TUNABLE ALL-OPTICAL DELAY VIA NONLINEAR OPTICAL PROCESSES IN SEMICONDUCTOR QUANTUM WELLS

by

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A DISSERTATION

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“Tunable All-optical Delay Via Nonlinear Optical Processes in Semiconductor Quantum Wells,” a dissertation prepared by Susanta Kumar Sarkar in partial fulfillment of the requirements for the Doctor of Philosophy degree in the Department of Physics. This dissertation has been approved and accepted by:

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The dramatic experimental demonstration of slow light in atomic vapors via electromagnetically induced transparency (EIT) has stimulated considerable interest in the dynamic control of the group velocity of light and in the development of tunable all-optical delays. This dissertation presents experimental studies of all-optical tunable delays via nonlinear optical processes in semiconductor quantum wells (QWs). Two different approaches have been pursued. The first employs EIT arising from electron spin coherence in semiconductors and the second is based on efficient carrier induced exciton dephasing in QWs.

The EIT-based approach takes advantage of the spin-orbit coupling in the valence band to couple two electron spin states to a common light-hole valance band state in a GaAs QW waveguide. Induced transparency due to electron spin coherence in optical absorption has been demonstrated by investigating both the polarization and magnetic
field dependence of the induced transparency. Signature of Rabi oscillations for excitonic transitions has also been observed. The relatively small transparency, however, has limited the fractional delay that can be achieved. Nevertheless, our studies have shown a novel approach for inducing and manipulating electron spin coherence in a semiconductor with neither external nor effective internal magnetic fields.

We have overcome the shortcomings of the EIT-based approach by exploiting unique incoherent nonlinear optical processes in semiconductors. In this approach, a control laser beam injects free carriers above the band gap of a GaAs QW. Strong Coulomb interactions between excitons and free carriers lead to highly efficient broadening and bleaching of the exciton absorption resonance, effectively modifying the group velocity of a signal pulse propagating near the exciton resonance. Fractional delay exceeding 200% has been obtained for an 8 ps optical pulse tuned near the heavy-hole exciton resonance, representing an improvement of more than one order of magnitude in terms of both fractional delay and signal bandwidth for tunable optical delays in semiconductors. In addition, pulse reshaping and pulse breakup observed near the exciton absorption line center also motivate further investigation of coherent propagation effects such as self-induced transparency in semiconductors.
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CHAPTER I

INTRODUCTION

Motivation

Group velocity of light has been a topic of great interest over many decades [2, 38]. Still it remains an active area of research even today [29, 1-3, 7-11, 35]. Study of group velocity of light is fueled by two primary reasons. On the one hand, the meaning and the interpretation of group velocity is a critical issue in fundamental physics [38]. Important reason for this is that the theory of relativity dictates that speed of an object can not be greater than c (the speed of light in vacuum). An important question to ask then is what it means to have group velocity more than c or even negative. Does the concept of group velocity still have meaning? When the meaning and interpretation of group velocity break down? [119] How superluminal (faster than c) group velocity can be related to propagation of information without violating causality? [4] On the other hand, many potential applications [5] of controllable group velocity have also generated lot of interests, particularly, in the area of telecommunications. These two reasons are complementary and together they enable better understanding of physics and improved performance of potential applications. Moreover, pulse propagation effects have the potential of being used as a spectroscopic tool to study various physical mechanisms [100].

Over the past several years, intense research has been done to use nonlinear optical methods to gain unprecedented control over the pulse propagation through the material systems. The dramatic experimental demonstration of slow light [12] in atomic vapors via electromagnetically
transparency (EIT) has stimulated considerable interest in the dynamic control of the group velocity of light. The group velocity \( v_g \) is defined by the velocity at which the peak of an optical pulse propagates through a medium and is given by

\[
v_g = \frac{c}{n + \nu \frac{dn}{d\nu}}
\]

