be between 1s states of E1-HH1 and E2-HH1 ($\Psi_{2HH1}^{3/2,1s}$) excitons (see Fig.5.5a). However, when we detect the a light-hole exciton, the coupling is between exciton states associated with E1-LH1 ($\Psi_{1LH1}^{-1/2,1s}$) and E2-LH1 ($\Psi_{2LH1}^{-1/2,1s}$) (Fig.5.5b).
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5.6 Absorption spectra of wide quantum wells, hole Coulomb mixing effects in the excitonic intraband dressing

In narrow QW’s because the hole subbands are well separated from each other, the exciton states are either heavy- or light-hole in nature. Under this condition the hole Coulomb interaction can be ignored and one can ascribe each absorption peak to a specific spin index \( \nu \). Therefore the infrared field couples pure states according to the selection rules discussed in section 5.3.2.

As mentioned before, however, the hole Coulomb mixing could play a role in wide QW’s. To show this in the excitonic intraband dressing of QW’s we consider the structure shown in Fig. 5.3. Here, since HH1 and LH2 are relatively far from each other, we ignore mixing of the HH exciton associated with \( \xi = 3/2 \) \((b = 0 \text{ in Eq. (5.24)})\). For the LH exciton with \( \xi = -1/2 \), where this effect is significant, we use Eq. (5.25). Based on Ref. [60] in this structure the shared oscillator strengths for each component of \( \Psi_{1}^{-1/2} \) \((\Psi_{1HH1}^{-1/2,s}, \Psi_{1HH1}^{-1/2,d-}, \text{and } \Psi_{1HH2}^{-1/2,p^+})\) are approximately \( 4 \times 10^{-5} \text{ A}^{-2} \), \( 0.5 \times 10^{-5} \text{ A}^{-2} \), and \( 1.4 \times 10^{-5} \text{ A}^{-2} \), respectively. The energy spacing between the first and fourth conduction subbands is 143 meV.

The absorption spectrum of the structure in the absence of the infrared field is shown in Fig. 5.6. Here the solid line shows the total absorption spectrum of the QW. To clarify how different components of the exciton contribute to this spectrum we also show the contribution of the HH1 (dashed line), LH1 (dotted line), and HH2 (dashed-dotted line) exciton states separately. As Fig. 5.6 shows since we do not consider any Coulomb mixing for the E1-HH1 exciton, none of the forbidden states \( (m = \pm 1, \pm 2...) \) contribute to this state. As expected, the contribution the allowed transition to \( \Psi_{1HH1}^{-1/2,s} \) in the high energy peak is high. Here, however, \( \Psi_{1HH1}^{-1/2,d-} \) and \( \Psi_{1HH2}^{-1/2,p^+} \) also contribute. Although that of the former is more than that of the latter. For the range of energy we consider, the continuum mainly arises from the E1-HH1 exciton.
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Figure 5.6: Linear interband response of a 20 nm GaAs/Al_{0.3}Ga_{0.7}As quantum well to an in-plane polarized laser. The dashed line represents $\Psi_{1HH_1}^{3/2,1s}$ and $\Psi_{1HH_1}^{-1/2,d-}$ contributions, the dotted line that of $\Psi_{1LH_1}^{-1/2,s}$, and the dashed-dotted line that of $\Psi_{1HH_1}^{-1/2,p+}$. The solid line shows the sum of all these contributions including that of the $\Psi_{1HH_1}^{3/2,s}$. $E_p$ refers to the probe photon energy and $E_{e1}$ to that of the exciton energy from E1 subband energy.
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Now let us consider interaction of this QW with an infrared field resonant with the E1-E4 transition or the excitonic transitions between $\Psi_1^f$ and $\Psi_4^f$. Since the E4 is very close to the conduction band edge, it is broad. For this reason we consider the phenomenological dephasing rates of exciton states corresponding to E4-HH1, E4-LH1, and E4-HH2 to be 10 meV. The dephasing rates corresponding to the exciton states associated with E1 are considered to be 1 meV.

The results of calculations when the infrared field is resonant with the transitions between $\Psi_{1HH1}^{3/2,1s}$ and $\Psi_{4HH1}^{3/2,1s}$, and between $\Psi_1^{-1/2}$ and $\Psi_4^{-1/2}$ are shown in Fig.5.7. Based on the selection rules obtained in section 5.3.2 this causes a simultaneous coupling of the corresponding components of these states. In other words, $\Psi_{1HH1}^{\xi,1s}$ with $\Psi_{4HH1}^{\xi,1s}$; $\Psi_{1HH1}^{\xi,d-}$ with $\Psi_{4HH1}^{\xi,d-}$; and $\Psi_{1HH2}^{\xi,p+}$ with $\Psi_{4HH2}^{\xi,p+}$ are coupled. Here the net effect of the infrared coupling process is seen as broadening of the exciton peaks. For high field intensity it may be possible to detect a doublet associated with the coupling of $\Psi_{1HH1}^{3/2,1s}$ and $\Psi_{4HH1}^{3/2,1s}$ excitons. However, this doublet could be very broad and in practice one might only see a pure broadening of the exciton peaks.

5.7 Intraband excitonic dressing effects on the emission spectra of interband excitons

An issue which is of great importance for both fundamental physics and practical applications is the alteration of the photoluminescence (PL) spectrum of a QW in frequency and/or intensity. This can be achieved by applying an electric field to the QW [62], or by enhancing or inhibiting spontaneous emission using optical microcavities [63]. Another method of changing the PL spectrum is to use optical fields. This process depends strongly on the polarization of the optical field. For intense far infrared fields polarized in the QW plane the effect has been found to be quenching and red shifting of the PL spectra [64]. If the optical field is polarized along the QW growth direction, however, the intraband excitonic coupling discussed earlier in this chapter can occur. In this section
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Figure 5.7: Absorption spectrum of a quantum well similar to that in Fig. 5.6 in the presence of a resonant infrared field. Here the solid, dashed, and dotted lines refer, respectively, to the total absorption when $\Omega = 0$, $\Omega = 2$ ps$^{-1}$, and $\Omega = 10$ ps$^{-1}$.

we theoretically discuss how such a coupling could change the PL spectra of a QW. In chapter 11 we extend this investigation to more complex systems by introducing non-radiative decay mechanism and multi-state coupling of excitons. In the present section we also theoretically address some of the experimental results reported earlier [67]. The results of this section have been published in Ref. [30].

Note that in general the decay process in QW's depends on the carrier-carrier and carrier-phonon scattering processes, QW layer roughness (interface fluctuation), and other effects. However, since here we are looking for the linear PL spectra at low interband excitation, the carrier-carrier scattering effects are ignored. In fact, we are dealing with the emission spectra of uncorrelated single excitons (intrinsic case). In the present system these excitons are optically mixed to another set of excitons by an IR field.

Under these conditions the linear PL spectrum caused by the decay of the $\Psi_{1HH1}^a$ and $\Psi_{2HH1}^a$ bound states can be found in terms of the two-time correlations of the system polarization, $< p_{j\alpha}^{-}(t)p_{j\alpha}^{+}(t') >$ in which $t > t'$. Here $p_{j\alpha}^{-}$ and $p_{j\alpha}^{+}$ are the negative
and positive frequency parts of the polarization associated with the αth states of these excitonic sets and \( j \) refers to \( HH1 \). The negative part is given by

\[
p_{ja}(t) = \mu_{1j}^a a_{1ja}^\dagger b_{1ja} + \mu_{2j}^a a_{2ja}^\dagger b_{2ja}.
\]

The second term here refers to the nondiagonal transition (E2-HH1 emission). The result of the calculations is:

\[
<p_-(z)p_+(\infty) > = |\mu_{1HH1}^\alpha|^2 \left[ R_{33}^\alpha(z)\Phi_3^\alpha(\infty) + R_{87}^\alpha(z)\Phi_6^\alpha(\infty) + R_{68}^\alpha(z)\Phi_1(\infty) \right] + \\
|\mu_{2HH1}^\alpha|^2 \left[ R_{33}^\alpha(z')\Phi_9^\alpha(\infty) + R_{37}^\alpha(z')\Phi_5^\alpha(\infty) + \\
R_{38}^\alpha(z')\Phi_4^\alpha(\infty) \right],
\]

where \( z' = z - i\omega \).

Here we consider narrow QW's, therefore the hole Coulomb mixing effects are ignored. Also due to the fact that the oscillator strengths of the excited states of excitons are decreased by \((n - \frac{1}{2})^{-3}\) and because of low temperature and low intensity pump we only consider emission of 1s states [65]. This means that one can refer only to the excitonic index \( \beta = 1s \) (recall \( \alpha \equiv (\xi, \beta) \)). Here also \( \xi = 3/2 \) which are dropped in the following for simplicity. We consider the structures to be inhomogeneously broadened. For this reason the homogeneous line width \((1/T_{1j}^{s} \text{ in Eq. (5.47)})\) is considered to be 0.6 meV. This includes both the acoustic phonon and layer interface fluctuation quasi-elastic scattering processes with bound excitons [66]. We also follow the standard method for dealing with such cases by considering the exciton emission spectrum as a convolution of a Gaussian line-shape with homogeneous emission spectra.

### 5.7.1 PL spectrum of IR-coupled QW's with Ξ configurations, diagonal E1-HH1 emission

In this subsection we study the PL spectrum of a symmetric single QW coupled by an IR field. Here the parities of the subbands are well-defined, and therefore, only the E1-HH1
exciton can decay radiatively ($\frac{1}{\tau^2} = 0$ in Eq. (5.45)). We start our investigation with a GaAs/ Ga$_{0.7}$Al$_{0.3}$As QW structure having a width of the order of 7 nm. In such a structure the first and second bound conduction subbands are separated from each other by $\sim$ 132 meV, close to the CO$_2$ laser photon energies. Also the binding energy of the 1s state of E1-HH1 is assumed to be 10 meV. We consider $\frac{1}{\tau^2} = 5$ ps$^{-1}$ and $\mu_{12} = e \times 2$ nm. Taking this value for the dipole moment, we estimate the IR intensity as $I=0.06 \times \Omega^2$ MW/cm$^2$ ($\Omega$ in ps$^{-1}$). For this structure we consider the FWHM of the Gaussian line-shape to be 4 meV. Also note that since here the E1-HH1 exciton decay is detected, the system has a $\Xi$ configuration similar to that shown in Fig.2.2a.

Fig.5.8 shows the way in which the linear PL spectrum changes when the IR field is resonant with the E1-HH1 to E2-HH1 transitions. The initial effect of the IR field is seen to be broadening of the PL line (dashed line). By further increasing the IR intensity the spectrum develops a doublet structure centered at the uncoupled central emission frequency (dotted line). The doubling becomes magnified by a further increase in the IR intensity (dashed-dotted line).

5.7.2 PL emission of IR-coupled QW’s with $\Lambda$ configurations, nondiagonal E2-HH1 emission

In this subsection we study the nondiagonal PL spectrum in asymmetric QW structures. In such structures the interband transition of electrons in E2 to HH1 is possible to some extent ($\frac{1}{\tau^2} \neq 0$). Therefore, we expect to observe the radiative recombination of E2-HH1 excitons once the IR field is turned on. Here we apply our formalism to explain the experiment done in Ref. [67] and discuss and predict more interesting phenomena. Note that since E2-HH1 emission is detected here the system now has a $\Lambda$ configuration such as that shown in Fig.2.2b.

To investigate the experiment we use the parameters suggested in Ref. [67] for the GaAl/ Al$_{0.3}$Ga$_{0.7}$As structure. This means that we consider the ratio of the emission
Figure 5.8: 1s PL spectrum of E1-HH1 exciton in an IR-coupled QW with a resonant Ξ configuration. The solid line corresponds to $\Omega = 0$, the dashed line to $\Omega = 1 \text{ ps}^{-1}$, the dotted line to $\Omega = 2 \text{ ps}^{-1}$, and the dashed-dotted line to $\Omega = 10 \text{ ps}^{-1}$. $E_r$ and $E_{e1}$ refer to energies of the emitted photons and that between the first conduction and valence subbands respectively. Here $\frac{1}{\tau_{j1}} \sim 1 \text{ ps}^{-1}$ and $\frac{1}{\tau_{j2}} \sim 6 \text{ ps}^{-1}$. 
oscillator strengths of E2-HH1 to E1-HH1 ($\frac{f_2}{f_1}$) to be equal to 0.024, and the IR field to be resonant with transition between these two states. Also we consider $\tau_2^{\sigma'}=2.5$ ps, as obtained in Ref. [67], and the FWHM of the Gaussian function equal to 1.5 meV. The theory is applied such that $\mu_{12}$ is considered to be a fitting parameter. Consistent with our theory, the experiment reference has been done in low temperature and the interband laser intensity was chosen to be low.

Calculation of the real part of Eq. (5.59) for such a structure leads to the PL spectra of E2-HH1 as shown in Fig.5.9. One can see that for $\Omega = 0$ ($I=0$) there exists no emission from E2-HH1 exciton recombination (since no carrier exists in the second conduction subband). But when we increase the laser intensity to $\Omega = 0.08$ ps$^{-1}$ (3.2 kW/cm$^2$) we see emission of E2-HH1 excitons (solid line). Further increase in the field intensity causes further increase in the intensity of such a process (see dashed line for $I=13$ kW/cm$^2$ with $\Omega = 0.16$ ps$^{-1}$). The interesting result here resides in the ratio of the intensity of E2-HH1 transition ($I_2$) to that of the E1-HH1 transition ($I_1$) when they are simultaneously measured. Based on the experiment for an intensity of the IR field around $\sim 15$ kW/cm$^2$, this ratio ($I_2/I_1$) should be $\sim 0.001$. Using the inset of Fig.5.9 which shows the intensity of E1-HH1 ($I_1$) simultaneous to the emission of E2-HH1, we find this ratio similar to that of the experiment. Such a consistency between the theory and experiment was found by putting $\mu_{12}=e \times 0.7$ nm ($I=500 \times \Omega^2$ kW/cm$^2$). One may find this value for the dipole moment a little small compared to those measured previously in symmetric QW. This can, however, be related to the fact that the quantum well here is asymmetric. Note that in Ref. [67] a cw CO$_2$ laser with low intensity was used. As will be shown in chapter 11, such fields do not cause any significant quenching. Therefore, although here we have not included the non-radiative decay of the exciton the results are found to be consistent with those obtained in the experiment. Also here we considered the pump rate to be small such that the linear response approximation remained valid. Therefore the value of this rate did not play any dynamic role in the emission spectra.
Figure 5.9: The nondiagonal exciton recombination of E2-HH1 in an IR-coupled QW with a resonant A configuration. The solid line corresponds to emission at $\Omega=0.08$ ps$^{-1}$ and the dashed line to $\Omega=0.16$ ps$^{-1}$. The inset shows the corresponding E1-HH1 emission. Here $\frac{1}{T_{ij}} \sim 1$ ps$^{-1}$ and $\frac{1}{T_{ij}} \sim 1.5$ ps$^{-1}$.

The results presented in Fig.5.9 were limited to the intensities tested in the experiment. Now let us go forward and study the E2-HH1 radiation dynamics at higher IR intensities keeping other parameters the same as those of Fig.5.9. One can see in Fig.5.10 that increasing the field intensity causes a further increase in the peak amplitude of the emitted light of E2-HH1 (solid line). However, a further increase in the IR intensity causes development of an “emission hole” or dip at the line-center frequency. This is accompanied by a further increase of the emission intensity (dashed line). Meanwhile the “emission hole” becomes deeper while the emission peak amplitudes reach a saturated limit (dotted line). Finally when we apply high field intensities the “emission hole” reaches a minimum and becomes virtually a dark line. At this condition further increase in the IR intensity causes further separation of the symmetric peaks of emission with no change in their shapes.
5.7.3 Damping Rate Effects and Characteristic Limits

The evolutions of PL spectra studied in the present section revealed two different kinds of dynamics. The first type which happens both in E1-HH1 and E2-HH1 exciton emissions is a measure of the quantum interfering processes. They can be characterized in terms of the damping rates of the excitons and are scaled by the Rabi frequency. Based on our calculations this kind of dynamic falls into three characteristic limits. When we detect the E1-HH1 exciton emission ($\Xi$ configuration) these limits are: (i) $\Omega \ll \frac{1}{T_{2j}}$ (the low-field limit) in which the coupling effect is seen as broadening of the E1-HH1 PL spectrum, (ii) $\Omega \sim \frac{1}{T_{2j}}$, where the spectrum starts to develop a doublet, and (iii) $\Omega \gg \frac{1}{T_{2j}}$ or the high-field limit where the doublet becomes well resolved. Any further increase in the IR intensity beyond the high-field limit only causes more separation of the doublet peaks.

A similar dynamic can also be obtained in the evolution of the E2-HH1 exciton decay. But, such a dynamic is convoluted by a second type of dynamic caused by the rate in
which E2-HH1 excitons are generated. This rate which only is present in the E2-HH1 emission decay depends on the damping rate of these excitons \( \frac{1}{T_{j2}} \) and \( \Omega \). Therefore in contrast to the E1-HH1 emission which was characterized by a single parameter \( \frac{1}{T_{j1}} \), emission of E2-HH1 should be characterized by two parameters, \( \frac{1}{T_{j1}} \) and \( \frac{1}{T_{j2}} \). The former parameter provides the same information as \( \frac{1}{T_{j1}} \) did for E1-HH1. Considering Fig.5.10 it gives the limits in which E2-HH1 spectrum develops as a growing single peak \( \Omega << \frac{1}{T_{j1}} \), solid line ), or starts to develop a dip or “emission hole” \( \Omega \sim \frac{1}{T_{j1}} \), dashed line ), or generates a well-resolved doublet or dark line \( \Omega >> \frac{1}{T_{j1}} \), dashed-dotted line). On the other hand, the amount of increase of the emission intensity of this state is characterized by \( \frac{1}{T_{j2}} \). If \( \Omega << \frac{1}{T_{j2}} \), any increase of \( \Omega \) generates a linear increase in the E2-HH1 exciton emission, for \( \Omega \sim \frac{1}{T_{j2}} \) this reaches a saturated limit, and at the condition \( \Omega >> \frac{1}{T_{j2}} \) it becomes constant.

We also note that the relative magnitudes of \( \frac{1}{T_{j1}} \) and \( \frac{1}{T_{j2}} \) are important in the evolution of the E1-HH1 and E2-HH1 emission spectra. In QW’s, since \( \frac{1}{T_{j1}} > \frac{1}{T_{j2}} \) the basic development of the E1-HH1 emission should be mostly broadening. The corresponding dynamics of the E2-HH1 emission, however, is the development of a single growing spectrum followed by an “emission hole”. In other words, the conditions \( \Omega < \frac{1}{T_{j2}} \) and \( \Omega \sim \frac{1}{T_{j1}} \) are respectively likely regimes for the E1-HH1 and E2-HH1 evolution. This subject would be revealed to some extend in our experimental analyses in chapter 11.

5.7.4 Structuring processes in the exciton emission spectra

Two interesting features seen above were broadening followed by doubling of the E1-HH1 emission spectrum and generation of an emission hole followed by a dark line in that of E2-HH1. These features can be related to the dark line effect previously discussed in atomic systems with \( \Xi \) or \( \Lambda \) configuration in chapter 3. Comparing the \( \Xi \) system discussed in this chapter and that in chapter 3, one finds out that the metastable state in the former was substituted by a relatively broad state in the latter. Therefore as shown in Fig.5.8,
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the effect became broadening of the emission spectrum followed by the resolution of a doublet (Autler-Townes doublet) [68]. These processes can also be interpreted in terms of quantum interferences between various transition paths of the system, similar to that in chapter 3.

It is, however, more transparent to study these quantum interferences in the nondiagonal PL emission spectra of the Λ system (Fig.5.10). To do this we use our structuring model explained in chapters 3 and 4. Based on this model the emission dynamics of each discrete state of E2-HH1 can be considered as if it is structured by the optical embedding of a discrete E1-HH1 state into its line-center vicinity, similar to LICS. In fact here the relatively narrow E1-HH1 acts as a bound state and the broad discrete state of E2-HH1 as an “effective continuum”. To see this clearly we consider the evolution of non-diagonal emission of excitons considering two discrete E1-HH1 and E2-HH1 states with \( \frac{1}{\tau_{ij}} = 0.001 \) ps\(^{-1}\) (unrealistic for quantum wells) and \( \frac{1}{\tau_{ij}} = 5 \) ps\(^{-1}\). This has the benefit of showing the effects of damping rates and establishing a close correspondence to our discussions in chapters 3 and 4. The effects of actual damping rates of excitons enter as resolution parameters and cause line broadening in the results obtained at this limit of damping rates. Also the case of the inhomogeneous broadening effect can be obtained by speculating the results of the resonant and off-resonant coupling cases discussed in this subsection.

Under these conditions the results of detection of a single E2-HH1 discrete state PL spectrum are shown in Fig.5.11. Here we see that the PL spectrum from its early development has a dark line. This is because \( \Omega \) reaches the limit of \( \Omega >> \frac{1}{\tau_{ij}} \) very early. Therefore, the phase of the dynamic “emission hole” is virtually skipped. Any increase in the field intensity makes the dark line wider and their corresponding peaks stronger and shifts them farther away from each other. Since at resonant coupling the photon energy of the laser is at the line peak of the E2-HH1 spectrum, the symmetry seen by the coupling field requires the induced structure to be the generation of a “hole” within the “effective continuum”. The inset of Fig.5.11 shows this in more detail at the earlier
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Figure 5.11: The nondiagonal exciton recombination of a discrete E2-HH1 exciton in the presence of a resonant IR field. The solid line corresponds to $\Omega = 1$, dashed line to $\Omega = 3$ ps$^{-1}$, and dotted line to $\Omega = 8$ ps$^{-1}$. $E_2$ refers to the energy of the bound 1s E2-HH1 exciton and $\frac{1}{\tau_{11}} = 0.001$ ps$^{-1}$ and $\frac{1}{\tau_{12}} = 5$ ps$^{-1}$. The inset shows a similar process on the earlier stage of coupling. The solid line here corresponds to $\Omega = 0.1$, dashed line to $\Omega = 0.2$ ps$^{-1}$, and dotted line to $\Omega = 0.3$ ps$^{-1}$.

...stages of the coupling process at the vicinity of the line-center frequency. In fact within the low-field limit any increase in the field intensity increases the magnitude of emission intensity while the dark line remains Lorentzian and becomes wider and deeper. This is a characteristic feature of a symmetrically structuring process. At this limit the “effective continuum” acts as the flat continuum of LICS [52].

To describe the effect of detuned coupling we consider a case where $\Delta = +3$ meV. The result of the detection of the E2-HH1 PL spectrum with the same parameters as those in Fig.5.11 is shown in Fig.5.12. In contrast to Fig.5.11, we see here the effect of the IR field as development of an asymmetric spectrum. For low IR intensities the spectrum includes an essential spectrum with a narrow structure at its tail (solid line). Increasing the IR intensity makes the essential peak and structure larger in amplitudes and shifts them away from each other. This process is accompanied by narrowing the main peak...
Figure 5.12: 1s PL spectrum of a discrete E2-HH1 exciton in the presence of an IR field with $\Delta = +3$ meV. All other specifications are the same as in Fig.5.11. The inset shows the structured parts of the emission spectra at an earlier stage of the coupling process. The solid line here corresponds to $\Omega = 0.1$, dashed line to $\Omega = 0.2 \text{ ps}^{-1}$, and the dotted line to $\Omega = 0.3 \text{ ps}^{-1}$.

and broadening the structure until they become identical and Lorentzian at the high-field limit. In the inset of Fig.5.12 we show the evolution of the structured part of Fig.5.12 at the early stage of its evolution. This shows an asymmetric coupling between these two states, as the the IR field now sees the E2-HH1 spectrum asymmetrically. Increasing the IR intensity causes an increase in the amplitude of the structure emission accompanied by a definite amount of optical Stark shift. Note that here one can use similar paths as those in chapter 3 to discuss quantum interferences.
Chapter 6

Multi-subband mixing of excitons in quantum wells

6.1 Introduction

The discussion of the intraband exciton coupling presented in the preceding chapter dealt with two sets of exciton states coupled by a nearly resonant infrared field. In order to make such a two-level model valid, we paid particular attention to the band structure of the QW. This assumption may not be valid for some other structures. For example, using a structure consisting of 8.5 nm GaAs wells and Al$_{0.3}$Ga$_{0.7}$As barriers, Fröhlich et al. reported a peculiar modification of the interband absorption spectrum [11]. This occurred when a CO$_2$ laser was used to couple the ground and first conduction subbands off-resonantly. They found that employment of a two-level coupling model (the $\Xi$ system) leaves these results unexplained. In the present chapter we explain the reason of this failure by presenting the results of a new model which are in much better agreement with the experiment. This model is based on the multi-subband excitonic coupling processes. For the case of the 8.5 nm wide GaAs QW for example, since the E1-E2 and E2-E3 transition energies are $\sim$112 and 140 meV [77], such processes are caused by mixing of the three sets of excitons associated with these subbands. Fig.6.1 shows the schematic diagram of such a QW structure. Note that here E3 is a quasi-bound subband [77]. Also since the QW is narrow we ignored the effects of the hole Coulomb mixing. The results of this chapter have been published in Ref. [25].
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Figure 6.1: Schematic diagram of the energy levels of a GaAs/Al$_{0.3}$Ga$_{0.7}$As quantum well structure with 8.5 nm well width.
6.2 Excitonic Infrared coupling of a quantum well with a $\Xi_-$ configuration

We are interested in the multi-subband excitonic mixing processes caused by a single infrared field polarized along the growth direction. For this reason we consider a QW structure such as that shown in Fig. 6.1. As mentioned in the introduction the energy separations between the three conduction subbands are such that they can be coupled by a single infrared field, forming a detuned $\Xi_-$ configuration (Fig. 2.3a). Under these conditions the infrared-QW interaction can be presented by

$$H_1 = - \sum_k \{ \mu_{12} E(t) a^\dagger_{2,k} a_{1,k} + \mu_{23} E(t) a^\dagger_{3,k} a_{2,k} + H.C. \}. \quad (6.1)$$

Here $\mu_{12}$ and $\mu_{23}$ are the electric dipole matrix elements of transitions between the conduction subbands.

Taking steps similar to those in chapters 2 and 5, the (complex) near bandgap linear interband response of the IR coupled system is found as follows:

$$\Gamma(i\omega_p) = \sum_{\beta;j=HH1,LH1} |\mu_{1j}|^2 \{ R^{j\alpha}_{10,9}(i\omega_p) \Phi_{12}^{j\alpha}(\infty) + R^{j\alpha}_{10,10}(i\omega_p) \Phi_{7}^{j\alpha}(\infty)$$

$$+ R^{j\alpha}_{10,11}(i\omega_p) \Phi_{3}^{j\alpha}(\infty) + R^{j\alpha}_{10,13}(i\omega_p) \Phi_{8}^{j\alpha}(\infty) - R^{j\alpha}_{10,5}(i\omega_p) \Phi_{14}^{j\alpha}(\infty)$$

$$- R^{j\alpha}_{10,6}(i\omega_p) \Phi_{15}^{j\alpha}(\infty) - R^{j\alpha}_{10,7}(i\alpha) \Phi_{12}^{j\alpha}(\infty) - R^{j\alpha}_{10,10}(i\omega_p) \Phi_{8}^{j\alpha}(\infty) \} \quad (6.2)$$

Here $R^{j\alpha}_{ml}(i\omega_p)$ are the elements of the matrix $R^{j\alpha}(i\omega_p) = (i\omega_p I - L^{j\alpha})^{-1}$ in which $I$ refers to the identity matrix. Note that similar to the preceding chapter here $\alpha \equiv (\xi, \beta)$. Here because of the interband excitation selection rules discussed in the previous chapter and the fact that the QW is narrow, $\beta$ sums only over s-states of excitons. Also, as mentioned in the previous chapter, for $j = HH1$ and $LH1$, $\xi$ takes 3/2 and $-1/2$, respectively. The linear absorption spectrum of the QW is given by the real part of $\Gamma(i\omega_p)$ in which $\omega_p$ refers to the probe field frequency.
6.3 One-field generated multi-exciton coupling in quantum wells

We now use the results of the preceding section to estimate the effects of the multi-subband excitonic coupling on the interband absorption spectrum of a GaAs/Ga_{0.7}Al_{0.3}As QW structure with an 8.5 nm well width. The following spectra were obtained assuming dephasing rates of bound excitons associated with $HH1$ and various electron subbands (E1, E2 and E3) to be $\frac{1}{T_{E1-HH1}^{\text{deph}}} = 0.5 \text{ ps}^{-1}$, $\frac{1}{T_{E2-HH1}^{\text{deph}}} = 2.5 \text{ ps}^{-1}$ and $\frac{1}{T_{E3-HH1}^{\text{deph}}} = 15 \text{ ps}^{-1}$, and those corresponding to inter-exciton-state transitions ($i - HH1 \rightarrow i' - HH1$) to be $\frac{1}{T_{\text{interexciton}}^{\text{deph}}} = 3 \text{ ps}^{-1}$, $\frac{1}{T_{E1-E2}^{\text{deph}}} = 15.5 \text{ ps}^{-1}$, and $\frac{1}{T_{E2-E3}^{\text{deph}}} = 17.5 \text{ ps}^{-1}$. The corresponding rates involving $LH1$ were also assumed to be respectively $1.5 \text{ ps}^{-1}$, $10 \text{ ps}^{-1}$, $20 \text{ ps}^{-1}$; $11.5 \text{ ps}^{-1}$, $21.5 \text{ ps}^{-1}$, and $30 \text{ ps}^{-1}$. The corresponding dephasing rates of the exciton continua are considered to be the same as those of the light-hole excitons. The large values of dephasing rates involving $E3$ are related to the fact that this subband is quasi-bound in this particular structure. We also include a 4 meV (full width at half maximum) inhomogeneous broadening of the bound states.

The solid line in Fig.6.2 shows the linear interband absorption spectrum in the absence of the IR field, calculated using the above parameters. Assuming an infrared field with a photon energy of $\sim 124 \text{ meV}$ (a CO$_2$ laser line), $\Delta_1 = -12 \text{ meV}$ and $\Delta_2 = -16 \text{ meV}$, where $\Delta_1$ and $\Delta_2$ are the detunings of the field for transitions between E1-E2 and E2-E3, respectively. The dotted spectrum is that calculated when the Rabi frequency ($\mu_{12}E/\hbar$) due to this field is $\Omega = 15 \text{ ps}^{-1}$ (or $E = 4.83 \times 10^6 \text{ Vm}^{-1}$ with the assumption of $\mu_{12} = e \times 2 \text{ nm}$), but when only the E1-E2 coupling is included (the $\Xi$ system). Both of the bound states are split into doublets, the centroids of each being red shifted from the undressed states due to the -12 meV detuning of the IR field (note the low energy LH exciton peak is barely resolvable, near -11 meV). The dashed line in Fig.6.2 shows the result of the full calculation, self consistently including both E1-E2 and E2-E3 coupling (the $\Xi$ system). At this intensity, the main effects of including the additional coupling are to increase the red-shifts of the doublets, and to introduce significant broadening (recall
Figure 6.2: Absorption spectrum of the infrared-coupled quantum well using the two- (Ξ system) and three-conduction-subband (Ξ_ system) coupling models (dotted and dashed lines) for Ω=15 ps^{-1}. The solid line corresponds Ω = 0. The peaks are labeled based on their heavy-hole (HH) and light-hole (LH) natures. Here $E_p$ refers to the probe field photon energy and $E_{c1}$ to the energy of $E_1$.

that the dephasing rate of $E3$ is large, ~ 40 ps^{-1}).

A similar set of curves are shown in Fig.6.3, but with Ω =30 ps^{-1} (or $E = 9.6 \times 10^6$ Vm^{-1} with the assumption of $\mu_{12} = \mu_{23} = e \times 2$ nm). This is nearly correspond to one of the intensities used in Ref. [11]. While the dotted spectrum, associated with just 1-2 coupling, again consists of two red-shifted doublets (the low energy component of the LH doublet now being clearly resolved near -20 meV) split further apart by the stronger coupling field, the dashed curve corresponding to the full calculation is no longer as simple. While the “light-hole peaks”, near -26 meV and 14 meV, might still be approximated as red-shifted and broadened versions of their dotted counterparts, the heavy-hole peaks clearly can not. The peak near -7 meV is red shifted much more from the 6 meV peak in the $E_1 - E_2$ coupled calculation than is the low energy peak near -43 meV, only 7 meV below its dotted counterpart. There are more differences associated with the field-induced broadening as well.
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Figure 6.3: Absorption spectrum of the infrared-coupled quantum well using the two (Ξ system) three-conduction-subband (Ξ- system) coupling models (dotted and dashed lines) for Ω = 30. All other parameters are the same as those of Fig.6.2.

The above calculations were intended to be reasonably realistic, and as such, the spectra in Figs.6.2 and 6.3 correspond to the dressed superpositions of bound and continuum heavy hole, and bound and continuum light hole excitons. Note that the relatively weak blue shift of the dominant heavy hole bound state at higher intensities is in better agreement with experiment [11] than previous calculations based on the $E1 - E2$ coupling alone (Ξ system). The extensive broadening seen in these figures is also more consistent with the experiment [11]. To explain the origin of this result more clearly, and to more generally describe the fundamentally different behavior of the two and three coupled level systems, we now consider the heavy hole bound state in isolation and neglect inhomogeneous broadening. Fig.6.4 shows the calculated absorption near the interband threshold for a single 1s heavy hole exciton under the same conditions used to generate Figs.6.2 and 6.3. The solid, dotted and dashed curves in Fig.6.4 correspond, respectively, to $\Omega = 0$ (solid line), 15 ps$^{-1}$ (dashed line) and 30 ps$^{-1}$ (dotted line) in the Ξ- system.

The results in Fig.6.4 are clearly analogous to those obtained for the atomic systems
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Figure 6.4: Absorption spectrum of 1s bound heavy hole exciton in isolation without any inhomogeneous broadening for various coupling field intensities in the Ξ_ system. The solid line corresponds to $\Omega = 0$, the dashed line to $\Omega = 15 \text{ ps}^{-1}$, and dotted line to $\Omega = 30 \text{ ps}^{-1}$. The inset shows full spectrum of $\Omega = 0$ compare to that of $\Omega = 15 \text{ ps}^{-1}$.

in chapter 4. Here infrared coupling of the E1-HH1 and E2-HH1 and E3-HH1 excitons leads to one- and two-photon coupling effects. Considering the transition energies of the E1-E2 and E2-E3 transition the same for simplicity, the eigen-states associated with these coupling mechanisms are

\begin{align*}
|\tau >_{1s} &= -\frac{1}{\sqrt{2}}[|E1-HH1 >_{1s} -|E3-HH1 >_{1s}], \\
|s >_{1s} &= \frac{1}{\sqrt{2}}[\frac{1}{\sqrt{2}}|E1-HH1 >_{1s} +|E2-HH1 >_{1s} + \frac{1}{\sqrt{2}}|E3-HH1 >_{1s}], \\
|t >_{1s} &= \frac{1}{\sqrt{2}}[\frac{1}{\sqrt{2}}|E1-HH1 >_{1s} -|E2-HH1 >_{1s} + \frac{1}{\sqrt{2}}|E3-HH1 >_{1s}],
\end{align*}

where $|E1-HH1 >_{1s}$, $|E2-HH1 >_{1s}$, and $|E3-HH1 >_{1s}$ are the 1s eigen-states of the total Hamiltonian of the QW in the absence of the IR for the heavy hole exciton (Eq. (6.1)).

Note that similar to the atomic systems in Chapter 4 the position of the $|\tau >_{1s}$ state
is fixed at the uncoupled exciton energy,

\[ H_1|s >_{1s} = 0, \]  \hspace{1cm} (6.6)

while the |s >_{1s} and |t >_{1s} states split symmetrically

\[ H_1|s >_{1s} = \hbar \sqrt{2} \Omega|s >_{1s}, \]  \hspace{1cm} (6.7)

\[ H_1|t >_{1s} = -\hbar \sqrt{2} \Omega|t >_{1s}. \]  \hspace{1cm} (6.8)

Note that the idealized behavior is somewhat masked in the more realistic simulations, largely due to the finite damping associated with real QW states. This damping smears out the triplet when \( \Omega < 1/T_{ij} \). However, Eqs. (6.3)-(6.8) still provide insight as to the role of damping, since they clearly predict that the central feature should be independent of the damping of E2-HH1, while the two side lobes should depend on the damping of all three transitions.
Chapter 7

Intraband nonlinear optical processes in n-doped QW's

7.1 Introduction

In chapters 5 and 6 we showed how the intraband excitonic coupling of a QW can modify its interband absorption and emission spectra. As mentioned before, QW's can support a different class of nonlinear dynamics when both the coupling and probing occur within the same band. In particular, for the case of the conduction band (all-conduction-band schemes), this has received a great amount of attention in recent years. This is because these schemes are promising candidates for optical devices, such as infrared photodetectors [3], frequency converters [69], infrared and far-infrared lasers [5, 70], etc. They are also important for study of the semiconductor physics including intersubband electron relaxation times [71], many body effects, etc. In particular, all-conduction-band schemes are appealing hosts for investigation of the optical nonlinear effects in QW semiconductors. This is because of their large transition dipole moments along the growth direction [2], and the fact that for small electron wave-vectors the conduction subband dispersions are similar. These lead to atomic-like non-linear phenomena in QW's.

Motivated by these interesting features, we have done a comprehensive in-depth study of the nonlinear effects in infrared-coupled n-doped QW's with all-conduction-band schemes. This work has been done in two parts: In the first part we considered the cases where the intense infrared fields did not affect the partially populated ground subbands. Therefore the electron distributions in these subbands remained Fermi-Dirac type in the presence of the intense IR fields. This is the subject of this chapter. Here we study how infrared coupling and dressing of upper conduction subbands of n-doped QW's
change their linear intersubband absorption spectra. We are not aware of similar studies done by others. In the second part we studied the cases where the ground subbands were also significantly affected by the IR fields. Here a large number of electrons were excited into the upper subbands, causing different types of nonlinear effects. In chapters 8 and 9 we discuss these cases in detail. The results of the present chapter have been published in Refs. [23, 26, 29].

7.2 All-conduction-band schemes in n-doped QW's

To identify the all-conduction band schemes here we use the same notations as those used for the case of the atomic systems. Two generic simple schemes in symmetric QW's with $\Lambda$ and $\Xi$ configurations are shown in Fig.7.1a and 7.1b, respectively. Here two strongly driven upper subbands are monitored weakly from the ground subband. As shown in this figure one of the subbands can be in the continuum (quasi-bound subband). As will be shown later, employing such subbands enhances our ability to manipulate coherent effects in QW's.

In practice, however, one can consider asymmetric quantum wells where the wave functions and energies of the subbands can be engineered. Here since the subband parties are not well-defined, a probe field can detect each of the upper subbands from the ground subband. Using these properties we can consider $\Pi$, $K$, or $\Xi_-\!$ configurations similar to those discussed in the chapter 4. In the asymmetric structure shown in Fig.7.2 we have a $\Xi_-\!$ system when we monitor the ground to the first upper subband transition (a), and (b) $K$ or (c) $\Pi$ system when the ground to third upper or second upper subband is detected, respectively.

7.3 Conduction band infrared coupling with $\Xi_-\!$, $K$, and $\Pi$ configurations

To study $\Xi_-\!$, $K$ and $\Pi$ systems in QW's we consider lightly n-doped asymmetric QW's with energy levels such as those shown in Fig.7.2. The infrared fields are all polarized
Figure 7.1: Schematic diagrams of (a) $\Lambda$ system, and (b) $\Xi$ system in symmetric QW's. The shaded area refers to the quasi-bound subband, and $\omega_1$ and $\omega_p$ are the coupling and probe field frequencies, respectively.
Figure 7.2: Schematic diagrams of (a) $\Xi_\gamma$, (b) $K$, and (c) $\Pi$ in an asymmetric QW.
along the growth direction of the QW and are quite off-resonance from transitions between the other excited subbands and from those between the ground to the higher energy subbands. Under these conditions the total interaction Hamiltonian in the interaction picture is:

\[ H_{\text{int}} = H_1' + H_2', \]  

(7.1)

where within electric-dipole and rotating-wave approximations we have:

\[ H_1' = \hbar \sum_k \Omega_{13} \{ e^{i\Delta_k t} a_{3,k}^\dagger a_{1,k} + e^{-i\Delta_k t} a_{1,k}^\dagger a_{3,k} \}, \]  

(7.2)

\[ H_2' = \hbar \sum_k \Omega_{23} \{ e^{i\Delta_k t} a_{3,k}^\dagger a_{2,k} + e^{-i\Delta_k t} a_{2,k}^\dagger a_{3,k} \}. \]  

(7.3)

Here \( \Omega_{13} = -\mu_{13} E_1/\hbar \) and \( \Omega_{23} = -\mu_{23} E_2/\hbar \) are the Rabi frequencies. \( E_1 \) and \( E_2 \) refer to the amplitudes of infrared fields with frequencies \( \omega_1 \) and \( \omega_2 \) respectively. The detunings are:

\[ \Delta_k^1 = \omega_k^3 - \omega_1^3 - \omega_1, \]  

(7.4)

\[ \Delta_k^2 = \omega_k^3 - \omega_2^3 - \omega_2, \]  

(7.5)

where \( \omega_i^k \)'s are defined as follow

\[ \omega_i^k = E_i^c(i, k)/\hbar. \]  

(7.6)

Here \( E_i^c(i, k) \)'s are the excited conduction subband energies and \( i \) (subband index) takes 1, 2, and 3.

Having the total Hamiltonian of the system which includes the infrared-QW interaction term (Eq. (7.1)) as well as electron-electron, electron-phonon interactions etc., we can obtain the equations of motion of the system, in general. However, since here we are dealing with the weakly probing of the ground to upper subbands, the dynamic damping effects caused by the electron-electron scattering processes are not significant. Note that these processes still play key rules in the ground subband damping. This is
because a weak transition associated with the probe field leaves a quasi-hole in the Fermi distribution of the ground subband. This hole is rapidly damped by the electron-electron scattering, which is the dominant damping process in this subband. To see this and other aspects of the system let us consider the equations of motion at a specific $k$:

$$\frac{\partial \rho^{k}}{\partial t} = -\frac{i}{\hbar} [H_{\text{int}}, \rho^{k}] + \frac{\partial \rho^{k}}{\partial t} |_{\text{lincoh}} e^{-e} + \frac{\partial \rho^{k}}{\partial t} |_{\text{lincoh}} e^{-p}. \tag{7.7}$$

Here $\frac{\partial \rho^{k}}{\partial t} |_{\text{incoh}}$ refers to the incoherent processes caused by electron-electron scattering. Within our approximations this term only affects the ground subband. Also since this subband is not significantly affected by the IR fields we can apply relaxation time approximation (RTA):

$$\frac{\partial \rho^{k}_{00}}{\partial t} |_{\text{lincoh}} e^{-e} = -\Gamma_{0} e^{-e}(k)(\rho^{k}_{00} - \rho^{0k}_{00}). \tag{7.8}$$

Here $\rho^{0k}_{00}$ refers to the equilibrium probability of finding electrons in the states $|0, k \rangle$ and therefore it is represented by a Fermi distribution. $\Gamma_{0} e^{-e}(k)$ is the energy relaxation rate of the Fermi sea distortion or quasi-hole ($\delta \rho^{0k} = \rho^{0k} - \rho^{0k}_{00}$). Note that RTA is valid as long as the ground subband is not significantly distorted. In the presence of a strong laser field nearly resonant with the ground to an upper subband this approximation becomes invalid. This will be discussed in detail in the next chapter.