where \( c \) is the velocity of light in vacuum, \( n \) is the refractive index of the medium at the center frequency \( \nu \) of the light pulse. As shown in Figure 1, dispersion curve \((n \text{ vs. } \nu)\) can lead to different values of the denominator \( n + \nu (dn/d\nu) \) at different frequencies. Controlling group velocity of light then essentially means controlling dispersion curve. For normal dispersion \( dn/d\nu > 0 \) and the group velocity is less than \( c \). But if the medium has an absorption line near \( \nu \), dispersion becomes anomalous and group velocity can be larger than \( c \), or even negative because of \( dn/d\nu < 0 \) in the anomalous dispersion region. Group velocities faster \([14-15]\) than the speed of light in vacuum (superluminal), slower than the speed of a car \([12, 13, 15-16]\), stopped light \([17-19]\), and even negative group velocities \([39]\) have all been experimentally achieved. The ability to control group velocity of light is not only interesting in understanding fundamental physics of governing pulse propagation in material systems, it also motivates possible novel laboratory and commercial applications based on tunable optical delay.

Time taken by an optical pulse through a medium is known as group delay, which is given by \( t_g = L/v_g \). Larger delay is possible by making either \( L \) or \( v_g \) large. In practice, however, requirement of acceptable absorption loss and dispersion induced pulse broadening limits the effective length of \( L \). Hence, tunable group velocity \( v_g \) is the convenient choice for tunable group delay \( t_g \). Tunable optical delay are of critical importance for applications such as all-optical buffers \([21, 27]\), controllable delay lines \([22]\), true time delay for phased array antennas in radio
frequency communications [20], optical signal processing [21], quantum memory where state of photon is transferred to state of matter [90-91], optical coherence tomography [6].

![Graphs showing absorption coefficient α and refractive index n of a medium as frequency are related.](image)

**FIGURE 1.** Absorption coefficient $\alpha$ and refractive index $n$ of a medium as frequency are related. When absorption in (a) is modified as in (b), there is corresponding change in dispersion ($n$ vs. $v$) curve. Dispersion curve engineering is the basis for the control of the group velocity of light.

One of the most important applications of tunable delay is optical buffering as explained in Figure 2. In an $N \times N$ switch or router, any input can be connected to any output. However, if two or more data packets arrive at the switch at the same time, data will be lost. A solution is optical buffer where one can hold data using delay and release on demand by optical switching. Existing technology mainly uses electronic switch and buffer for telecommunications and this limits the speed of communications. All-optical switch and all-optical buffer based on tunable delay light can improve both the cost and the speed of communications significantly.
Even though the concepts of phase, group, information, energy velocity have been studied over many decades [38, 39, 120], dramatic control over the pulse propagation has been demonstrated only recently [12,15]. The wide scope of applications where tunable propagation of an optical pulse or other modulation of an optical carrier at a very low group velocity, has motivated different approaches for generating them.

**FIGURE 2.** All-optical buffer based on tunable optical delay. (a) N X N optical switch. (b) Optical buffer can be used to prevent two data packets arriving at the optical switch at the same time.

There are many mechanisms for achieving slow light, essentially all of which mean dispersion curve engineering and create narrow spectral regions with high dispersion as shown in Figure 1. Schemes can be divided into two categories: material dispersion and waveguide dispersion. Waveguide dispersion mechanisms in coupled resonator structures (CRS) [30] based on photonic crystals, Coupled Resonator Optical Waveguides, and other micro-resonator structures modify the
spatial component (k-vector) of a propagating wave. The other approach is based on nonlinear optical effects [3, 5]. To realize tunable optical delay various nonlinear optical effects have been pursued based on material dispersion mechanisms such as Electromagnetically Induced Transparency (EIT) [12], Coherent Population Oscillation (CPO) [15-16, 31-34], stimulated Brillouin and Raman scatterings [24-27], optical wavelength conversion and various Four Wave Mixing (FWM) [23-28] schemes produce a rapid change in refractive index as a function of optical frequency. In EIT and CPO, optical interactions between a signal and a pump induce a narrow transparency window within an absorption resonance. Tunable optical delay can be achieved via the pump intensity dependence of the spectral width as well as the depth of the transparency window. In stimulated light scattering, the parametric gain depends on the intensity of the pump beam, which can be used to generate tunable optical delay for the signal beam. In optical wavelength conversion, the spectral dependence of the group velocity in a fiber provides a convenient mechanism for tunable optical delay.