The third term in Eq. (7.7) represents the electron-phonon scattering process’ contribution to the damping of the electronic system. This depends on the kinetic energy of the electrons and which subband they are in. For the ground subband, if the Fermi energy is less than the energy of longitudinal-optical phonons (LO-phonons) the only such process is electron-acoustic phonon scattering. This process is weak and diffusive [72]. If the first upper subband is separated from the ground subband by an energy greater (smaller) than the LO-phonon energy electron decays by emission of LO-phonons (acoustic phonons). The damping caused by the electron-phonon scattering processes are thus represented in our model by

$$\frac{d \rho^{k}_{00}}{dt} |_{\text{lincoh}} e^{-e} = \frac{d \rho^{k}_{00}}{dt} |_{\text{diff}}, \tag{7.9}$$
and
\[ \frac{d\rho_{ii}^{k}}{dt}_{\text{incoh}} = -\Gamma_{i}^{e-p}(\rho_{ii}^{k} - \rho_{ii}^{0k}). \] (7.10)
Here \( i \) takes 1, 2, and 3, and \( \Gamma_{i}^{e-p} \) refers to the intersubband transition rate of an electron in the \( i \)th subband due to emission of phonons. Eq. (7.9) explicitly refers to the diffusive nature of electron energy relaxation in the first subband. For the off-diagonal terms we have
\[ \frac{d\rho_{ij}^{k}}{dt}_{\text{incoh}} = -\gamma_{ij}^{p}\rho_{ij}^{k}, \] (7.11)
where \( \gamma_{ij}^{p} \) is the dephasing rate caused by the electron-phonon scattering process. This is given by
\[ \gamma_{ij}^{p} = \frac{1}{2}(\Gamma_{i}^{e-p} + \Gamma_{j}^{e-p}), \] (7.12)
where \( i \) and \( j \) run over all subband indices. To find the dephasing rates associated with other transitions, \( \gamma_{0j} \), we follow the same method but include the contribution of electron-electron scattering.

After applying Eqs. (7.8)-(7.12) to Eq. (7.7), the equations of motion of the system can be obtained. The results after taking steps similar to those explained in chapter 2, are a set of equations which can be written in a formal form as that in Eq. (2.36). Here, however, the matrices are labeled by \( k \)'s, i.e. we have \( L^{k}, K^{k} \) and \( \Phi^{k} \). To proceed with the calculation we use linear response theory. To do this for the \( \Xi_{-} \) system, for example, we apply this theory to a specified \( k \) and calculate the unequal-time correlation functions \( \langle [p_{k}^{+}(t'), p_{k}^{-}(t)] \rangle \). Here \( p_{k}^{-} \) and \( p_{k}^{+} \) are the negative and positive frequency components of the polarization of the \( \Xi_{-} \) system given by:
\[ p_{k}^{\pm}(t) = \mu_{01}^{\pm} a_{0,k}^{\dagger} a_{1,k}. \] (7.13)

The imaginary and real parts of the susceptibility with a QW with a \( \Xi_{-} \) configuration can then be, respectively, related to the real and imaginary parts of the following two-time correlation function:
\[ \sum_{k} \rho_{00}^{0k} \langle [p_{k}^{+}(t'), p_{k}^{-}(t', z)] \rangle |_{z = i\omega} . \] (7.14)
Calculation of $< [p_k^{+}(t'), p_k^{-}(t', z)] >$'s in Eq. (7.14) are similar to those discussed in chapter 2. The results have also the same formal forms as that seen in Eq. (2.96). The same are correct for the systems with K and Π configurations after making proper changes.

### 7.4 Intraband laser-induced transparency in quantum wells

As mentioned before, one important feature of the all-conduction-band optics in QW's is that they can present atomic-like dynamics. As we show in this section one can even choose the QW parameters such that the transitions from the ground to an upper subband show a laser-induced transparency (LIT) similar to that seen in the case of atomic systems (Fig.3.1). To see this let us consider an asymmetric QW similar to that shown in Fig.7.3. The advantage of this structure is that the two coupled upper subbands are detectable. This means that both Λ and Ξ systems can be found in a single system by tuning the probe field frequency. Another advantage of such a structure is that we can choose its parameters (such as widths of layers, material compositions, etc.) to get the most desirable damping rate for each subband. Here we consider the 0-1 transition energy less than those of LO-phonons. Therefore electrons in $|1, k >$ can only decay by emitting acoustic phonons. This makes this subband very narrow ($\Gamma_0^{e-p}=0.1$ ps$^{-1}$). On the other hand since $|2, k >$ is close to the continuum edge it is very broad with $\Gamma_2^{e-p} \sim 30$ ps$^{-1}$.

For simplicity in this chapter we consider an average value for $\Gamma_0^{e-p}(k)$. Since the electron density and distribution are not changed by the fields and the effective masses of electrons in various subbands are considered to be the same, this does not cause a significant error. $\Gamma_0^{e-p} \sim 3$ ps$^{-1}$ which is much larger than that caused by electron-acoustic phonon scattering ($\sim 0.1$ ps$^{-1}$).

The results for the 0-2 transition (Λ system) are shown in Fig.7.4. When the infrared intensity is zero (no IR coupling field) the absorption spectrum associated with this
transition has a Lorentzian line-shape (solid line). However, in the presence of the field with $\Omega = 5 \text{ ps}^{-1}$, a transparency hole is generated at the spectrum line-center. The hole widens and deepens as the field intensity increases. Note that the effects of the infrared coupling here are very similar to those seen in an atomic system (Fig.3.1). The main difference between these two systems’ dynamics is the order of magnitude of the coupling field intensities required to induce a coherent effect.

The results, when the probe field monitors the 0-1 transition ($\Xi$ configuration), are shown in Fig.7.5 under the same conditions as those of Fig.7.4. Here, in contrast to the $\Lambda$ system, the main effect of the coupling process is seen as a pure broadening of the absorption peak. In other words, LIT does not occur.

These effects can be explained in terms of the developments presented in chapters 3 and 4. They show how damping rates affect the development of quantum interferences. In the case of the $\Lambda$ system since the quantum interferences are scaled to the lower (narrow) subband, they occur at the very early stage of the coupling process. In the case of the $\Xi$ system, however, the interferences are scaled to the higher (broad) subband. Therefore, they are virtually suppressed for all field intensities.
Figure 7.4: LIT in the conduction band of a QW with a $\Lambda$ configuration. The solid line corresponds to $\Omega=0$, the long dashed line to $\Omega=5$ ps$^{-1}$, the short dashed line to $\Omega=10$ ps$^{-1}$, and the dashed-dotted line to $\Omega=30$ ps$^{-1}$.

Figure 7.5: Absorption spectrum of a $\Xi$ system in the conduction band of a QW for various field intensities. All other specifications are the same as those in Fig.7.4.
7.5 Coherent manipulation of the intersubband transitions in the absence of intense electron excitations

The intersubband LIT discussed in the preceding section showed how one can induce quantum interferences similar to those discussed in chapter 3. In this section we expand this concept and show how one can coherently manipulate the intersubband transitions in n-doped QW's in the absence of extensive electron excitation. This section is analogous to the atomic systems discussed in chapter 4. Here we consider $\Gamma_0^{-}=7$, $\Gamma_1=3$, $\Gamma_2=10$ and $\Gamma_3=40$ ps$^{-1}$.

7.5.1 Dynamics in the \( \Pi \) system

In order to illustrate the infrared coupling effects in the \( \Pi \) system, we consider a quantum well structure such as that shown in Fig.7.2c. With the assumption of the resonant coupling ($\Delta_2 = \Delta_1 = 0$) and $\Omega_{23} = \Omega_{13} \equiv \Omega$, the evolution of the absorption spectrum of $|2, k \rangle$ is shown in Fig.7.6 for various coupling field intensities. Based on this evolution three characteristic values for the coupling field intensities are found. When $\Omega << \gamma_{01}, \gamma_{02}$ and $\gamma_{03}$, the effect of the coupling fields is to broaden the Lorentzian absorption line of the uncoupled spectrum. This process seems to continue even when $\Omega$ becomes close to $\gamma_{01}$ and $\gamma_{02}$ but much less than $\gamma_{03}$ (dashed line). For values of $\Omega$ close to these dephasing rates (here $\gamma_{03}$) the absorption spectrum starts to develop three maxima, one at the line-center (unshifted) and the others at either side shifted proportional to $\Omega$ (dotted line). At this stage further increase in $\Omega$ causes further enhancement and finer resolution of these peaks until a limit is reached at which any increase in the value of $\Omega$ does not change the central peak but only moves the sideband peaks by $\sqrt{2}\Omega$ away from the uncoupled line-center.

Now let us consider the effects of detuning the coupling fields from their corresponding transitions. We first start with the case in which both fields are detuned equally from the $|3, k \rangle$ line-center. Fig.7.7 shows the evolution of $|2, k \rangle$ for the same parameters as
Figure 7.6: Intersubband absorption of a QW with a resonant $\Pi$ configuration. The Rabi frequencies are $\Omega = 0 \ \text{ps}^{-1}$ (solid line), $\Omega = 7 \ \text{ps}^{-1}$ (dashed line), $\Omega = 20 \ \text{ps}^{-1}$ (dotted line) and $\Omega = 40 \ \text{ps}^{-1}$ (dashed-dotted line). $E_p$ and $E_{02}$ refer to energies of the probe and ground to $|2, k>$ respectively.
Figure 7.7: Intersubband absorption a II system in a QW for $\Delta_1 = \Delta_2 = 10$ meV and various coupling fields intensities. The solid line represents the uncoupled spectrum, the dashed line $\Omega = 20$ ps$^{-1}$ and the dotted line $\Omega=50$ ps$^{-1}$. All other specifications are the same as Fig.7.6.

those of Fig.7.6 and various intensities of the two driving fields at $\Delta_1 = \Delta_2=10$ meV. As one can see, at the beginning the spectrum is asymmetric, but as the intensities increase it develops a triple line spectrum and ultimately becomes symmetric in the high-field limit. In other words, the detuning loses its effects at this limit.

If we now detune the fields equally but with opposite signs, $\Delta_2 = -\Delta_1=10$ meV from the $|3, k>$ line-center, the spectrum at $|2, k>$ no longer develops a symmetric triplet even at high field intensities (see Fig.7.8). Instead a detuned peak which becomes fixed in energy (-10 meV from $|2, k>$ line-center) and two other peaks with variable energies and peak magnitudes are generated. The one which has a fixed energy becomes also nearly fixed in magnitude at high field intensities.

The nature of these nonlinear optics and the coupling mechanisms responsible for
Figure 7.8: Intersubband absorption a Π system in a QW for $\Delta_2 = -\Delta_1 = 10$ meV and various coupling fields intensities. The solid line represents the uncoupled spectrum, the dashed line $\Omega = 25$ ps$^{-1}$ and the dotted line $\Omega = 50$ ps$^{-1}$. All other specifications are the same as Fig. 7.6.
them are identical to the corresponding ones discussed for the atomic systems in chapter 4. These results, however, provide useful information for practical tests of these systems in QW's.

7.5.2 Dynamics in the K system

To study the corresponding evolution of $|3, k >$ we now tune the probe field to become nearly resonant with the 0-3 transition, i.e. switching to the K system (Fig.7.2b). The results of calculation for similar structure as that in Fig.7.6 are shown in Fig.7.9. Note that similar to the case of Fig.7.6, if $\Omega << \gamma_0$, the net effect is just to broaden the absorption spectrum (not shown). But in contrast to Fig.7.6, at intensities in which Fig.7.6 apparently continues its broadening, Fig.7.9 has already developed a line doubling (dashed line). Here this effect occurs when $\Omega$ becomes close to $\gamma_0$. When we reach the high-field limit the doubled peaks remain unchanged in shape but move by $\sqrt{2}\Omega$ away from the uncoupled $|3, k >$ line-center.

Under the detuned coupling conditions similar to that of Fig.7.8, the spectrum of $|3, k >$ shows a central line in addition to two asymmetric sideband peaks (see Fig.7.10). By increasing the coupling field intensities the central peak vanishes while the sideband peaks grow and become symmetric and shifted from the line-center of $|3, k >$.

7.5.3 Dynamics in the $\Xi_-$ system

To study evolution of a system with a $\Xi_-$ configuration, we consider a system as that shown in Fig.7.2a. The results of calculations for a structure the same as that in Fig.7.6 are shown in Fig.7.11 when $\Omega_{12} = \Omega_{23} \equiv \Omega$ and $\Delta_1 = \Delta_2 = 0$. Note that similar to Fig.7.6, at the early stage of coupling the absorption spectrum becomes broadened (dashed line). But in contrast to Fig.7.6, before the triplet effect evolves, it has already developed a doublet (dotted line). This process continues until $\Omega$ becomes close to $\gamma_0$ (here larger than $\gamma_0$, $\gamma_0$) when the triplet effect shows up.
Figure 7.9: Intersubband absorption of the 0-3 transition in a QW with a K configuration. All specifications are the same as those of Fig. 7.6. $E_{03}$ refers to energy of the ground to $|3, k >$. 
These phenomena can be explained in terms of the competition between one- and two-photon coupling effects addressed in section 4.5. We briefly discuss this in the next section.

7.6 Damping rates effects

In chapter 4 we showed that when three levels of a system were strongly coupled together, their dynamics were governed by one- and two-photon couplings. It is clear that the same mechanisms are also responsible for the dynamics of the systems discussed in the preceding section. Here, however, related to the intrinsic effects caused by the electron-electron scattering processes in the ground subband, it is useful to highlight several facts:

(i) In a QW structure, the Π and Ξ systems develop into similar spectra. The differences are manifested in their dynamics before reaching the high-field limit.

(ii) The widths $W_c$, and amplitudes, $A_c$, of the central peaks depend only on the
Figure 7.11: Intersubband absorption of the 0-1 transition in a quantum well $\Xi_-$ system for various coupling field intensities. The solid line corresponds to $\Omega=0$ ps$^{-1}$, the dashed line to $\Omega=3$ ps$^{-1}$, the dotted line to $\Omega=10$ ps$^{-1}$, and the dashed-dotted line to $\Omega=40$ ps$^{-1}$. Here $\Gamma_0^{\text{reg}}=7$, $\Gamma_1=3$, $\Gamma_2=6$, and $\Gamma_3=40$ ps$^{-1}$. 
dephasing rates of $|1, k>$ and $|2, k>$ and can be obtained from the following relations:

$$W_c = \gamma_{01} + \gamma_{02},$$

(7.15)

$$\frac{A_0}{A_c} = \frac{\gamma_{01} + \gamma_{02}}{\gamma_{02}}.$$  

(7.16)

Here $A_0$ refers to the uncoupled peak amplitude of the $\Pi$ system spectrum.

(iii) The sideband peaks depend on relaxation rates of all excited subbands. We obtain the following relations for the sideband widths, $W_s$, and peak amplitudes, $A_s$:

$$W_s = \frac{\gamma_{01} + 2\gamma_{03} + \gamma_{02}}{4},$$

(7.17)

$$\frac{A_0}{A_s} = \frac{\gamma_{01} + 2\gamma_{03} + \gamma_{02}}{\gamma_{02}}.$$  

(7.18)

Here again Eq.(7.18) gives the ratio of uncoupled peak, $A_0$, to the sideband peak amplitude, $A_s$, in a $\Pi$ system. Eqs. (7.15)-(7.18) are valid for the $\Xi_-$ system if we replace $\gamma_{01}$ and $\gamma_{02}$ with $\gamma_{03}$ and $\gamma_{01}$, respectively.

The basic dynamics of the spectra seen in the preceding section are similar to those discussed in section 4.4. Briefly, if $\gamma_{01} << \gamma_{03}$, as it is in the case of Fig.7.6, $\Omega$ reaches $\gamma_{01}$ much sooner than $\gamma_{03}$. Therefore the two-photon process with central peak manifestation appears first. This explains why in Fig.7.6, for $\Omega$'s close to $\gamma_{01}$ but far from $\gamma_{03}$, no significant change in $|2, k>$ can be seen (dashed line). But once $\Omega$ reaches $\gamma_{03}$ and one-photon process becomes significant the sideband peaks become resolved (dotted line). This process is confirmed by the evolution of Fig.7.11 in the $\Xi_-$ system. Here since $\gamma_{03}$ is larger than $\gamma_{02}$, the ratio $\frac{\gamma_{02}}{\Omega}$ becomes close to unity sooner than $\frac{\gamma_{03}}{\Omega}$. Therefore when we detect $|1, k>$, doubling which is a sign of one-photon process becomes dominant at the early stage of coupling. When we are close to the high-field limit, however, the two types of coupling mechanisms show up (Fig.7.11, dashed line).

The story of the $K$ system in which only the one-photon process can happen is rather different. Here we find that the system evolution can be described by the following
Chapter 7. Intraband nonlinear optical processes in n-doped QW's

parameter:

\[ \gamma_{\text{eff}} = \frac{\gamma_01 \gamma_02}{\gamma_01 + \gamma_02} \]  

(7.19)

If \( \Omega \) becomes close to \( \gamma_{\text{eff}} \), the \( |3, k> \) spectrum starts to resolve a doubling effect while if \( \Omega << \gamma_{\text{eff}} \) only broadening happens. Since the Fig.7.9 evolution is scaled by \( \gamma_{\text{eff}} \) which is in general less than \( \gamma_01 \) and \( \gamma_02 \), the doubling effect happens here earlier than that in Fig.7.6. In the limits in which the dephasing rates of \( |1, k> \) or \( |2, k> \) are very different, the coupling of \( |3, k> \) with the broader subband is weak and therefore its evolution is determined mainly by the smaller subband dephasing rate.

7.7 Coherent destruction effect

The concept of coherent destruction effect was discussed in Sect.3.5. We found that the coherent processes generated in the two-field-coupled systems depend on the relative intensities of the coupling fields. When one field becomes more intense than the other, it destroys the coherences generated by the weaker field. Here we discuss this phenomenon in the \( \Pi \) system in some detail.

Consider a \( \Pi \) system where the two infrared fields are resonant with their corresponding transitions but with different intensities (\( \Omega_{13} \neq \Omega_{23} \)). First we put \( \Omega_{13} = 40 \text{ ps}^{-1} \) and change \( \Omega_{23} \) such that \( 1 \leq \frac{\Omega_{23}}{\Omega_{13}} \), i.e. taking \( \Omega_{13} \leq \Omega_{23} \). Fig.7.12 shows the way in which the absorption spectrum of the transition to \( |2, k> \) changes when this ratio goes from unity to larger values. When \( \frac{\Omega_{23}}{\Omega_{13}} = 1 \) we observe a triplet spectrum (solid line). But by increasing \( \Omega_{23} \) the central peak is vanishing while the sideband peaks become stronger and moving away from the uncoupled peak line-center. Ultimately at the limit of \( \frac{\Omega_{23}}{\Omega_{13}} >> 1 \) the central peak has disappeared and the remaining part is just an illustration of the two-subband coupling of \( |2, k> \) with \( |3, k> \) by the infrared field with frequency \( \omega_2 \). Note that if \( \Omega_{13} >> \Omega_{23} \) occurs, \( |2, k> \) becomes decoupled from the rest of the system. As a result, the absorption spectrum associated with the 0-2 transition becomes similar to that when there is no field.
Figure 7.12: Intersubband absorption of a \( \Pi \) system for various values of \( \frac{\Omega_{23}}{\Omega_{13}} \). The solid line corresponds to \( \frac{\Omega_{23}}{\Omega_{13}} = 1 \) and the dashed line to \( \frac{\Omega_{23}}{\Omega_{13}} = 2 \). The dotted line is the uncoupled absorption spectrum. All other specifications are the same as Fig. 7.6.
Chapter 8

Electron-electron scattering effects on the QW intersubband transitions

8.1 Introduction

The coupling processes discussed in the preceding chapter were caused by intense laser fields nearly resonant with the transitions between the upper subbands of n-doped QW's. Here the ground subbands which contained Fermi-Dirac type electron distributions were left unchanged. In this chapter and the following one we study the cases which involve with strong photo-excitation of electrons from the ground into upper subbands. These studies have applications in semiconductor lasers in mid and far infrared (THz) frequency range, infrared detectors, etc. Also they can be used to study semiconductor physics, such as infrared-driven two-dimensional electron gas systems, coherent control of the electron populations, intersubband scattering rates of electrons, etc. Note that since here the intersubband transitions cause significant changes in the real populations, electron-electron scattering effects play a major role in the evolution of the system. In this chapter we show how these scattering processes renormalize the polarization dephasing rates associated with the intersubband transitions. It will also be shown that a simple two-subband system can demonstrate various interesting effects, such as steady-state non-Fermi (non-equilibrium) electron distribution, tunable gain at far and mid infrared ranges and quantum interference effects. In the following chapter we extend these developments to explore more exotic nonlinear optics in n-doped QW's. The results of this chapter have been reported in Ref. [24].
8.1.1 Failure of relaxation time approximation in strongly driven n-doped QW’s

There are many theoretical reports on how intense laser fields change the optical properties of QW’s when they are nearly resonant with the ground to upper subband transitions [16, 17, 74]. In most of these reports, however, dephasing rates of the polarization associated with these transitions and damping of electrons in the ground subband were attributed to electron-phonon scattering. As a result, the relaxation time approximation (RTA) was extensively used, although the intensity of the infrared fields were not small. Even in the literature where the electron-electron scattering effects have been studied, the intersubband polarization decay rates were introduced phenomenologically, using constant parameters [79, 80]. We will show in this chapter that when we are dealing with extensive electron excitation, application of RTA and phenomenological dephasing rates may give rise to unrealistic results.

8.1.2 The realistic picture

Intersubband transitions involving the ground subband are strongly affected by the electron-electron scattering process. This implies that the actual dephasing rates depend on the electron wave-vector (k), distribution function, and density. Even when the infrared field only weakly perturbs the ground subband distribution function so that the RTA is valid (the weak-field regime), the strong k-dependence implies that one can not consider the damping rates to be constant for all values of k, in principle. However, since for small k’s electrons in different conduction subbands have nearly the same effective masses, this does not give rise to any significant effect. Therefore in the preceding chapter we treated the electron-electron scattering effects using constant parameters estimated from the electron-electron scattering rates at all k’s.

When the IR intensity is not weak, however, three major issues are raised which to our knowledge also remain unexplored: (i) The IR field generates non-Fermi-Dirac
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(non-equilibrium) steady-state electron distribution functions in the coupled subbands. This is in contrast to some previous investigations where the distributions were assumed to be Fermi type. This study is the intersubband analog of the cw semiconductor lasers where electron-hole pairs are generated through injection pumping and removed by stimulated emission at a fixed frequency [86]. In such a case the frequency-selective carrier removal tends to cause a kinetic hole in the quasi-equilibrium carrier distribution. (ii) Saturation effects can be seen once the higher subband is significantly occupied. This case has been studied in the past by employing the RTA and ignoring electron-electron scattering [16, 73, 74]. (iii) One can use an additional weak IR field to induce quantum interference and gain processes in a simple two-subband QW system. This concept is very appealing for generation of far-infrared lasers in QW’s.

The goal of this chapter is to address these issues by considering a rigorous treatment of intersubband transitions in n-doped QW’s including the mutual effects of the screened electron-electron scattering process and the intersubband transitions. We show that the damping rates of electrons and polarization not only depend on the temperature, the electron distribution function, the density, the effective mass, and the QW parameters, but also on the IR intensity and frequency. In the strong-field regime we also discuss the quantum interference and gain processes when the strongly driven n-doped QW is probed by a weak field with the same polarization as the driving field. We also discuss generation of gain with hidden inversion (Raman gain), and gain caused by stimulated Rayleigh scattering.
8.2 Electron-electron scattering effects in an infrared-coupled n-doped QW with two conduction subbands

Consider a quasi-two dimensional electron system in an n-doped QW. The Hamiltonian of such a system has the following form:

\[ H_0 = \sum_{i,k} E^c(i,k) a_{i,k}^\dagger a_{i,k} + \sum_{i,j,j',k,k'} V_{ii'jj'}(q,\omega') a_{i',k}^\dagger a_{j,k}^\dagger a_{j',q} a_{i,k}. \]  \hspace{1cm} (8.1)

Here \( V_{ii'jj'}(q,\omega') \) refers to the various electron-electron scattering processes which can occur in the n-doped QW. \( \hbar \omega' \) refers to the transfer of energy by such processes. In the two-conduction-subband system considered in this chapter we can decompose these processes into the following terms:

1. \( V_{1111} \) (\( V_{2222} \)) is the Coulomb interaction associated with the scattering of electrons within the first (second) subband. Here initial and final states of the scattered electrons are confined in one subband. This means that the intrasubband electron-electron scattering processes cause intrasubband transitions. These processes are responsible for the energy and population redistributions of electrons within a subband.

2. \( V_{1122} \) and \( V_{2211} \) are related to the Coulomb interactions of electrons in the first subband with those in the second subband. Here the initial and final transition states of an electron are all within one subband. In other words, the Coulomb intersubband interaction results in intrasubband transitions. This type of interaction causes energy transfer between subbands without transferring the electrons. \( V_{1122} \) and \( V_{2211} \) are responsible for excitonic effects in the intersubband transitions of n-doped QW's.

3. \( V_{2112} \) and \( V_{1221} \) are related to the processes where electrons in different subbands interact with each other and exchange their subband indices. In other words, intersubband Coulomb interaction causes intersubband transitions. Here also there
is no net population transfer, but energy transfer may occur. These processes are responsible for depolarization effects in the intersubband transitions of n-doped QW's.

4. $V_{2121}$ ($V_{1212}$) is associated with the scattering of two electrons with initial states in the first (second) subband and final states in second (first) subband. Here the Coulomb intrasubband transition cause intersubband transitions. Here both energy and population can be transferred.

Let us now consider the interaction of such a QW with an intense IR field. This field is considered to be nearly resonant with the transition between the first and the second conduction subbands (1-2) and quite off-resonant with all other transitions. We assume that in the absence of the IR field only the ground subband is occupied, with a Fermi-Dirac distribution. In the rotating-wave and dipole approximations the coupling of this electronic system with the IR field, $E(t) = E e^{-i\omega t}$, polarized along the QW growth direction, $z$, can be described by the Hamiltonian

$$H_{\text{int}} = \hbar \sum_k \{ \mu_{12} E(t) a_{1,k}^\dagger a_{2,k} + \mu_{12}^* E^*(t) a_{2,k}^\dagger a_{1,k} \}. \quad (8.2)$$

Here $\mu_{12}$ is the electric dipole moment for the transition between the first and second conduction subbands along the $z$ direction, which is assumed to be $k$-independent.

The density matrix of this system in the presence of the IR field can be obtained from

$$\frac{\partial \rho_k^k}{\partial t} = -\frac{i}{\hbar} [H_0 + H_{\text{int}}, \rho_k^k]. \quad (8.3)$$

Substitution of Eqs. (8.1) and (8.2) in this equation and including the terms corresponding to electron-phonon interaction to the Hamiltonian lead to the most general forms of the equations of motion of the system. Such a treatment has already been developed extensively for the cases involving interband transitions [88, 89, 90]. In general, the electron-electron and electron-phonon interaction terms in the systems involving interband or intersubband transitions are responsible for coherent and incoherent processes.
The coherent contributions lead to renormalization of the Rabi frequency (internal field terms) and the subband energies. The latter is caused by the Hartree energy and the real part of the exchange contributions. These two contributions are caused by the electrostatic Coulomb interaction among spatially separated electrons in different states and by Pauli-exclusion, respectively. In Ref. [88] Schmitt-Rink et al have shown that in the presence of high excitation the effects of the internal field can be insignificant. Therefore since in this chapter we also deal with highly doped QW's, the Coulomb screening is strong and we can ignore the internal field effects. The Hartree energies and exchange effects, however, are not smeared out by screening. In the absence of the infrared field when all electrons are in the ground subband the exchange effect decreases the energy of this subband. This enhances the transition energy between this subband and an upper one. Under the same condition the Hartree energy increases the subband energy, therefore decreases the transition energy.

In addition to these effects, the excitonic effects can also decrease the subband transition energies. The overall effects of excitons and those caused by the Hartree and exchange terms are around several meV. Also one may destroy the effect of the other, to some extend. For example, Bloss in Ref. [92] has shown that the Hartree and exchange effects in a GaAs/Al$_{0.3}$Ga$_{0.7}$As QW with 8.5 well width and $10^{12}$ cm$^{-2}$ density can cause $\sim$1.5 meV increase in the first to second conduction subband transition energy. The exciton effects, however, decrease this energy by about 2 meV (note these effects are caused by the Coulomb interaction between electrons in the first conduction subband and the photo-excited electrons in the upper subband). Based on these and the fact that in this chapter we are dealing with broad subbands, we do not expect that the overall effects of Hartree, exchange and excitons to be significant in the coupling process. On the other hand the depolarization effect caused by the collective presence of the electrons tends to enhance the transition energy significantly. This effect, however, depends on the total
density of the electrons [92]. However, since the electron density remains constant during the coupling process, we do not expect that the depolarization effect plays a major dynamic role. Its effects can, however, be included in the detuning of the IR field. One should note that this can only be a first order consideration. One may find some effects due to removal of some of the electrons from one subband to another by the intense IR field.

The incoherent contributions of the interaction processes lead to the damping of the system. In the following we treat the effects caused by electron-phonon scattering in a phenomenological way. Those caused by electron-electron scattering, however, are described by the Boltzmann equation. In this chapter we numerically solve this equation when it is coupled to optical Bloch equations due to the presence of an intense infrared field. This is done within the Born approximation including the screening of the Coulomb potential in dynamical random-phase approximation (RPA).

8.3 Optical Bloch and Boltzmann equations

Coupled optical Bloch and Boltzmann equations have already been solved by others to tackle various issues. For example, Binder et al have solved these equations to study the evolution of quasi-holes and steady-state non-equilibrium electron distribution functions in highly inverted bulk semiconductors [86]. Also by solving the Boltzmann and rate equations Lee et al have studied the time evolution of non-equilibrium electron distribution functions in the first and second conduction subbands in an n-doped QW after excitation by an infrared pulse [75]. They have shown how such functions become Fermi-like with time. Also in Ref. [91] Lyo has calculated relaxation rates of quasi-holes by solving the Boltzmann equation numerically for electrons and holes with Fermi distributions. In the following we apply the coupled optical Bloch and Boltzmann equations to a two-subband n-doped QW. The QW interacts with an intense infrared laser polarized...
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along its growth direction. Within the approximation discussed in the preceding section, for highly doped samples (electron density larger than $10^{11}$ cm$^{-2}$) the optical Bloch equations have the following forms:

$$\frac{d\rho_{11}^k}{dt} = -i\Omega_{12}(\rho_{21}^k - \rho_{12}^k) + \frac{d\rho_{11}^k}{dt} \mid_{\text{le-e}} + \frac{d\rho_{11}^k}{dt} \mid_{\text{lincoh}} + \sum_{k'} P_{kk'}, \quad (8.4)$$

$$\frac{d\rho_{22}^k}{dt} = i\Omega_{12}(\rho_{21}^k - \rho_{12}^k) + \frac{d\rho_{22}^k}{dt} \mid_{\text{le-e}} + \frac{d\rho_{22}^k}{dt} \mid_{\text{lincoh}}, \quad (8.5)$$

and

$$\frac{d\rho_{21}^k}{dt} = -i[\Delta_k - i\gamma_{12}^p](\rho_{21}^k - \rho_{12}^k) + \frac{d\rho_{21}^k}{dt} \mid_{\text{lincoh}}. \quad (8.6)$$

Here $\Omega_{12}$ is the Rabi frequency of the IR field and $\Delta_k$ is its detuning given by

$$\Delta_k = \frac{E^c(2, k)}{\hbar} - \frac{E^c(1, k)}{\hbar} - \omega. \quad (8.7)$$

In Eq. (8.4) the Boltzmann equation for the first subband is given by [86]:

$$\frac{d\rho_{11}^k}{dt} \mid_{\text{le-e}} = \Gamma_{1}^\text{in}(k, \rho_{11}^k)(1 - \rho_{11}^k) - \Gamma_{1}^\text{out}(k, \rho_{11}^k)\rho_{11}^k, \quad (8.8)$$

where $\Gamma_{1}^\text{in}(k, \rho_{11}^k)$ and $\Gamma_{1}^\text{out}(k, \rho_{11}^k)$ are the scattering in and out rates at $|1, k>$ due to the electron-electron scattering process. Within Born approximation they are given by

$$\Gamma_{1}^\text{in}(k, \rho_{11}^k) = \frac{4\pi}{\hbar} \sum_{p, q} \rho_{11}^p \rho_{11}^{k-q} (1 - \rho_{11}^{p-q}) |V_1(q, \omega')|^2 \times \delta(E^c(1, k) + E^c(1, p - q) - E^c(1, k - q) - E^c(1, p)), \quad (8.9)$$

and

$$\Gamma_{1}^\text{out}(k, \rho_{11}^k) = \frac{4\pi}{\hbar} \sum_{p, q} (1 - \rho_{11}^p) \rho_{11}^{k-q} (1 - \rho_{11}^{k-q}) |V_1(q, \omega')|^2 \times \delta(E^c(1, k) + E^c(1, p - q) - E^c(1, k - q) - E^c(1, p)). \quad (8.10)$$

Here $V_1(q, \omega')$ is the screened electron-electron interaction given by

$$V_1(q, \omega') = \frac{v_q F_1(q)}{\epsilon_1(q, \omega')}, \quad (8.11)$$
where \( v_q = \frac{2\pi e^2}{S q e_b} \), \( e \) is the electric charge, \( S \) the area of the QW, and \( e_b \) is the bulk dielectric constant which is assumed to be the same in the QW and in the barrier. Also \( \hbar \omega' = E_c^\ast(1, k - q) - E_c^\ast(1, k) \), and the form factor \( F_1(q) \) is given by

\[
F_1(q) = \int \int |\phi_1(z)\phi_1(z')|^2 e^{-q|z - z'|} dz dz'.
\] (8.12)

Here \( \phi_1(z) \) is the confinement wave function for the first conduction subband. Within the RPA the dynamical screening due to electron gas is given by (see [87] and [86])

\[
\epsilon_1(q, \omega) = 1 + v_q F_1(q) \chi_1(q, \omega),
\] (8.13)

where

\[
\chi_1(q, \omega) = 2 \sum_{p, \delta = 0} \frac{\rho_{11}^p - \rho_{11}^{p-q}}{E_c^\ast(1, p - q) - E_c^\ast(1, p) + \hbar \omega - i\delta}.
\] (8.14)

In general the contribution of the electron-electron scattering process to the dephasing rates of the system polarization or non-diagonal elements of the density matrix is determined according to [81], [82]

\[
\frac{\partial \rho_{ij}^k}{\partial t}|_{e-e} = \sum_{k'} (\gamma_{ij}^{kk'} \rho_{ij}^{k'} - \gamma_{ij}^{k'k} \rho_{ij}^{k'}) \quad i \neq j.
\] (8.15)

Here the first and second terms on the right side represent the non-diagonal and diagonal scattering rates of polarization, respectively. Eq. (8.15) shows that the decay of polarization at \( k \) depend not only on its loss at this wave-vector, but also on the decay of polarization at other states with different wave-vectors (\( k' \)). In the limit of high electron densities, however, one can neglect this term due to strong screening, as shown in Ref. [81]. Therefore in Eq. (8.6) we consider only the diagonal term as given by:

\[
\frac{\partial \rho_{21}^k}{\partial t}|_{e-e} = -\gamma_{12}^{e=e}(k) \rho_{21}^k.
\] (8.16)

Here \( \gamma_{12}^{e=e}(k) \) is

\[
\gamma_{12}^{e=e}(k) = \frac{1}{2} [\Gamma_1^{out}(k) + \Gamma_1^{in}(k)].
\] (8.17)
Note that in the above discussion we ignored the electron-electron scattering processes in the second subband,
\[
\frac{d\rho_{22}^k}{dt}\big|_{\text{e-e}} = 0.
\] (8.18)

This was justified based on the assumption that the upper subband is separated from the ground subband by an energy much larger than those of LO-phonons. Therefore, the former is much closer to the conduction band edge than the latter. As a result the states in the upper subband are much more extended than those in the ground subband. For example, in the structure studied in this chapter (GaAs/Ga\textsubscript{0.7}Al\textsubscript{0.3}As quantum well structure with 6.5 nm well width) the upper subband is far from the continuum edge by an energy less than 40 meV while the ground subband is separated from this edge by about 190 meV. Note that under these conditions the electrons in the upper subband undergo very fast intersubband transitions by emitting LO-phonons.

Returning to Eqs. (8.4) and (8.5), the terms \(\frac{d\rho_{22}^k}{dt}\big|_{\text{e-p}}\) represent the electron-phonon scattering process' contribution to the damping of the electronic system. As was discussed in the preceding chapter (Eqs. (7.9)-(7.12)), since the electrons in the ground subband are scattered by acoustic phonons, we have
\[
\frac{d\rho_{11}^k}{dt}\big|_{\text{e-p}} = \frac{d\rho_{11}^k}{dt}\big|_{\text{diff}}. 
\] (8.19)

For the second subband, electron-LO-phonon scattering lead to
\[
\frac{d\rho_{22}^k}{dt}\big|_{\text{e-p}} = -\Gamma_{22}^p\rho_{22}. 
\] (8.20)

In Eq.(8.4) \(P_{kk'}\) is the scattering-in rate of \(|1, k\rangle\) due to relaxation of electrons out of states in the second conduction subband with wave-vector \(k'\). This term includes both the intersubband transition, and the subsequent cascade in the ground subband due to LO-phonon decay. Since here the electron probability is calculated in each subband, in contrast to previous studies which only calculated \(\rho_{22} - \rho_{11}\) [16, 73], it is crucial to consider such a term. In general Eq. (8.4) can have a complicated form since \(P_{kk'}\) couples
states with different $k$'s. However, in the case where the energy spacing of the subbands is nearly equal to an integer number of LO-phonon energies ($E^c(2, k) - E^c(1, k) = n\hbar\Omega_{LO}$) the problem becomes tractable. In this case we can impose the condition that

$$\rho_{11}^k + \rho_{22}^k \sim \rho_{11}^{0k}. \quad (8.21)$$

Here $\rho_{11}^{0k}$ is the Fermi-Dirac distribution of the ground subband in the absence of any IR field. This yields $P_{kk'} \sim \Gamma_2^p \rho_{22}^{kk'} \delta_{kk'}$. Note that in a more realistic approach one may relax the condition of Eq. (8.21) and $E^c(2, k) - E^c(1, k) = n\hbar\Omega_{LO}$. This alters the distribution functions calculated in this chapter to some extent. The basic features discussed here, such as non-Fermi-Dirac distribution functions, $k$-dependent intersubband excitation, and field-dependent polarization dephasing rates, however, remain valid.

To study the linear response of the system discussed above, we consider an optical probe field with the same polarization as the strong field, similar to those considered in the previous chapters. The absorption spectrum of the weak probe field is found to have the form

$$A(\omega_p) = Re \sum_k \rho_{11}^k < [\hat{p}_k^+(t'), \hat{p}_k^-(z, t')] > |_{z=i\omega_p}, \quad (8.22)$$

where under steady state conditions

$$< [\hat{p}_k^+(\infty), \hat{p}_k^-(z)] > = \mu_0^2 \{\rho_{21}^{k1}(\infty)[R_{32}^k(z) - R_{31}^k(z)] - R_{33}^k[\rho_{11}^{k}(\infty) - \rho_{22}^{k}(\infty)]\}. \quad (8.23)$$

Here $R_{ij}^k(z)$ are the elements of the matrix

$$R^k(z) = \begin{pmatrix}
  z + \Gamma_2^p & 0 & -i\Omega & i\Omega \\
  0 & z + \Gamma_2^p & i\Omega & -i\Omega \\
  i\Omega & -i\Omega & z - i[\Delta_k + i(\gamma_{12}^{e-e}(k) + \gamma_{12}^p)] & 0 \\
  -i\Omega & i\Omega & 0 & z + i[\Delta_k - i(\gamma_{12}^{e-e}(k) + \gamma_{12}^p)]
\end{pmatrix}^{-1}. \quad (8.24)$$
8.4 Electron-electron scattering effects on the intersubband polarization dephasing rates in the weak-field regime

In this section we study how the electron-electron scattering process contributes to the polarization dephasing rate $\gamma_{12}^{e-e}(k)$ in the weak-field limit. In this limit, depletion of the ground subband population by the IR field is ignored, and $\gamma_{12}^{e-e}(k) = \frac{1}{2}(\Gamma_1^{\text{out}} + \Gamma_1^{\text{in}})$ is evaluated using the zero-field population. Here $(\Gamma_1^{\text{out}} + \Gamma_1^{\text{in}})$ is the damping rate of quasi-holes (QH’s) or kinetic holes, generated by transitions out of the ground subband. To do this we put $\Omega = 0$ in Eqs. (8.4)-(8.6). Therefore our formalism becomes similar to that of Lyo’s [91], although he did not use realistic structures for temperatures above 0 K.

We first consider an n-doped GaAs/Al$_x$Ga$_{1-x}$As QW with 6.5 nm well width and $x=0.3$. The effective mass of the electrons is taken as 0.07$m_0$ ($m_0$ is the free electron mass), and the electron density is $7 \times 10^{11}$ cm$^{-2}$ ($\epsilon_f=24$ meV at 0 K). In Fig.8.1 we show $\gamma_{12}^{e-e}(k)$ for various temperatures. For T=4 K, $\gamma_{12}^{e-e}(k)$ decreases from a maximum value at $k=0$ to $\sim 0$ at $k = k_f=0.21$ nm$^{-1}$ and then increases for $k > k_f$. This feature, which has been predicted by Binder for bulk semiconductors, becomes less significant at higher temperatures. At 100 and 200 K (dashed and dotted lines) $\gamma_{12}^{e-e}(k)$ is significantly higher for all $k$. The reason is that at these temperatures the Fermi distributions are expanded over larger number of states. This allows more extensive scattering processes and causes enhancement of $\gamma_{12}^{e-e}(k)$. For T=0 K (not shown) the damping rate is very close to that shown for T=4 K, for $k < k_f$.

Next we examine how $\gamma_{12}^{e-e}(k)$ changes with conduction band offset. We consider $\Delta E_c = 107$ meV and $\Delta E_c = 213$ meV, which correspond to $x=0.15$ and $x=0.3$ respectively, and $\Delta E_c = 10$ eV. The last case is not realistic but is an approximation to the infinite well model used by others [91]. The material structure is the same as that of Fig.8.1 with the same electron density and well width. The temperature is 4 K. Fig.8.2
Figure 8.1: Polarization dephasing rates caused by electron-electron scattering for various temperatures for Al$_{0.3}$Ga$_{0.7}$As with 6.5 nm well width. Density of electrons is $7 \times 10^{11}$ cm$^{-2}$[Ref. [24]].
shows that for states far from the Fermi edge, increasing the value of the offset causes an increase in $\gamma_{12}^{e-e}(k)$. This is because as the band offset increases, the electrons become more confined and the strength of their interaction with other electrons increases.

Figure 8.3 shows the effect of electron density on $\gamma_{12}^{e-e}(k)$ at 4 K, using the same structure as in Fig.8.1. All of the densities are chosen such that the Fermi energy is less than the LO-phonon energy ($n=5\times10^{11}$ cm$^{-2}$ with $\varepsilon_f=17$ meV; $n=2\times10^{11}$ cm$^{-2}$ with $\varepsilon_f=6.8$ meV). As the electron density decreases $\gamma_{12}^{e-e}(k)$ is significantly reduced over a large range of $k$. In the vicinity of the Fermi energy, however, there is a slight increase in the dephasing rate.
Figure 8.3: Polarization dephasing rates caused by electron-electron scattering for various electron densities for Al$_{0.3}$Ga$_{0.7}$As with 6.5 nm well width at T=4 K. All other specifications are the same as those of Fig.8.1[Ref. [24]].
8.5 Field-dependent electron-electron scattering effects in the intersubband transition of quantum wells and steady-state non-equilibrium distribution functions

To study the electron-electron scattering effects in the intersubband transition of a QW under an intense IR field, we assume that this field does not vary significantly on the scale of the system’s characteristic dephasing time, allowing steady-state solution of Eqs. (8.4)-(8.6). From Eqs. (8.4) and (8.5),

\[
\frac{d\rho_{11}^k}{dt} + \frac{d\rho_{22}^k}{dt} = \frac{d\rho_{11}^k|\text{le-e}}{dt} + \frac{d\rho_{11}^k|\text{incoh}}{dt} + \sum_{k'} P_{kk'} - \Gamma_{22}^p \rho_{22}^k. \tag{8.25}
\]

In the steady state, the right-hand side of Eq. (8.25) is zero. Since we consider \( P_{kk'} \sim \Gamma_{22}^p \rho_{22}^k \), therefore

\[
\frac{d\rho_{11}^k|\text{incoh}}{dt} + \frac{d\rho_{11}^k|\text{diff}}{dt} = 0. \tag{8.26}
\]

Using this equation we find a solution for Eqs. (8.4)-(8.6) which simultaneously satisfies Eqs. (8.9)-(8.10) for a given field intensity. To do this we use the equilibrium density matrix in Eqs. (8.9) and (8.10) to calculate \( \gamma_{12} \) (Eq. (8.17)). These are put into Eqs. (8.4)-(8.6) to calculate a new density matrix which reflects the presence of the optical field. The new density matrix was then put back in Eqs. (8.9)-(8.10) to calculate a new \( \gamma_{12} \). This cycle continues until the values are converged. Note that, as mentioned before, here we ignored the coherent effects caused by the electron-electron and polarization scattering processes. Therefore the distributions also do not reflect the effects of these processes. As shown in Ref. [89], however, one can self-consistently include these effects using a Monte-Carlo method.

To do the calculations we consider the same QW structure as used for Fig.8.1 with \( E^c(1,0)=55 \) meV and \( E^c(2,0)=199 \) meV, and take \( T=4 \) K, \( \Gamma_2^p =5 \) ps\(^{-1} \), and \( \Gamma_1^p=0.1 \) ps\(^{-1} \). The electron density is \( 7.4 \times 10^{11} \) cm\(^{-2} \). Fig.8.4a shows the ground state electron distribution for \( \Delta_k = 0 \). In the presence of an IR field such that \( \Omega_{12}=2 \) ps\(^{-1} \) (dashed lines), there is significant depletion of the initial Fermi distribution in \( \rho_{11}^k \). This depletion
is nonuniform in \( k \), however: states close to the Fermi energy are depleted more than others. As the field intensity is increased, \( \rho_{11}^k \) undergoes approximately linear depletion until \( \Omega_{12} \simeq 5 \) ps\(^{-1} \) (dotted line), where it begins to saturate. Note that now \( \Omega_{12} \) is close to the width of the second subband (\( \Gamma_2^P \)). Further increase in \( \Omega_{12} \) causes less change in \( \rho_{11} \), as shown. Note that the non-equilibrium distribution functions obtained here are in steady state. When the field intensity becomes zero we expect the electrons in the second subband decay very fast into the ground subband as the distribution in the latter becomes thermalized toward a Fermi distribution. The time evolution of a similar inverted system but with a wide well has been studied by Lee et al [75].

Fig. 8.4c shows the corresponding evolution of the electron distribution in the second subband. One can see that at low field intensity the electron distribution is not flat. As expected, increasing the infrared intensity causes the excited and ground states to become more equally populated. In the high-field limit where \( \Omega_{12} \gg \gamma_{12} + \gamma_{12}^P \) we have \( \rho_{11}^k = \rho_{22}^k = 0.5 \) for all \( k \).

The non-flatnesses of the electron distributions seen in Figs. 8.4a and 8.4c are results of the electron-electron scattering process in the ground subband. To see this in more detail, we show in Fig. 8.5 the dephasing rates of polarization corresponding to the distribution functions shown in these figures. The solid line in this figure shows the rate corresponding to the undisturbed Fermi distribution (\( \Omega_{12} = 0 \)). As the field intensity increases, the damping rates increase dramatically. For \( \Omega_{12} = 2 \) ps\(^{-1} \), for example, the rate is increased by more than a factor of two. In other words, the IR field extensively broadens the ground subband states via an enhancement of the electron-electron scattering process. This process saturates in the high-field limit, however.

The inset of Fig. 8.5 shows the separate contributions due to scattering in and out for \( \Omega_{12} = 10 \) ps\(^{-1} \). For a Fermi distribution at \( T = 4 \) K, \( \Gamma_1^{in} \) dominates for \( k < k_f \), while \( \Gamma_1^{out} \) dominates for \( k > k_f \). This is a direct result of the Pauli exclusion principle. For the
Figure 8.4: Electron distribution functions. The solid lines correspond to $\Omega_{12}=0$ ps$^{-1}$, dashed lines to $\Omega_{12}=2$ ps$^{-1}$, the dotted line to $\Omega_{12}=5$ ps$^{-1}$, and dashed-dotted line to $\Omega_{12}=10$ ps$^{-1}$. (a) and (b) show the distributions in the first subband for $\Delta=0$ and $\Delta=10$ ps$^{-1}$, respectively. (c) and (d) show the distributions in the second subband for the same respective cases. The electron density is $7\times10^{11}$ cm$^{-2}$ [Ref. [24]].
Figure 8.5: Polarization dephasing rates caused by electron-electron scattering corresponding to Fig.8.4a. Inset shows the contributions due to scattering in (dashed-dotted line) and scattering out (dashed line) for $\Omega=10$ ps$^{-1}$ [Ref. [24]].
case shown, however, this principle has less effect and both scattering terms contribute to the dephasing rate of a state in the ground subband. Such a state therefore becomes increasingly broad as the IR field intensifies. This process, however, depends very much on the density of electrons in the subband. At saturation, the electron density in the ground subband drops to half of its value in the uncoupled case, and the broadening reaches its maximum.

Now we consider the effect of detuning the infrared field on the intersubband transition and the electron-electron scattering process. We use the same structure as in Fig.8.4, but with $\Delta_k=10 \text{ ps}^{-1}$. As shown in Figs.8.4b and 8.4d, the essential effect of detuning is the expected reduction of the transition rates. For similar $\Omega_{12}$'s a smaller change in $\rho_{11}$ and $\rho_{22}$ is seen than for resonant coupling. There exists a distinct feature which was not expected, however. In Fig.8.4b we see that, in contrast to Fig.8.4a, when $\Omega_{12}=2$ and 5 ps$^{-1}$ the distribution is less depleted near the Fermi edge than away from it. Therefore in Fig.8.4d, the distribution has a shoulder close to $k_f$, rather than a peak as in Fig.8.4b. This can be understood by noting that when the energy spacings are similar, the detuning of a field has much more effect on transitions which are associated with smaller dephasing rates or narrower transition states. Therefore, in detuned coupling conditions, if the field intensity is low enough that the k-dependence of $\gamma_{12}^{e-e}$ is significant, the states close to $k_f$ should show less excitation than those with $k \sim 0$. 
Chapter 9

Coherent effects in infrared-coupled three-subband systems

9.1 Introduction

In chapter 7 we discussed how one can manipulate the intersubband transitions between the ground and upper subbands of n-doped QW's. In chapter 8 we included the effect of the electron-electron scattering processes when a laser field caused extensive electron excitations. This was done in a simple two-subband system. Many interesting nonlinear effects, however, occur in systems where two strong fields mix the ground and two upper subbands. Here depending on frequencies of the fields and band structures we may have a $\Xi_d$ or a $\Lambda_d$ configuration (see Figs.9.1 and 9.6). These systems can lead to coherent control of the electron populations in the conduction subbands. As shown in this chapter this can lead to coherent population trapping (CPT) where most of the conduction band population is coherently trapped in the two lower subbands of the $\Lambda_d$ system. CPT and the nonlinear gains generated in this system can play a role in the development of mid- and far-infrared lasers in QW's. The $\Xi_d$ system, on the other hand, illustrates several new effects such as enhancement of the stimulated Rayleigh scattering, gain and absorption caused by transitions between multi-photon states, etc. Note that QW systems with two conduction band transitions have already been used to study second harmonic generation and frequency-sum processes [99, 100]. The results of this chapter have been published in Refs. [27, 28].
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Figure 9.1: Schematic diagram of a generic Ξ_3 system in an asymmetric QW. ω_1 and ω_2 refer to the coupling field frequency and ω_p to that of the probe field.

9.2 Optical Three-Conduction-Subband Mixing in n-doped Quantum Wells with a Ξ_3 configuration

We consider here an n-doped QW structure which contains three conduction subbands mixed by two IR fields polarized along its growth direction. The first field, with frequency ω_1, couples the partially populated ground subband (|1, k>) to the second subband (|2, k>), and the second field, with frequency ω_2, couples the second subband to the third one (|3, k>) forming a Ξ_3 configuration (see Fig.9.1). Similar to the previous discussions, to have the equations of motion we should write down the interaction Hamiltonian of this system. Considering the same level of approximations as those in chapter 8 and after doing some algebra the equations of motion of the system are obtained as follows:

\[
\frac{d\rho_{11}^k}{dt} = -i\Omega_{12}(\rho_{21}^k - \rho_{12}^k) + \frac{d\rho_{11}^k}{dt}\bigg|_{\text{incoh}} + \frac{\partial \rho_{11}^k}{\partial t}\bigg|_{\text{diff}} + \sum_{k'} P_{1kk'}^k
\]

\[
\frac{d\rho_{22}^k}{dt} = i\Omega_{12}(\rho_{21}^k - \rho_{12}^k) - i\Omega_{23}(\rho_{32}^k - \rho_{23}^k) - \Gamma_2^p\rho_{22}^k + \sum_{k'} P_{2kk'}^k
\]

\[
\frac{d\rho_{33}^k}{dt} = -i\Omega_{23}(\rho_{32}^k - \rho_{23}^k) - \Gamma_3^p\rho_{33}^k
\]

\[
\frac{d\rho_{12}^k}{dt} = i[\Delta_1^k + i\gamma_{12}^{p-e}(k) + i\gamma_{12}^{-e+p}(k)]\rho_{12}^k + i\Omega_{12}\rho_{13}^k + i\Omega_{12}(\rho_{11}^k - \rho_{22}^k)
\]
\[
\frac{dp_{31}^k}{dt} = -i[\delta^k - i\gamma_{13}^k \rho_{31}^k] - i\Omega_{23} \rho_{21}^k + i\Omega_{12} \rho_{32}^k
\] (9.5)

\[
\frac{dp_{23}^k}{dt} = i[\Delta_2^k + i\gamma_{23}^k \rho_{23}^k - i\Omega_{23} (\rho_{33}^k - \rho_{22}^k) - i\Omega_{12} \rho_{13}^k]
\] (9.6)

Here \(\Omega_{12} = -\mu_{12} E_1/\hbar\) and \(\Omega_{23} = -\mu_{23} E_2/\hbar\) are the Rabi frequencies of the fields in which \(E_1\) and \(E_2\) are the amplitudes of the IR fields with frequencies \(\omega_1\) and \(\omega_2\), respectively. \(P_{i^kk'}\) is population rate of \(|i, k>\) due to relaxation of electrons out of states in higher subbands with wave-vector \(k'\). The one-photon detunings, \(\Delta_1^k\) and \(\Delta_2^k\), and two-photon detuning, \(\delta^k\), in Eqs. (9.4)-(9.6) are given by:

\[
\Delta_1^k = \omega_2^k - \omega_1^k - \omega_1,
\] (9.7)

\[
\Delta_2^k = \omega_2^k - \omega_2^k - \omega_2,
\] (9.8)

\[
\delta^k = \omega_3^k - \omega_1^k - \omega_1 - \omega_2,
\] (9.9)

where \(\omega_i^k\) is \(E_c(i, k)/\hbar\). Because we assume the effective masses of the coupled subbands are equal we suppress the \(k\) indices for these detunings below.

In our system electrons in the upper subbands decay to lower energy subbands by emission of LO-phonons. These intersubband transitions leave some electrons in states with \(k > k_{LO}\). Such electrons undergo a series of very fast LO-phonon decays within the same subband (intrasubband transitions) until they reach a state with \(k < k_{LO}\). They may then undergo another intersubband decay (if they are in the second subband), or may be scattered by acoustic phonons and other electrons. This is reflected in Eqs. (9.1) and (9.2) by the terms \(P_{i^kk'}\) in which \(k\) and \(k'\) refer to the initial and final wave-vector of the electron. Similar to chapter 8, the problem becomes tractable when the energy spacing of the subbands is nearly equal to an integer number of LO-phonon energies \((E_c(2, k) - E_c(1, k)) = n\hbar\Omega_{LO}\) and \((E_c(3, k) - E_c(2, k)) = n'\hbar\Omega_{LO}\), \(n\) and \(n'\) are integers). Under these conditions the electron ends up in a state with the same wave-vector as it started with, and therefore \(P_{1^kk'} = \Gamma_1^p \rho_{22}^k \delta_{kk'}\) and \(P_{2^kk'} = \Gamma_2^p \rho_{33}^k \delta_{kk'}\). Then adding Eqs.
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(9.1)-(9.3) gives
\[
\frac{d\rho_{11}^k}{dt} + \frac{d\rho_{22}^k}{dt} + \frac{d\rho_{33}^k}{dt} = \frac{d\rho_{11}^k}{dt} |_{\text{e-e}} + \frac{d\rho_{11}^k}{dt} |_{\text{diff}}. \tag{9.10}
\]

In the steady state, therefore, Eq. (9.10) becomes equal to zero. Because of the very fast intrasubband LO-phonon decay, we can ignore the population of electrons in states with \( k > k_{LO} \). Then
\[
\rho_{11}^k + \rho_{22}^k + \rho_{33}^k \sim \rho_{11}^0. \tag{9.11}
\]

Imposing Eqs. (9.10)-(9.11) on Eqs. (9.1)-(9.6) linearizes the latter. They can then be written in the following compact matrix form
\[
\frac{d\Phi^k}{dt} = L^k \Phi^k + K^k = 0. \tag{9.12}
\]

Similar to Eqs. (5.50)-(5-52), \( \Phi^k \)'s here refer to the density matrix elements.

To study the linear absorption and gain spectra of the system discussed above we use linear response theory. To do this we consider an optical probe field with the same polarization as those of the strong fields but with tunable frequency. Using the formalism described in chapter 2, for the three possible transitions we obtain the absorption (or gain) spectrum from
\[
A_{\omega_p} = A_{12}(\omega_p) + A_{13}(\omega_p) + A_{23}(\omega_p), \tag{9.13}
\]

where
\[
A_{12}(\omega_p) = \mu_{12}^2 \text{Re} \sum_k \rho_{11}^k \{ [R_{8,1}^k(z_1) - R_{8,7}^k(z_1)]\rho_{21}^k + R_{8,8}^k(z_1)\rho_{11}^k - \rho_{22}^k \}
+ R_{8,4}^k(z_1)\rho_{31}^k - R_{8,3}^k(z_1)\rho_{23}^k, \tag{9.14}
\]

\[
A_{13}(\omega_p) = \mu_{13}^2 \text{Re} \sum_k \rho_{11}^k \{ [R_{3,2}^k(z_3)\rho_{21}^k + R_{3,3}^k(z_3)\rho_{11}^k
+ [R_{3,9}^k(z_3) + R_{3,7}^k(z_3)]\rho_{31}^k - R_{3,3}^k\rho_{33}^k - R_{3,8}^k(z_3)\rho_{32}^k \}, \tag{9.15}
\]

\[
A_{23}(\omega_p) = \mu_{23}^2 \text{Re} \sum_k \rho_{22}^k \{ [R_{2,2}^k(z_2)\rho_{22}^k + R_{2,3}^k(z_2)\rho_{12}^k - R_{2,1}^k(z_2)\rho_{32}^k
- R_{2,2}^k(z_2)\rho_{33}^k - R_{2,6}^k(z_2)\rho_{31}^k \}. \tag{9.16}
\]
Here \( z_1 = z - i\omega_1, z_2 = z - i\omega_2 \) and \( z_3 = z - i(\omega_1 + \omega_2) \). Also we have

\[
R_{ij}^k(z_l) = (z_l I - L^k)_{ij}^{-1},
\]

where \( l = 1, 2, \) and \( 3 \), and \( I \) refers to the identity matrix. Eqs. (9.14)-(9.17) are obtained considering the temporal width of the IR fields to be large compared to the typical dephasing time of the system.

### 9.3 Linear Absorption and Gain Spectra in the \( \Xi_d \) system

In the previous section we presented the formalism required to study the interaction of two IR fields with a quantum well with three conduction subbands. For the sake of practical application we confine our analysis to the case where the IR fields are generated by a single \( \text{CO}_2 \) laser, and the QW’s three conduction subbands are nearly equally separated from each other. This structure contains a 5 nm GaAs layer followed by a 4nm Ga0.9Al0.1As layer and a Ga0.6Al0.4As barrier. This structure satisfies the condition \( E_{12} = E_{23} = n\hbar\Omega_{LO} \) (\( E_{12} \) and \( E_{23} \) are the energies of the first to second and the second to third subband transitions). We assume that this structure is n-doped with an electron density of \( 7 \times 10^{11} \text{ cm}^{-2} \). Also we consider \( \mu_{12} = e \times 2.1 \text{ nm}, \mu_{23} = e \times 3.0 \text{ nm}, \mu_{13} = e \times 0.38 \text{ nm} \) \([96] \), and \( \Gamma_{2e-p} = 5 \text{ ps}^{-1} \) and \( \Gamma_{3e-p} = 10 \text{ ps}^{-1} \). Since here we consider only one field, we have \( \omega_1 = \omega_2 \) and \( \Omega_{12} \sim \Omega_{23} = \Omega \). The temperature of the QW is considered to be 4 K.

We now consider a weak probe field to detect the three possible transitions in the system. To do this we solve Eqs. (9.14)-(9.17). Here since the energy spacings between the 1-2 and 2-3 transitions are assumed to be nearly equal, their linear responses occur in the same range of frequencies. For the sake of clarity of discussion and generalization to systems where these two transitions have different energies, we show the response due to each transition, as well as the total response.

Consider the absorption spectra associated with the 1-2 and 2-3 transitions. As Fig.9.2a shows, in the presence of very weak fields (\( \Omega = 0.01 \text{ ps}^{-1} \)) we see that the 1-2
transition spectrum is a standard Lorentzian line and that the 2-3 transition does not contribute. For $\Omega = 1 \text{ ps}^{-1}$ (not shown), however, the spectrum of the 1-2 transition becomes broadened and suppressed in amplitude while that of the 2-3 transition starts to appear. For $\Omega = 5 \text{ ps}^{-1}$, as Fig.9.2b shows, we see the 1-2 transition spectrum becomes double-peaked (solid line). That of the 2-3 transition shows a similar evolution but at an earlier stage (dashed line). As we increase the Rabi frequencies the spectrum associated with the 1-2 transition develops further until it becomes negative in two regions. In fact, as shown in Fig.9.2c, for $\Omega = 40 \text{ ps}^{-1}$ this spectrum consists of two gain peaks at $-\sqrt{2}\Omega$ and $+\sqrt{2}\Omega$ and two dispersive features with zeros at $-2\sqrt{2}\Omega$ and $+2\sqrt{2}\Omega$ (solid line). On the other hand, the evolution of the spectrum associated with the 2-3 transition is very different. After an initial broadening and doubling the spectrum develops two absorption peaks at $\pm\sqrt{2}\Omega$ and two dispersive features, similar to those of the 1-2 transition, at $\pm2\sqrt{2}\Omega$ (dashed line). The only regions where the total spectrum shows gain are in the vicinity of the dispersion frequencies. The dotted lines in these figures represent the total response of the system.

To study the effect of detuning in these spectra we consider the case where $\Delta_1 = \Delta_2 = 10 \text{ ps}^{-1}$. In Fig.9.3a we show the spectra associated with the 1-2 and 2-3 transitions and their sum for $\Omega = 5 \text{ ps}^{-1}$. As one can see, the 1-2 transition spectrum here mainly consists of a strong peak with two small structures at lower frequencies (solid line). That of the 2-3 transition, however, mainly shows a broad peak with some small bumps (dashed line). For $\Omega = 20 \text{ ps}^{-1}$, however, both transition spectra develop into exotic shapes, as shown in Fig.9.3b. Here the spectrum of the 1-2 transition has two gain peaks, one at $\sim 40 \text{ ps}^{-1}$ and the other at $\sim 70 \text{ ps}^{-1}$, as well as two absorption peaks. Under the same conditions the 2-3 transition spectrum also shows two regions of gain and three of absorption.

By increasing the field intensity such that $\Omega = 40 \text{ ps}^{-1}$ one sees (Fig.9.3c) that the spectrum of the 1-2 transition has three gain peaks: two at frequencies lower than that
Figure 9.2: Linear absorption of the resonantly-driven quantum well for frequencies near $\omega_{12}$. The solid curves represent absorption due to the 1-2 transition. The dashed lines represent that due to the 2-3 transition, and the dotted ones to the total response. Rabi frequencies are: (1) $\Omega=0.01 \text{ ps}^{-1}$, (b) $\Omega=5 \text{ ps}^{-1}$, and (c) $\Omega=40 \text{ ps}^{-1}$ [Ref. [27]].
Figure 9.3: Linear absorption of the quantum well with $\Delta_1 = \Delta_2 = 10\, \text{ps}^{-1}$ for frequencies near $\omega_{12}$. The solid curves represent absorption due to the 1-2 transition. The dashed lines represent that due to the 2-3 transition, and the dotted ones to the total response. Rabi frequencies are: (1) $\Omega=5\, \text{ps}^{-1}$, (b) $\Omega=20\, \text{ps}^{-1}$, and (c) $\Omega=40\, \text{ps}^{-1}$ [Ref. [27]].
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of the 1-2 transition ($\sim 70$ ps$^{-1}$ and $\sim 120$ ps$^{-1}$) and one at a higher frequency ($\sim 45$ ps$^{-1}$). The spectrum also has an absorption peak at $\sim 105$ ps$^{-1}$. Under the same conditions the spectrum associated with the 2-3 transition shows only one region of gain. In addition to the three absorption peaks seen at positive frequencies, a new absorption peak is generated at a frequency below that of the 1-2 line center. As we will show in the following section, these features are drastically changed as one changes the detuning and/or damping rates of $|3\rangle$. This suggests fundamental differences between the 1-2 and 2-3 transitions.

9.4 Quantum Interference effects and Hyper-Raman transitions between one- and two-photon dressed states

In the preceding section we presented our results regarding the nonlinear effects induced in linear response near the 1-2, 2-3 and 1-3 transitions in an n-doped QW. In order to understand these results we adopt in this section a dressed-state picture. This picture is useful for examining the coupling mechanisms in the system and the origin of the gain and absorption spectra. For the sake of clarity and generality we consider here a resonant $\Xi_d$ system created by two IR fields with different frequencies. Based on this picture, in the presence of these fields but in the absence of interaction with the QW, the QW-field states at the energy of each subband are triply degenerate. At the energy of $|1, k\rangle$ these states are $|1, n\omega_1, n'\omega_2 >_k, |2, (n-1)\omega_1, n'\omega_2 >_k, and |3, (n-1)\omega_1, (n'-1)\omega_2 >_k$. At the energy of $|2, k\rangle$ the degenerate triplet states are $|1, (n+1)\omega_1, n'\omega_2 >_k, |2, n\omega_1, n'\omega_2 >_k, and |3, n\omega_1, (n'-1)\omega_2 >_k$, and at that of $|3, k\rangle$ they are $|1, (n+1)\omega_1, (n'+1)\omega_2 >_k, |2, n\omega_1, (n'+1)\omega_2 >_k, and |3, n\omega_1, n'\omega_2 >_k$. However, once the coupling between the IR fields and the QW are considered these degeneracies are removed and each state becomes a triplet. In the case of resonant coupling and $\Omega_{12} = \Omega_{23}$, $|1, k\rangle$ is substituted by three states as follows [35]:
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|1, t >_k = \frac{1}{\sqrt{2}} \left[ \frac{1}{\sqrt{2}} |1, n\omega_1, n'\omega_2 >_k + |2, (n-1)\omega_1, n'\omega_2 >_k + \frac{1}{\sqrt{2}} |3, (n-1)\omega_1, (n'-1)\omega_2 >_k \right], (9.18)

|1, s >_k = \frac{1}{\sqrt{2}} \left[ \frac{1}{\sqrt{2}} |1, n\omega_1, n'\omega_2 >_k - |2, (n-1)\omega_1, n'\omega_2 >_k + \frac{1}{\sqrt{2}} |3, (n-1)\omega_1, (n'-1)\omega_2 >_k \right], (9.19)

and

|1, r >_k = \frac{1}{\sqrt{2}} [-|1, n\omega_1, n'\omega_2 >_k + |3, (n-1)\omega_1, (n'-1)\omega_2 >_k] (9.20)

Under similar conditions for |2, k > we have

|2, t >_k = \frac{1}{\sqrt{2}} \left[ \frac{1}{\sqrt{2}} |1, (n+1)\omega_1, n'\omega_2 >_k + |2, n\omega_1, n'\omega_2 >_k + \frac{1}{\sqrt{2}} |3, n\omega_1, (n'-1)\omega_2 >_k \right], (9.21)

|2, s >_k = \frac{1}{\sqrt{2}} \left[ \frac{1}{\sqrt{2}} |1, (n+1)\omega_1, n'\omega_2 >_k - |2, n\omega_1, n'\omega_2 >_k + \frac{1}{\sqrt{2}} |3, n\omega_1, (n'-1)\omega_2 >_k \right], (9.22)

and

|2, r >_k = \frac{1}{\sqrt{2}} [-|1, (n+1)\omega_1, n'\omega_2 >_k + |3, n\omega_1, (n'-1)\omega_2 >_k], (9.23)

and for |3, k >

|3, t >_k = \frac{1}{\sqrt{2}} \left[ \frac{1}{\sqrt{2}} |1, (n+1)\omega_1, (n'+1)\omega_2 >_k + |2, n\omega_1, (n'+1)\omega_2 >_k + \frac{1}{\sqrt{2}} |3, n\omega_1, n'\omega_2 >_k \right], (9.24)

|3, s >_k = \frac{1}{\sqrt{2}} \left[ \frac{1}{\sqrt{2}} |1, (n+1)\omega_1, (n'+1)\omega_2 >_k - |2, n\omega_1, (n'+1)\omega_2 >_k + \frac{1}{\sqrt{2}} |3, n\omega_1, n'\omega_2 >_k \right], (9.25)

and

|3, r >_k = \frac{1}{\sqrt{2}} [-|1, (n+1)\omega_1, (n'+1)\omega_2 >_k + |3, n\omega_1, n'\omega_2 >_k]. (9.26)

Here |i, t >_k and |i, s >_k (i=1,2, and 3) are one-photon dressed states which are the results of mixing of all three subbands and |i, r >_k are the result of a two-photon dressing process between QW-field states associated with |1, k > and |3, k >.

Using this picture we can explain the spectra seen in the previous sections at high field intensities. First, consider the spectra shown in Fig.9.2c. The figure shows two gain
peaks and two dispersive features in the 1-2 transition spectrum. As shown in Fig. 9.4a the gain peaks are the results of three-photon processes. In each of these processes two pump field photons with frequency $\omega_1$ are absorbed and one photon is emitted with a frequency of either $\omega_1 - \sqrt{2} \Omega$ or $\omega_1 + \sqrt{2} \Omega$. It is important to note that these processes start from one-photon states $|1, t >_k$ and $|1, s >_k$ and end up in the two-photon state $|2, r >_k$, respectively. Therefore the gain peaks are the result of decay from $|2, t >$ and $|2, s >$ into $|1, r >$. This shows that the gain is not the result of inversion between $|1, k >$ and $|2, k >$ but rather between the dressed states. In fact it is caused by transition between $|2, n\omega_1, n'\omega_2 >_k$ and $|3, (n - 1)\omega_1, (n' - 1)\omega_2 >_k$. Such a gain process is an extension of the hyper Raman gain discussed in Ref. [24] for a two-subband system.

The story is rather different for the dispersive parts of the 1-2 transition spectrum. The transitions between the dressed states which are responsible for these features are shown in Figs. 9.4b and 9.4c. Here absorption of two pump field photons with frequency $\omega_1$ is accompanied by transitions between two one-photon states. For the photon emitted with frequency $\omega_1 - 2\sqrt{2} \Omega$ these states are $|2, t >_k$ and $|1, s >_k$ (Fig. 9.4b), while for the photon with frequency $\omega_1 + 2\sqrt{2} \Omega$ they are $|2, s >$ and $|1, t >$ (Fig. 9.4c). Based on Eqs. (9.18)-(9.23) such transitions involve transitions between various components of these states, some of which generate gain and some absorption. This causes quantum interferences, resulting in the dispersive features [33].

Now consider the spectrum associated with the 2-3 transition. As Fig. 9.2c shows this spectrum shows two absorption peaks at the frequencies where the 1-2 transition has gain. However, both transitions show dispersive features near $\pm 2\sqrt{2} \Omega$. As shown schematically in Fig. 9.4a, the absorption peaks in the 2-3 transition are associated with transitions between the two-photon state $|2, r >_k$, and two upper one-photon states, $|3, t >_k$ and $|3, s >_k$, with frequencies $\omega_2 - \sqrt{2} \Omega$ and $\omega_2 + \sqrt{2} \Omega$, respectively. In fact these processes can be considered to be a continuation of those considered for the 1-2 transition. Adding these two types of transitions together we see a transition between
Figure 9.4: Schematics of gain and absorption processes in a $\Xi_d$ system. (a) shows the hyper Raman process in the 1-2 transition, and pure absorption in the 2-3 transition. (b) shows the mechanisms causing dispersion features in the 1-2 and 2-3 transitions at $\omega_1 - 2\sqrt{2}\Omega$ and $\omega_2 + 2\sqrt{2}\Omega$, respectively, as does (c) at $\omega_1 + 2\sqrt{2}\Omega$ and $\omega_2 - 2\sqrt{2}\Omega$. Solid lines correspond to absorption/emission of the probe frequency. The direction of the arrows indicates whether the process is gain, absorption, or a mixture of the two [Ref. [27]].
\( |1, t >_k \) and \( |3, t >_k \) and between \( |1, s >_k \) and \( |3, s >_k \). The mechanisms responsible for the dispersive features seen in the 2-3 transition spectrum are very similar to those of the 1-2 transition. Here also the transitions associated with these features (Figs.9.4b and 9.4c) complete full transitions between the one-photon states \( |1, t >_k \) and \( |3, t >_k \) and between \( |1, s >_k \) and \( |3, s >_k \).

### 9.5 Extension of Stimulated Rayleigh Scattering, Coherent and non-Coherent Processes

Our results in Sect.9.3 showed that the responses associated with 1-2 and 2-3 transitions were different. In Sect.9.4 we explained that the absorption and gain spectra associated with these transitions at \( \sqrt{2\Omega} \) and \( -\sqrt{2\Omega} \) arise from three-photon and one-photon processes, respectively. Detuning of the field revealed other characteristic features of these two transitions. In fact the spectrum associated with the 1-2 transition under the conditions of Fig.9.3b shows some similarity to the corresponding one in a two-level atomic system (Mollow spectrum) or a two-subband QW system in chapter 8. Specifically, there is a dispersive feature at the frequency of the coupling field, with gain peaks on one side and absorption peaks on the other. This was not seen in the case of the 2-3 transition (see Figs.9.3b and 9.3c). In this section we discuss these characteristic features in more detail.

To gain some insight into the origin of this spectrum we consider the 1-2 transition of a system similar to that of Fig.9.3 but with \( \Delta_1 = \Delta_2 = 40 \text{ ps}^{-1} \) and \( \Gamma_3 = 40 \text{ ps}^{-1} \). This could be a QW structure where \( |3, k > \) is located in the continuum (a quasi-bound subband). For \( \Omega = 10 \text{ ps}^{-1} \) (Fig.9.5a) there is a large blue-shifted peak, a kink close to \(-40 \text{ ps}^{-1}\), and a very small dip at \(-80 \text{ ps}^{-1}\) (not shown). Under these conditions the response of the 2-3 transition is very small. For \( \Omega = 20 \text{ ps}^{-1} \), as shown in Fig.9.5b, the kink at \(-40 \text{ ps}^{-1}\) in the 1-2 transition spectrum (solid line) becomes a dispersive feature. Also the dip at \(-80 \text{ ps}^{-1}\) grows larger and becomes a gain peak. The response of the 2-3 transition...
now becomes significant (dashed line), however, we see here that at the frequency where the 1-2 transition presents a dispersive feature, the 2-3 transition shows a small negative dip. When we increase the field intensity such that $\Omega=40 \text{ ps}^{-1}$ the dispersive feature in the 1-2 transition remains unchanged in frequency and the small dip now becomes a strong gain peak shifted to $\sim 120 \text{ ps}^{-1}$ (Fig.9.5c). The spectrum associated with the 2-3 transition, however, shows that the small dip at -40 ps$^{-1}$ becomes stronger without any change in frequency. The dotted lines in Fig.9.5 show the superposition of the 1-2 and 2-3 transition spectra for the case where these two transitions have the same energies.

To understand these phenomena we note that the gain peaks seen at -40 ps$^{-1}$ have the characteristic feature that their frequency is the same as that of the coupling field, regardless of field intensity. On the other hand, the dips or gain seen at lower frequencies become red shifted as the field intensity increases. The former is characteristic of simulated Rayleigh scattering between the one- and two-photon dressed states associated with one subband and those of another subband. The latter phenomenon is characteristic of simulated Raman scattering (or hyper-Raman scattering) between these states. However, at -40 ps$^{-1}$ in the 1-2 transition, there is a quantum interference process between the gain and absorption transitions. The competition between these two processes can either completely diminish both or make one or the other transition dominant at different frequencies [28]. This results in the dispersive profiles. For the 2-3 transition, however, there exists no such interfering process and the Rayleigh gain is not coherently destroyed by any other transition. For this reason we see its effect as a sharp gain peak.

9.6 Coherent Population Trapping and Quantum Interference Features in Infrared-Driven n-doped QW’s with $\Lambda_d$ configurations

In the previous sections we studied various features of an IR-driven QW with a $\Xi_d$ configuration. The damping rate of the second subband in this system ($|2, k>$) was either equal or smaller than those of two other subbands. Because of this many interesting
Figure 9.5: Linear absorption of the quantum well with $\Gamma_3=40$ ps$^{-1}$ and $\Delta_1 = \Delta_2 = 40$ ps$^{-1}$; other parameters are the same as in Fig. 9.3. The solid curves represent absorption due to the 1-2 transition. The dashed lines represent that due to the 2-3 transition, and the dotted ones to the total response. Rabi frequencies are: (1) $\Omega=10$ ps$^{-1}$, (b) $\Omega=20$ ps$^{-1}$, and (c) $\Omega=40$ ps$^{-1}$ [Ref. [27]].
effects generated by multi-photon coupling processes were suppressed. In order to investigate some of these effects, in this section we present our results for a $\Lambda_d$ system (Fig. 9.6). Here the subband corresponding to the second subband in the $\Xi_d$ can be very broad.

The equations of motion of the $\Lambda_d$ system can be obtained taking steps similar to those in the previous chapters. Also to make the system dynamics tractable, like the case of $\Xi_d$ system, we consider a QW structure where the energies corresponding to the 1-3 and 2-3 transitions ($E_{13}$ and $E_{23}$) are integer multiples of the LO-phonon energy (Fig. 9.6). Specifically we consider a structure which contains a 3.8 nm GaAs layer followed by 8 nm Al$_{0.25}$Ga$_{0.75}$As, confined by Al$_{0.39}$Ga$_{0.61}$As barriers. In this structure $E_{13}=214$ meV and $E_{23}=73$ meV, both of which are nearly integer multiples of the LO-phonon energy. After some algebra we find the following expressions for the contributions of the 1-2, 2-3 and 1-3 transitions in the absorption spectra of the $\Lambda_d$ system:

\begin{align}
A_{12}^{\Lambda_d} (\omega_p) &= \mu_{12}^2 Re \sum_k \rho_{11}^k \{ [R_{3,9}^k(z) - R_{3,7}^k(z) - R_{3,8}^k(z)] \rho_{31}^k + R_{3,2}^k(z) \rho_{21}^k + \\
&\quad - R_{3,3}^k(z)[\rho_{11}^k - \rho_{33}^k] \} + R_{3,3}^k(z)[\rho_{11}^k - \rho_{33}^k]
\end{align}

\begin{align}
A_{23}^{\Lambda_d} (\omega_p) &= \mu_{23}^2 Re \sum_k \rho_{22}^k \{ [R_{4,1}^k(z') - R_{4,9}^k(z')] \rho_{23}^k + R_{4,4}^k(z') \rho_{33}^k + R_{4,8}^k(z') \rho_{13}^k - \\
&\quad - R_{4,4}^k(z') \rho_{22}^k - R_{4,5}^k(z') \rho_{23}^k \}
\end{align}

and

\begin{align}
A_{13}^{\Lambda_d} (\omega_p) &= \mu_{13}^2 Re \sum_k \rho_{11}^k \{ [R_{8,1}^k(z'') - R_{8,7}^k(z'')] \rho_{21}^k + [\rho_{11}^k - \rho_{22}^k] R_{8,8}^k(z'') + \\
&\quad - R_{8,4}^k(z'') \rho_{31}^k - R_{8,3}^k(z'') \rho_{23}^k \}
\end{align}

Here $z = i(\omega_p - \omega_1 + \omega_2)$, $z' = i(\omega_p - \omega_2)$ and $z'' = i(\omega_p - \omega_1)$. 

\[
A_{12}^{\Lambda_d} (\omega_p) = \mu_{12}^2 Re \sum_k \rho_{11}^k \{ [R_{3,9}^k(z) - R_{3,7}^k(z) - R_{3,8}^k(z)] \rho_{31}^k + R_{3,2}^k(z) \rho_{21}^k + \\
&\quad - R_{3,3}^k(z)[\rho_{11}^k - \rho_{33}^k] \}
\]

\[
A_{23}^{\Lambda_d} (\omega_p) = \mu_{23}^2 Re \sum_k \rho_{22}^k \{ [R_{4,1}^k(z') - R_{4,9}^k(z')] \rho_{23}^k + R_{4,4}^k(z') \rho_{33}^k + R_{4,8}^k(z') \rho_{13}^k - \\
&\quad - R_{4,4}^k(z') \rho_{22}^k - R_{4,5}^k(z') \rho_{23}^k \}
\]

\[
A_{13}^{\Lambda_d} (\omega_p) = \mu_{13}^2 Re \sum_k \rho_{11}^k \{ [R_{8,1}^k(z'') - R_{8,7}^k(z'')] \rho_{21}^k + [\rho_{11}^k - \rho_{22}^k] R_{8,8}^k(z'') + \\
&\quad - R_{8,4}^k(z'') \rho_{31}^k - R_{8,3}^k(z'') \rho_{23}^k \}
\]
Chapter 9. Coherent effects in infrared-coupled three-subband systems

9.6.1 Coherent population trapping in QW’s

Consider the structure described in above is n-doped with an electron density of $7.3 \times 10^{11}$ cm$^{-2}$. We found the transition dipole moments $\mu_{12} = e \times 1.02$, $\mu_{13} = e \times 0.92$, and $\mu_{23} = e \times 2.55$ nm. The damping rates of $|2, k>$ and $|3, k>$ are considered to be $\Gamma_2^p = 5$ ps$^{-1}$ and $\Gamma_3^p = 30$ ps$^{-1}$, respectively. The effective mass for each layer is obtained from $m^* = [0.067(1 - x) + 0.15x]m_e$ where $x$ is the fraction of Al present and $m_e$ is the free electron mass [97]. The temperature of the QW is considered to be 4 K.

Figure 9.7a shows the evolution of $\rho_{ii}^k$ ($i = 1, 2$ and 3) at $k = k_f$ for $\Omega_{13} = \Omega_{23} = 20$ ps$^{-1}$ (corresponding to IR intensities of 113 and 14.9 MW/cm$^2$, respectively), $\Delta_2 = 0$, and various $\Delta_1$. Here $\Omega_{jj} \equiv -\mu_{jj}E_j/\hbar$ is the Rabi frequency and $\Delta_j$ is the detuning of the coupling field with frequency $\omega_j$. For $|\Delta_1| \gg 0$, the system dynamics are dominated by one-photon transitions of electrons into the second and third subbands and energy relaxation of electrons from the latter to the former. As a result, the electron populations reflect the subbands’ widths and energies. As $|\Delta_1|$ decreases, however, the population in the third subband becomes depleted despite the enhancement of the 1-3 coupling strength.
(dotted line). The ground subband population (solid line) increases, and when $\Delta_1=0$ most of the electron population is divided between the ground and second subbands.

To understand these results we recall that the coupling mechanism in a $\Lambda_d$ system is one-photon coupling of three subbands and two-photon coupling of the first and second subbands. Here one can construct the dressed states associated with these coupling processes similar to those in the $\Xi_d$ (Eqs. 9-18 to 9-26). Here since for the system of Fig.9.7a the intermediate state $|3,k>$ is relatively broad, when $|\Delta_1| \gg 0$ both forms of coupling are suppressed, and transitions between pairs of states are dominant. When $\Delta_1$ becomes close to zero, however, two-photon coupling becomes more significant and the population is transferred into $|1,r>_k$ and $|2,r>_k$, i.e. coherent population trapping (CPT) occurs [36]. The breadth of the intermediate state has a strong effect on the efficiency of CPT, since it causes suppression of one-photon coupling compared to two-photon coupling.

Fig.9.7b shows the complete electron distributions in the three coupled conduction subbands for $\Omega_1 = \Omega_2 = 0$ and $\Omega_{13} = \Omega_{23} = 20$ ps$^{-1}$. Under these conditions the populations in the first and second subbands are virtually equal, each being over 40% of the total. The slight non-flatness in the distributions is caused by the $k$-dependent effect of electron-electron scattering on the intersubband dephasing rates (see Fig.9.9). The scattering is enhanced here since the ground subband distribution is significantly different from the Fermi distribution.

9.6.2 Linear absorption spectra of a quantum well with a $\Lambda_d$ configuration

In the preceding subsection we studied the electron distributions of an IR-coupled QW system with a $\Lambda_d$ configuration. In this section we investigate the linear absorption spectra (Eqs. (9.26)-(9.28)) of this system when $\Omega_{13} = \Omega_{23} \equiv \Omega$. The structure considered is the same as in the preceding section.

As Fig.9.8a shows, when $\Omega$ increases from 1 to 5 ps$^{-1}$ the absorption due to the 1-2
Figure 9.7: Electron distributions in a $\Lambda_d$ system with $\Omega_{13} = \Omega_{23} = 20$ ps$^{-1}$. (113 MW/cm$^2$ at frequency $\omega_1$ and 14.9 MW/cm$^2$ at $\omega_2$) The solid, dashed, and dotted lines represent the populations in the first, second, and third subbands, respectively. (a) The effect of detuning one of the coupling fields on the Fermi level populations ($k = k_f$). (b) The complete electron distributions in the resonant coupling case. The electron density is $7.3 \times 10^{11}$ cm$^{-2}$ [ref. [28]].
transition is strongly suppressed. For $\Omega=20$ ps$^{-1}$ (the condition of Fig.9.7a) and higher the absorption virtually vanishes. This strong suppression occurs because most of the electron population is equally distributed between the first and second subbands due to CPT (Fig.9.7b).

The effects of CPT are seen in other parts of the absorption spectrum as well. At $\Omega = 5$ ps$^{-1}$, the contribution due to the 1-3 transition becomes spectrally doubled, as Fig.9.8b shows. For higher field intensities the two peaks become smaller and move away from the central frequency. These changes occur because when CPT happens, the ground population decreases from a Fermi distribution to a non-Fermi one with approximately 40% of the total population (Fig.9.7b). This tends to suppress the 1-3 absorption, but since the third subband is simultaneously losing its population to the other subbands, the absorption due to this transition remains noticeable. The doubling seen here is the result of transitions between the two-photon state in the ground subband ($|1, r >_k$) and the two one-photon states in the third subband ($|3, s >_k$ and $|3, t >_k$). Under resonant coupling, $|1, k >_r$ has the same energy as the uncoupled ground subband, and $|3, s >_k$ and $|3, t >_k$ have energies $\hbar \sqrt{\Omega^2_{13} + \Omega^2_{23}}$ above and below that of the uncoupled third subband, respectively. The one-photon states have less population than the two-photon state due to the dominance of two-photon coupling.

The 2-3 transition's contribution (Fig.9.8c) shows features which reflect the evolution of the 1-3 transition's dephasing rate ($\gamma^{\epsilon-\epsilon}_{13}(k)$) with varying field intensity. For $\Omega=1$ ps$^{-1}$ (inset), the spectrum shows a well-resolved LIT effect. As the field intensities increase the transparency hole diminishes and then is replaced by another hole with a small peak at its center. To explain these phenomena we show in Fig.9.9 the evolution of $\gamma^{\epsilon-\epsilon}_{13}(k)$ for various field intensities. For $\Omega=1$ ps$^{-1}$, $\gamma^{\epsilon-\epsilon}_{13}$ is small, especially for states with $k$ close to $k_f$. The coupling between the first and third subbands is therefore much stronger than that between the third and second. As a result, when one probes the 2-3 transition one sees a LIT effect similar to those studied in atomic or QW systems. In such systems a
Figure 9.8: Linear absorption of a weak probe field in a resonantly-coupled $\Lambda_d$ system. Parts (a), (b), and (c) show, respectively, the contributions due to the 1-2, 1-3, and 2-3 transitions. The Rabi frequencies corresponding to each line are solid: $\Omega=1$ ps$^{-1}$, dashed: $\Omega=5$ ps$^{-1}$, dotted: $\Omega=10$ ps$^{-1}$, dash-dotted: $\Omega=20$ ps$^{-1}$, dash-dot-dotted: $\Omega=40$ ps$^{-1}$ [Ref. [28]].
narrow or metastable state is coupled into the upper level of a transition. When this transition is detected by a probe field, a transparency hole is generated in its absorption spectrum.

Fig.9.9 shows that when $\Omega$ increases $\gamma_{13}^{e-e}$ becomes larger, and therefore the ground subband is no longer narrow. The fields then tend to couple the first and second subbands to the third one with comparable strengths. This establishes the two-photon coupling and CPT. Near $\Omega=5$ ps$^{-1}$ the mechanism of doublet generation in the 2-3 transition spectrum (Fig.9.8c) changes from weak detection of a strongly driven two-subband system, as discussed above, to transitions between multi-photon dressed states. As in Fig.9.8b, the doublet here is the result of weak detection of the two one-photon states from the two-photon state in the second subband ($|2, k\rangle$). The small central peak in the latter case might be the result of electron distribution evolution and vanishes the high field limit.
9.6.3 Field intensity effects on the coherent processes of intersubband transitions

The effects studied in the previous sections were associated with equal Rabi frequencies. One feature of the $\Lambda_d$ system, however, is that one can vary the frequencies and intensities of the fields without violating the assumptions of the theory. This allows one to explore the rich variety of nonlinear effects caused by quantum interference. In this section we study the case where $\Omega_{13} \neq \Omega_{23}$ but the fields remain resonant with the corresponding transitions. One significant effect of this is field-coherence destruction effect. This effect which was discussed in chapters 4 and 7, indicates large or small values of $\Omega_{23}/\Omega_{13}$ cause one state to be decoupled from the rest of the system. In fact the stronger field destroys the coherences generated by the weaker field, regardless of the absolute values of the field intensities.

The impact of field-coherence destruction on the 1-2 transition of the $\Lambda$ system is shown in Fig.9.10. The solid line in Fig.9.10a shows the linear response due to the 1-2 transition when $\Omega_{13} = \Omega_{23} = 5$ ps$^{-1}$ ($\tan\beta = \frac{\Omega_{23}}{\Omega_{13}} = 1$). When $\Omega_{23}$ is increased by four times ($\tan\beta = 4$), the spectrum becomes a doublet (dashed line). Comparing this spectrum with that for $\Omega_{23} = 20$ ps$^{-1}$ and $\Omega_{13} = 0$ (dotted line) illustrates that increasing $\Omega_{23}$ decreases the effect of the weaker field resonant with the 1-3 transition. This reduces the excitation of electrons out of the ground subband, increasing the absorption amplitude associated with the 1-2 transition. The doubling of the peak seen in Fig.9.10a indicates the start of LIT, as will be discussed below.

Fig.9.10b shows the inverse case for the same transition, i.e. $\Omega_{13} \geq \Omega_{23}$. Here again the solid line corresponds to $\Omega_{13} = \Omega_{23} = 5$ ps$^{-1}$. When $\Omega_{13}$ increases to 20 ps$^{-1}$ ($\tan\beta = 1/4$), the spectrum changes drastically, showing extensive gain with a dip at the line-center. This spectrum is almost indistinguishable from that for the case where $\Omega_{13} = 20$ ps$^{-1}$ and $\Omega_{23} = 0$ (dotted line). The hole in the gain spectrum here is a sign of quantum interference caused by destruction of the coherences involving the second subband, as
Figure 9.10: The contribution of the 1-2 transition to the linear absorption of a weak probe field. In both parts, the solid line corresponds to $\Omega_{13} = \Omega_{23} = 5 \text{ ps}^{-1}$. In (a) the dashed line is for $\Omega_{13} = 5 \text{ ps}^{-1}$ and $\Omega_{23} = 20 \text{ ps}^{-1}$, and the dotted line is for $\Omega_{13} = 0$ and $\Omega_{23} = 20 \text{ ps}^{-1}$. In (b) the dashed line is for $\Omega_{13} = 20 \text{ ps}^{-1}$ and $\Omega_{23} = 5 \text{ ps}^{-1}$, and the dotted line is for $\Omega_{13} = 20 \text{ ps}^{-1}$ and $\Omega_{23} = 0$. The insets show the interfering paths which lead to LIT or dark line generation [Ref. [28]].
discussed below. The gain is caused by the immense deposition of electrons in this subband, which is also responsible for the \( \tan \beta = 1/4 \) spectrum's strong similarity to the case where \( \Omega_{23} = 0 \) (dotted line).

Now consider the evolution of the 1-3 transition’s spectrum under the same conditions as Fig.9.10. As shown in Fig.9.11a, in this case when \( \Omega_{23} = \Omega_{13} = 5 \text{ ps}^{-1} \) the 1-3 transition spectrum exhibits a transparency hole. When \( \Omega_{23} \) is increased, the absorption amplitude increases and the hole becomes deeper. As was the case in Fig.9.10a, this is caused by quantum interferences which occur when the coherences involving the ground subband are destroyed. Figs.9.10a and 9.11a thus provide complementary information about the evolution of LIT in the two driven subbands of a single system. The LIT effect is weaker in the absorption associated with transitions to the narrower subband (Fig.9.10a), because differences in the damping rates of the coupled subbands suppress (enhance) the quantum interferences that cause LIT when the narrower (broader) subband is probed. The interference processes are discussed further in the following section.

Fig.9.11b shows the evolution of the absorption due to the 1-3 transition, under the same conditions as Fig.9.10b. While the latter shows a large amount of gain, here we only see a doublet which is suppressed with increasing field strength. This suppression is caused by deposition of most of the electrons in the second subband. The system consisting of the two driven subbands here is similar to that discussed in detail in the previous chapter.

### 9.6.4 Quantum interferences in a \( \Lambda_4 \) system

Coherent control of the intersubband transitions is primarily determined by the coupling mechanisms and configuration of the system. The relative strengths of the coupling mechanisms depend on the coupled subband widths and the field frequencies and intensities. We showed in the previous section that when one field was either weaker than the other or was absent altogether, quantum interferences could lead to LIT and dark line effects.
Figure 9.11: The contribution of the 1-3 transition to the linear absorption of a weak probe field. The curves represent the same conditions as those in Fig.9.10. The inset in (a) shows the interfering paths which lead to LIT in the spectrum shown [Ref. [28]].
In this section we discuss these effects in more detail.

The quantum interferences in Figs.9.10-9.11 are mainly determined by one-photon coupling of two conduction subbands, and the probe transitions. Similar to what discussed in chapter 3, here there are two types of interfering paths: direct ones created by the probe field, and indirect ones generated by both the probe and coupling fields. In the case of Fig.9.10a the direct path is the 1-2 transition, and the indirect one is the 1-3 transition through 2 (see Fig.9.10a inset). The interference paths for the inverse case (Fig.9.10b) are similar in origin, but the intermediate state of the indirect path is the ground subband (see inset). These interferences generate fixed-frequency LIT or dark line effects, depending on the population of the driven subbands.

The dynamics of the gain in Fig.9.10b are made clearer by considering various values of $\Omega_{13}$ with $\Omega_{23} = 0$. The resulting absorption spectra are shown in Fig.9.12. For small values of $\Omega_{13}$, there is a large absorption peak corresponding to the 1-2 transition. As $\Omega_{13}$ increases, the former peak is suppressed and eventually develops a transparency hole. Further increase in $\Omega_{13}$ produces a single gain peak which develops a dark line. Fig.9.13 shows the electron distributions in the three subbands for the case where $\Omega_{13}=20$ ps$^{-1}$. The second subband (dashed line) contains a large fraction of the total population—more than twice that in the ground subband (solid line). Note that Fig.9.12 shows how in a single system quantum interferences can cause both LIT and dark line effects, depending on the populations of the upper subbands and the field intensities.

The interferences affecting the spectra in Fig.9.10b are between the direct 1-3 and indirect 1-3-2 paths, as shown in the inset. In the case of Fig.9.11b, however, increasing $\Omega_{13}$ suppresses the indirect path. The interference phenomena in this case are mostly due to competition between absorption and gain. At higher field intensities this produces dispersive features in the 1-3 absorption spectrum.
Figure 9.12: The contribution of the 1-2 transition to the linear absorption of a weak probe field when only the first and third subbands are coupled. The Rabi frequencies corresponding to each line are solid: $\Omega_{13}=5 \text{ ps}^{-1}$, dashed: $\Omega_{13}=7 \text{ ps}^{-1}$, dotted: $\Omega_{13}=10 \text{ ps}^{-1}$, dash-dotted: $\Omega_{13}=20 \text{ ps}^{-1}$, dash-dot-dotted: $\Omega_{13}=40 \text{ ps}^{-1}$ [Ref. [28]].

Figure 9.13: Electron distributions for the system of Fig.9.12, with $\Omega_{13}=20 \text{ ps}^{-1}$ ($I=113 \text{ MW/cm}^2$). The solid, dashed, and dotted lines represent the populations in the first, second, and third subbands, respectively [Ref. [28]].
Chapter 10

Laser systems and optical setups

10.1 Introduction

In the previous chapters we theoretically studied nonlinear optics of atomic and QW systems caused by their near resonance interactions with strong laser fields. To put some of the results of these theories to test we also conducted several experiments. The remainder of this thesis contains the descriptions and analyses of these experiments. While the primary goals of these studies were to investigate the dynamics of the QW emission spectra in the presence of an intense CO₂ laser (related to our discussions in chapter 5), the results shed some light on other novel phenomena. Briefly, the experimental results have shown the following:

1. The usefulness and completeness of the theoretical developments presented in this thesis. We used these theories extensively in analyzing the experimental results.

2. The PL spectra of an infrared-coupled QW can become peculiarly quenched. We show that this is a sign of multi-level mixing of excitons and the infrared enhancement of their effective non-radiative decay rates.

In the following sections we briefly present an overview of the experimental setup and measurement techniques. The results and analyses will be discussed in chapter 11.

10.2 Outline of the experiment

To carry out the experiment we arranged an optical setup as shown in Fig.10.1. The essential parts of this setup were two laser systems (one to generate electron-hole pairs
Figure 10.1: Schematic diagram of the experimental setup. The circle represents the cryostat. The cold finger and sample are shown at the center of this circle.

and the other to couple the QW conduction subbands), an arrangement for the synchronization of these two laser pulses, low temperature utilities, and detection and analysis equipment.

10.2.1 Visible laser pulse generation system

The visible laser used to generate electron-hole pairs consisted of a Nd:YAG mode locked laser (not shown) and a Nd:YAG Regenerative Amplifier (RGA). The former laser generated a single mode quasi-continuous pulse train with 1.064 μm wavelength and 82 MHz
repetition rate. The pulse duration of this laser was about 70 ps. This laser was then used to seed the RGA unit. This increased the energy of the RGA pulses and reduced their jitter. After passing through a second harmonic crystal (KDP), the output of this laser was a train of ~100 mJ pulses with 0.532 µm wavelength. The repetition rate of this laser was between 3-10 Hz.

10.2.2 CO₂ hybrid laser

The other basic component of our setup was a CO₂ hybrid laser. In this laser a single mode continuous wave (CW) CO₂ laser shared a cavity with a transversely excited atmospheric (TEA) CO₂ laser. These two units had previously been built in our laboratory [98]. We combined them by putting a 100% reflecting flat mirror at one end and a grating at the other end. The grating allowed us to tune the laser output wavelength between 9.55 and 10.33 µm. The cavity length was 150 cm.

The CW laser gain medium was about 70 cm long, containing a lasing gas mixture of CO₂:N₂:He with 15:15:70 balance. The TEA section was a high pressure system which was originally designed to work between 1 and 7 atm. It contained a 50 cm gain medium with two salt windows built at Brewster angles. We found new optimized working conditions for this system. Under these conditions the CO₂ pulses had about 30 mJ energy and 0.1 Hz repetition. We used a lasing gas mixture CO₂:N₂:He with 6:6:88 balance at about 3 atm. We found that arcs between the main electrodes of the TEA laser caused a considerable amount of electrical noise which led to loss of the signal. Therefore, the operating conditions had to be optimized to minimize the amount of arcing. In addition to the TEA laser, the Krytron units also generated a large amount of noise. This was overcome with proper shielding.

The lasing of a TEA laser by itself results in multi-mode pulses with considerable jitter. A typical shape of such multi-mode pulses is shown in Fig. 10.2. In the presence of the CW laser, however, there are photons with the desired frequency. Therefore the
TEA pulses built up from these rather than from noise. This leads to single-mode pulses (see Fig.10.3).

### 10.2.3 Synchronization

In order to study the infrared-coupling effects on the PL emission spectrum of the QW, the CO$_2$ and visible laser pulses should reach the sample at the same time. To do this we used an adjustable electronic synchronization unit. This unit had two TTL outputs which could be independently adjusted within $0-3$ μs delay relative to the RGA output pulses. The TTL outputs were used to determine the triggering times of the hybrid CO$_2$ laser and the integrator compared to that of the RGA amplifier (see Fig.10.1). The CO$_2$ laser was triggered more than one microsecond earlier than the visible laser. This was the time required by the CO$_2$ laser to generate an output pulse after receiving the TTL signal.

As shown in Fig.10.1, the synchronization unit was triggered after receiving the RF signals of the mode locker. This unit locked on the 41.071 MHz clock of the mode locker and triggered the TTL signals. After a definite time determined by the delay units the
RGA was triggered. Note that since the repetition rate of the TTL signals did not match with that of the CO_2 laser, before triggering the laser, we used a frequency divider. This divider reduced the repetition rate of the TTL signals used to trigger this laser by 1/16. The output of this divider was then sent to a pulse generator to increase its amplitude by about 30 V, enough to trigger the Krytron unit. We adjusted the delay unit such that the narrow visible pulse fell within the CO_2 pulses on the sample location. Fig. 10.4 shows the simultaneous detection of these pulses at the location of the sample. Note that the visible pulse width was much narrower that the detector rise time (2 ns).

10.2.4 Experimental data collection system

To study the evolution of the PL spectrum in the presence of intraband excitonic coupling, one needs to frequency resolve the QW emission. This was done in our experiment using a 0.25 m monochromator. The output of the monochromator was then guided into an electromagnetically shielded room using fiber optics. Inside, it was detected by a GaAs PMT tube. The detected signals were then fed into an integrator where they were integrated over 5 ns. The output signal was then sent to a computer for storage and
Figure 10.4: Simultaneous detection of the synchronized visible and CO$_2$ laser pulses at the location of the sample.

analysis. As mentioned before the integrator was triggered by one of the channels of the RGA delay unit.

10.3 Sample characteristics

The sample was grown by Dr. Tiedje's research group in UBC using molecular beam epitaxy (MBE). It contained fifty 7.3 nm nominally undoped wells (GaAs) sandwiched between 18.1 nm barriers (Al$_{0.28}$Ga$_{0.72}$As). A buffer layer consisting of 440 nm GaAs followed by a smoothing superlattice separates this structure from the [100] plane of the GaAs substrate. The thickness of the semi-insulating substrate was 1 mm. Two edges of the sample were polished at 45°, making a wave-guide geometry (see Fig. 10.5), and then mounted on the cold finger of a cryostat. The conduction band of this structure has two subbands with energy spacing $\sim$121 meV at 77 K. The electronic energy scheme of this structure at this temperature is shown in Fig.10.6.
Figure 10.5: Wave-guide geometry of the QW sample.
Figure 10.6: The electronic energy scheme of the sample at 77 K [31].
Chapter 11

Emission spectra of excitons in the presence of an intense CO$_2$ laser

11.1 Introduction

Modification of the PL spectra of QW's using the conduction intersubband transitions has already been employed to study the evolution of electrons [101, 102] and the non-diagonal emission spectra of E2-HH1 excitons [67]. In chapter 5 we theoretically discussed the latter and showed its capability to reveal quantum interferences. In this chapter we experimentally and theoretically study how the emission spectra of the QW structure sample described in the preceding chapter evolves in the presence of an intense CO$_2$ laser. This laser coupled the E1-HH1 and E2-HH1 excitons. To analysis the results of this section we use our developments presented in chapters 2, 4 and 5.

11.2 Experiment

The studies of intraband excitonic coupling presented in chapter 5 were involved with certain conditions and approximations. Some of these approximations were related to ignoring the many-body effects caused by carrier-carrier scattering. To hold these approximations valid we attenuated the Nd:YAG laser such that the carrier densities were about $3 \times 10^9 \, \text{cm}^{-3}$. At this limit the dynamics of the system were mostly determined by excitons. The waist of this laser on the sample was about 1 mm. Under these conditions the PL spectra of the QW in the absence of the CO$_2$ laser for three different trials are shown in Fig.11.1 (open circles). The low-energy tails might be caused by the excitons localized by the layer interface roughness and other imperfections [104]. Because in terms of the interface morphology various lateral regions of the sample were not identical, the
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spectra shown in Fig.11.1 (open circles) are slightly different.

To see how these spectra are changed in the presence of the CO₂ laser, we adjusted timing of the system such that pulses of this laser and those of the visible laser reached the sample at the same time. The waist of the CO₂ laser on one of the facets of sample was ~1.5 mm and its intensity inside the sample was ~1 MW/cm². On its first total reflection inside the sample this laser influenced part of the QW which was pumped by the visible laser. The results of our observation for three different frequencies of the CO₂ laser with polarization along the growth direction are shown in Fig.11.1 (filled circles). Inspection of these results show that when the CO₂ laser photon energy was 121 meV, we see the maximum amount of quenching around the peak frequency (Fig.11.1b). When the CO₂ photon energy increases by 9 meV, we still see a significant quenching at this region (Fig.11.1a). However, as Fig.11.1c shows, the quenching decreases significantly when the photon energies are reduced by 4 meV compared to that in Fig.11.1b. This shows that the quenching process depends strongly on the frequency of the CO₂ laser. In the following we will show that this is a sign of mixing of the exciton states associated with E₁ and E₂ with different configurations.

To verify the role played by the CO₂ laser polarization in these dynamics we tried the experiment using the same CO₂ laser pulses but with in-plane polarization. Under similar conditions as those of Fig.11.1b the PL spectrum evolved as shown in Fig.11.2. Here the effect of the CO₂ laser is a slight quenching and some red shifting. We discuss this feature in the next section. Note that based on the beam geometry considered in the experiment when the incoming CO₂ laser is horizontally polarized, its electric field inside the sample has a component perpendicular to the growth direction. Therefore only half of the CO₂ laser intensity serves to couple the exciton states. In fact the results presented in Fig.11.1 are the net effects of the z-polarized CO₂ laser, i.e. the contributions of the in-plane polarization have been deducted.
Figure 11.1: PL spectra of the QW in the absence (open circles) and presence (filled circles) of the CO$_2$ laser with ~1 MW/cm$^2$ intensity and photon energies (a) 130, (b) 121, and (c) 117 meV. The solid and short dashed lines are the results of the theory. The long dashed line in (a) represents the result of the theory when $\mu_{1s-2s} = 0$. 
Figure 11.2: PL spectra of the QW in the absence (empty circles) and the presence (filled circles) of the CO\textsubscript{2} laser with in-plane polarization. Other specifications are the same as those in Fig.11.1b. Here the solid and dashed lines are the eye guide.

11.3 Analysis

The experimental results of the preceding section showed that the quenching of the emission spectra depends on the details of the frequency and polarization of the CO\textsubscript{2} laser. To analyse these results let us first describe the electronic picture of the coupling processes. Based on our energy level calculations and experimental measurements at 77 and 10 K we found the energy spacings between E\textsubscript{1} and E\textsubscript{2} at these temperatures equal to \(\sim 121\) and \(\sim 119\) meV, respectively, with less than 2 meV uncertainty (see Fig.10.6). Having these we come up with the electronic picture of the coupling process as that shown in Fig.11.3. Considering this picture one may attribute Fig.11.1b to the case where the CO\textsubscript{2} laser was near resonance with the E\textsubscript{1}-E\textsubscript{2} transition (Fig.11.3b). Also Figs.11.1a and 11.1c correspond to \(\Delta \sim 9\) and \(\Delta \sim -4\) meV detunings, respectively. As a result one expects to see stronger quenching in the latter case than in the former. This is, however, in contrast to our experimental results. In the following we investigate these phenomena.
Chapter 11. Emission spectra of excitons in the presence of an intense CO$_2$ laser

Figure 11.3: Schematic diagram of the electronic picture of the coupling process. (a), (b), (c) refer to the corresponding cases in Figs.11.1a, 11.1b, and 11.1c, respectively.

and show why the electronic picture fails.

To understand the dynamics shown in Fig.11.1 one has to consider the intraband exciton coupling processes caused by the CO$_2$ laser. We discussed these extensively in chapter 5 and addressed their selection rules. Based on these rules this laser can only couple heavy- or light-hole excitons with the same angular momenta. Therefore we expect that the coupling processes occur between the $\Psi^{n \Sigma}_{E_1-HH_1}$ and $\Psi^{n' \Sigma}_{E_2-HH_1}$ excitons ($n$ and $n'=1$ and 2). Note that, as discussed in chapter 5, here we have two types of transitions: diagonal ($n = n'$) and non-diagonal ($n \neq n'$). As Eq.(5.29) shows a non-diagonal transition is allowed because of the nonparabolicity of the HH1 subband. In fact this reduces the symmetry of the excitons states, making the non-diagonal transitions possible. In the following we estimate the dipole moment associated with such a transition.

Considering the intensity and frequencies of the CO$_2$ laser used in this experiment and the sample parameters, one can find three effective coupling configurations for the free excitons. As shown in Fig.11.4b, when the CO$_2$ laser is nearly resonant with the transitions between $\Psi^{1 \Sigma}_{E_1-HH_1}$ and $\Psi^{1 \Sigma}_{E_2-HH_1}$, $\Xi$ is the dominant coupling configuration of the system. Here $\Psi^{1 \Sigma}_{E_1-HH_1}$ decays radiatively while it is coupled to $\Psi^{1 \Sigma}_{E_2-HH_1}$. Also here
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The lower 1s and 2s states refer to those of $\Psi_{E_1-HH1}^{1s}$ and the upper ones to $\Psi_{E_2-HH1}^{1s}$. The dashed one-sided arrows refer to the LO-phonon decay of the E2-HH1 excitons. (a), (b), and (c) are called $V_-$, $\Xi$, and $\Lambda_-$ configurations, respectively.

Figure 11.4: Energy level schemes of the excitonic systems corresponding to Fig. 11.1. The lower 1s and 2s states refer to those of $\Psi_{E_1-HH1}^{1s}$ and the upper ones to $\Psi_{E_2-HH1}^{1s}$. The dashed one-sided arrows refer to the LO-phonon decay of the E2-HH1 excitons. (a), (b), and (c) are called $V_-$, $\Xi$, and $\Lambda_-$ configurations, respectively.

the energy spacings between $\Psi_{E_1-HH1}^{1s}$ and $\Psi_{E_2-HH1}^{1s}$ are similar to that between $E_1$ and $E_2$. Our developments in Sect. 5.7 refer to this specific case. When the laser photon energy is larger than these transition energies, $\Psi_{E_1-HH1}^{1s}$ can effectively be coupled to both $\Psi_{E_2-HH1}^{1s}$ and $\Psi_{E_2-HH1}^{2s}$ (Fig. 11.4a). In the case of smaller photon energies $\Psi_{E_2-HH1}^{1s}$ is coupled to $\Psi_{E_1-HH1}^{1s}$ and $\Psi_{E_1-HH1}^{2s}$ (Fig. 11.4c). We call the systems shown in Figs. 11.4a and 11.4c as the $V_-$ and $\Lambda_-$ systems, respectively. Note that in the $\Lambda_-$ and $\Xi$ systems only $\Psi_{E_2-HH1}^{1s}$ decays by emitting LO-phonons. In the $V_-$ system, however, both $\Psi_{E_2-HH1}^{1s}$ and $\Psi_{E_2-HH1}^{2s}$ emit LO-phonons. As we show in the following this plays a major role in the explanation of the experimental results. Also note that the $V_-$ and $\Lambda_-$ systems are analogous to the K and II systems discussed in chapters 2 and 4, respectively. Therefore, since we want to address the $V_-$ and $\Lambda_-$ systems at a phenomenological level, we can use the developments presented in these chapters. This is done using Eqs. (2.97) and (2.98) including the strong inhomogeneous broadening of the emission spectra.

To apply our model to the experiment we describe the inhomogeneously broadened emission spectra of the localized and free excitons by two Gaussian distributions. Adding
these two distributions we found a good fit to the emission spectra in the absence of the CO$_2$ laser (solid lines in Fig.11.1). Note that at 77 K the contributions of the free excitons are dominant. We considered the homogeneously broadened widths of these excitons were 1 meV and those of the localized excitons 0.4 meV. These values were estimated based on the specific nature of our sample. In samples with smoother interfaces and narrower PL widths (3-5 meV), the widths of the localized excitons have been estimated between 0.1-0.2 meV [105]. The line-width of the $\Psi_{E2-HH1}^{ss}$’s were considered to be 5 meV. This includes the fast decay of these excitons via emission of LO-phonons and the contribution of the layer interface roughness.

After extensive numerical calculations the results of the theory in the presence of the CO$_2$ laser are shown in Fig.11.1 (short dashed lines). In the case of Fig.11.1b the free excitons are coupled with $\Xi$ configurations. We reached a fairly good match with experiment by putting the non-radiative decay rate of these excitons equal to 0.15 ps$^{-1}$. Note that this rate corresponds to $1/\tau_2^{0}$ in Eq. (5.45). To understand such a fast decay process note that here the CO$_2$ laser effectively transfers excitons from their E$_1$-HH1 radiative states into the non-radiative E$_2$-HH1 states. The E$_2$-HH1 excitons are ionized very fast into electrons with large wave-vectors and holes at the bottom of the valence band. Before these electrons and holes bind together and relax into the radiative states of E$_1$-HH1 exciton, electrons have to undergo energy relaxation. This reduces their wave-vectors and makes their binding with holes more probable. However, since the effective masses of E$_1$ and E$_2$ are similar for small wave-vectors, the CO$_2$ laser can reexcite these electrons back into E$_2$ before they reach the bottom of conduction band (see Fig.11.5). Therefore the effective characteristic time of their relaxation enhances significantly. As a result, most of the electrons and holes are non-radiatively recombined before they could establish bound excitons and decay radiatively. Note that as shown in Fig.11.5 the ionization of the E$_2$-HH1 excitons and the subsequent reexcitation of electrons lead to the multiple LO-phonon decay process. In the following we discuss how such processes
Figure 11.5: Schematic diagram of energy relaxation of electrons in the Ξ system. The short curved and inclined arrows refer, respectively, to the intrasubband and intersubband transitions of electrons as they emit LO-phonons.

can be related to the heating of the sample.

For the case of Fig. 11.1a, however, since the CO₂ photon energy is relatively higher, the effective coupling configurations of the free excitons are $V_-$ types. Since in these systems both $\Psi_{E2-HH1}^{1s}$ and $\Psi_{E2-HH1}^{2s}$ decay very fast by emitting LO-phonons, we expect to see an effective quenching process. To apply the theory to experiment, here we considered the transition dipole moment for the non-diagonal transition between $\Psi_{E1-HH1}^{1s}$ and $\Psi_{E2-HH1}^{2s}$ ($\mu_{1s-2s}$) as a fitting parameter. Using the parameters of Fig. 11.1b, we found a good fit to the spectrum of Fig. 11.1a (short dashed line) assuming $\mu_{1s-2s} \sim \frac{\mu_{1s-1s}}{2} = e \times 1$ nm. Also to find out how much this dipole moment contributes to the coupling process, in another attempt we put $\mu_{1s-2s}=0$. In other words, we considered the system had a Ξ configuration with 9 meV detuning. As the long dashed line in Fig. 11.1a shows this made the result unrealistic.

To see the consistency of the theory and the estimated parameters (non-radiative decay rates of excitons and $\mu_{1s-2s}$), we now apply them to the Fig. 11.1c system without
any fitting parameter. Here since the field is detuned by -4 meV from the transition between $\Psi^{1s}_{E_1-HH_1}$ and $\Psi^{1s}_{E_2-HH_1}$, the system has a $\Lambda$- configuration (Figs.11.4c). As the short dashed line in Fig.11.1c shows the result is in a reasonable agreement with the experiment.

Fig.11.2 showed that the CO$_2$ laser with in-plane polarization could slightly modify the emission spectra of the QW. To estimate the effect of free carrier excitations in this case, we should look for the absorption coefficient of such excitations at 77 K. As Wu et al have shown for the CO$_2$ laser wavelengths and a QW structure similar to our sample this coefficient is $\sim$10 cm$^{-1}$ [103]. Considering this we found that the heating caused by such excitations in part of the QW affected by both visible and CO$_2$ lasers was insignificant. To estimate the amount of the heat generated in the GaAs substrate, we assumed the absorption coefficient of bulk GaAs at 10.6 $\mu$m equals 0.01 cm$^{-1}$. The temperature increase per CO$_2$ pulse was found to be less than 0.1 K. Such a temperature change can not cause any significant effect in the exciton dynamics. Note that in addition to the free carrier excitations, the emission spectra of infrared-coupled QW’s may also be affected by the second order effects of the infrared-QW interaction and by the infrared mixing of hole subbands and hole transitions. These effects may collectively be responsible for the quenching and red shifting of the emission spectrum in Fig.11.2.

In contrast to the case of the in-plane polarization, when the CO$_2$ is polarized along the growth direction we expect some effects due to heating. This can be understood by considering the electron dynamics after ionization of the E2-HH1 excitons. As Fig.11.5 shows since the E2-E1 transition energy is much larger than those of LO-phonons, each electron emits several phonons before recombining with a hole (radiatively or non-radiatively). These phonons could cause thermal ionization of the E1-HH1 excitons. The resulting electron-hole pairs may recombine non-radiatively, leading to some quenching. However, since such processes are initiated by the intraband excitonic transitions their rates are included in our estimate of the effective non-radiative decay rates of excitons. In
order to estimate the amount of heat generated in this case, we considered the absorption coefficient to be around 500 cm\(^{-1}\). This was estimated based on the absorption coefficient of a QW with \(\sim 10^{11}\) cm\(^{-2}\) electron density and a CO\(_2\) laser with \(\sim 1\) MW/cm\(^2\) intensity [16]. We found that relaxation of the photo-excited electrons increases the temperature of the part of the QW affected by both visible lasers by less than 1 K.

11.4 Coherent effects

In the preceding section we discussed the incoherent processes caused by different mixtures of the exciton states. We showed that the basic reason for having different amounts of quenching in Fig.11.1 was that for the three CO\(_2\) laser frequencies we had three different mixing configurations. In the case of Fig.11.1a the mixing configuration was \(V_-\) type with two channels of fast non-radiative decay. In the case of Fig.11.1b the configuration was a near resonant \(\Xi\) system, and for the case of Fig.11.1c it was a \(\Lambda_-\) configuration with inefficient quenching. In this section we discuss the coherent effects involved in the dynamics of these spectra. As expected, here these processes are involved with the multi-photon mixing of excitons. In chapter 4 we showed that these processes can lead to various effects, depending on the widths of the coupled levels. These could be Stark shifts, generation of a doublet or a triplet in the emission spectra, etc. To explore the coherent processes in the experimental data we make use of the developments presented in chapters 2 and 4.

Inspection of the features in Fig.11.1 shows a slight relative shift in the spectra associated with 130 and 117 meV photon energies. This shift originates from the coherent coupling of excitons with \(V_-\) and \(\Lambda_-\) configurations. One expects to observe these effects more distinct in samples with smoother interfaces and narrower emission widths. In the case of our sample we can distinguish the Stark shifts better if we consider those occur in the homogeneously broadened components of the free excitons in Fig.11.1. The dynamics of one of these components are shown in Fig.11.6 when the CO\(_2\) laser has 130
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Figure 11.6: The emission spectra of an homogeneously broadened free exciton component under the same conditions as those of Fig.11.1a (long dashed line) and Fig.11.1c (short dashed line). The solid line refers to the emission spectrum in the absence of the optical field.

(long dashed line) and 117 meV (short dashed line) photon energies. The blue and red shifted peaks are the Stark shifts caused by coherent mixing of excitons with V₀ and Λ₀ configurations, respectively. Note the amounts of quenching and shift seen in these cases are consistent with those in Figs.11.1a and 11.1c.

The Stark shifts seen in Fig.11.6 show that under the experimental conditions the contribution of coherent processes is small. Despite this for the sake of better understanding it is constructive to discuss the role played by the coupling mechanisms and coherent effects in the dynamics of the V₀ and Λ₀ systems. To do this we consider these systems when they are generated by two lasers near resonance with the 1s-1s and 1s-2s transitions (see Fig.11.7). The coherent effects in these systems can be discussed based on our developments in Sects. 4.4 and 4.5. There we showed that when in a Π or Ξ₀ system a broad state is coupled by two lasers to two narrower states, the two-photon coupling between the narrower states is dominant. This also occurs in the Λ₀ system. Here the
upper $\Psi_{E2-HH1}^{1s}$ state which is coupled to 1s and 2s states of E1-HH1 exciton is relatively broad. Therefore the system dynamics are mostly determined by the two-photon state which is immune from the non-radiative decay of E2-HH1 excitons. In fact, the larger is the rate of the scattering of LO-phonons with the E2-HH1 excitons, the less quenching occurs. This shows that the coherent effects in the $\Lambda_-$ system are against quenching of the emission spectra.

In the case of the $V_-$ system, however, the scenario is different. Here the laser fields couple two broad states (1s and 2s of E2-HH1) to a narrow lower energy state (1s of E1-HH1). Therefore, in contrast to the $\Lambda_-$ system, here the one-photon coupling between the exciton states is dominant. Here since excitons mainly undergo single transitions from 1s states of E1-HH1 to the 1s and 2s states of E2-HH1, their non-radiative decay rates increase drastically, causing extensive quenching.
Chapter 12

Conclusions and suggestions for future work

12.1 Introduction

In this chapter we briefly conclude and summarize the results of this thesis and present some suggestions for future work. These suggestions are basically proposed to experimentally test some theories developed in this thesis.

12.2 Summary and conclusions

Our primary task in this research program was to study the nonlinear coherent effects in the absorption spectra of QW's. The first attempt to observe such effects was made by Fröhlich et al. [11]. They showed that a CO₂ laser polarized along the growth direction of a QW could modify its absorption spectrum. The explanations of this experiment and almost all subsequent theoretical work, however, was tied to inadequate approximations and/or unrealistic treatments. Therefore, presenting proper a treatment of infrared coupling of excitons was our first goal. Briefly, our novel developments included:

1. A consistent theory for the coherent interaction of one or two infrared fields with undoped QW's. This theory properly dealt with the quantum mechanics of such interactions, selection rules for the infrared-induced excitonic transitions, effects of the Coulomb interaction between holes, etc.

2. Showing how, depending on the QW structural parameters, a single infrared field can cause multi-subband coupling of excitons. This major effect was overlooked by previous authors.
3. Coherent manipulation of the interband excitonic transitions. Here we showed that by employing two laser fields to drive multi-photon coupling processes, the absorption spectra of excitons can be controlled, to a large extent. The dephasing rates of coupled exciton states can be used to determine the strengths of these processes.

After addressing these issues we studied the effects of infrared-coupling of excitons on their emission spectra. This subject has not been previously discussed, to our knowledge. We first tackled this problem theoretically. Later we extended our investigations by conducting several experiments. Although the primary goals of these experiments were to test some of our predictions, the results went beyond that:

1. We observed, for the first time, that a CO$_2$ laser polarized along the growth direction of a QW can lead to a field-dependent quenching of the exciton emission spectra. Using our theoretical treatment we explained the mechanism responsible for this in detail.

2. We speculated on the existence of multi-photon coupling processes in the emission spectra of the QW.

The third part of this research was devoted to n-doped QW’s and the way they interact with intense laser fields. Here we theoretically studied coherent manipulation of the optical properties of QW’s both with and without large numbers of photo-excited electrons. In the former case, where the electron-electron scattering processes played major roles, we showed for the first time:

1. How the infrared field can influence the polarization dephasing rates associated with the intersubband transitions.

2. That the response of an infrared-coupled n-doped QW to a weak probe field can lead to various absorption and amplification processes. The latter can be the result
of nonlinear processes and quantum interferences or direct inversion. We studied these cases in various systems generated by the interactions of one or two intense infrared fields with n-doped QW's. We also showed how multi-photon coupling processes determine the dynamics of these systems.

This thesis also contained a comprehensive development of nonlinear optics of atoms. Although the primary purpose of these developments was to explain the dynamics of QW's and design their nonlinear optics, we found the results quite valuable in their own right. They include predictions of several new effects and an alternative picture to describe the near resonance interactions of lasers with matter. In contrast to the dressed-state picture in which the effects of damping rates are usually ignored by either considering the high-field limit or virtually putting the rates equal zero, this picture includes these rates consistently.

12.3 Suggestions for future research

Based on the theoretical and experimental investigations presented in thesis and the present equipment in our lab, one can perform the following experiments:

12.3.1 Coherent population trapping of excitons

Coherent population trapping of excitons and quantum interferences discussed in this thesis can be investigated using schemes slightly different than those explained in the previous chapters. One option is to study the interband emission spectrum of a QW in the presence of a visible laser such as Ti-sapphire and a CO$_2$ laser. The CO$_2$ laser couples the ground conduction subband to an upper subband and the visible laser couples the latter to a valence subband. One can even consider a case where the CO$_2$ laser couples the ground subband to the continuum and the visible laser is near resonance between a valence subband and this continuum. Detection of the E1-HH1 emission could also
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confirm some other theoretical predictions of this thesis, such as features of the laser-induced continuum structure.

12.3.2 Experimental observation of quantum interferences in non-diagonal excitonic transition

In our experiments we studied the diagonal interband transitions. Such transitions were involved with the first conduction and valence subbands. The results of Sect. 5.7, however, showed that quantum interferences are much more pronounced in non-diagonal transitions. As discussed in Sect.5.7.3, in these transitions the damping rate of excitons strongly supports the interference processes. Having this in mind, one can grow asymmetric structures in which the transition between the second conduction and first valence subbands is allowed. Therefore using experimental techniques similar to those we adopted in this thesis, one could observe the emission of the E2-HH1 excitons. This transition should develop a dark line effect at low field intensities. The oscillator strength of such a transition can be roughly 1/500 of that of the diagonal transition, therefore one needs a higher detection sensitivity. In this experiment one can also simultaneously detect the diagonal transition (E1-HH1 recombination).


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[87] For proof and detailed discussion refer to Many-Particle Physics by G.D. Mahan


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