The materials used in these studies range from atomic vapors [12-14], doped ions in crystals [15-16], optical fibers [24-28], biological bacteriorhodopsin [36] and semiconductors [31-35]. However, semiconductor based tunable optical delay devices are far more desirable for potential advantages including compactness, low power consumption, and opportunity for monolithic integration. Tunable slow light in semiconductors has been pursued using coherent nonlinear optical processes such as CPO and EIT. CPO has already been a widely studied nonlinear optical phenomenon in semiconductors [31-34]. Slow light based on CPO in GaAs QW has been demonstrated experimentally, but suffers from serious disadvantage that it is very inefficient at room temperature. EIT based on electron spin coherence, however, has the promise to be applicable at room temperature because of the robust electron spin coherence. Robustness of electron spin coherence at room temperature comes from the fact that electron spin interacts
weakly with phonons, and hence increased phonon modes at higher temperature does not contribute to electron spin dephasing. EIT arising from excitonic coherence has already been demonstrated in semiconductors [92-95, 108], but fragile excitonic coherence leads to restrictions similar to CPO at elevated temperature. Also, relatively weak transparency and wide transparency window due to excitonic coherence makes it not so promising for slow light applications. EIT based on electron spin coherence becomes a natural direction to pursue due to it viability for potential slow light based semiconductor devices at room temperature, and also due to potential spintronics device application based on electron spin. Thus, EIT based on electron spin coherence becomes a major thrust of our research presented in this dissertation. This dissertation presents experimental studies of all-optical tunable delays via nonlinear optical processes in semiconductors. Two different approaches have been pursued. The first employs EIT arising from electron spin coherence in semiconductors and the second is based on efficient carrier induced dephasing of dipole coherence (excitonic coherence) in QWs [47].

The EIT-based approach takes advantage of the spin-orbit coupling in the valence band [45] to couple two electron spin states in conduction band to a common light-hole valence band state in a GaAs QW waveguide. Induced transparency due to electron spin coherence in optical absorption has been demonstrated by investigating both the polarization and magnetic field dependence of the induced transparency [60]. The relatively small transparency, however, has limited the fractional delay that can be achieved. Fractional delay is a figure of merit of slow light schemes and is given by the ratio of pulse delay divided by the pulse width. Nevertheless, our studies have provided techniques of optical spectroscopy in semiconductor waveguides and novel way of inducing and manipulating electron spin coherence in semiconductors without either external or internal magnetic fields.
As it turns out, slow light based on coherent nonlinear optical processes such as CPO and EIT in semiconductors has some disadvantages. Even though longer coherence times are desirable for many applications, longer coherence times necessarily lead to narrower transparency window. Coherence times in semiconductors due to various dephasing mechanisms are considerably shorter than those in the atomic systems, but resulting transparency window is still narrow enough to limit the bandwidth of the signal pulse that can be slowed, thereby, limits the fractional delay or delay-bandwidth product. For device applications though higher delay-bandwidth is desirable [37]. On the other hand, shorter coherence times in semiconductors compared to atomic systems or solid state crystals lead to shallow transparency window. Hence, both narrowness in coherent optical process based approaches and shallowness due to shorter coherence times limit fractional delays. Various dephasing mechanisms in semiconductors make coherence times shorter and nonlinear optical processes can be quite complex [47-48]. Excitonic effects (bound electron-hole pair), many-body effects due to Coulomb interaction, exchange interactions all can significantly modify nonlinear optical response of a semiconductor as compared to atomic systems. Exciton-exciton, exciton-carrier interactions can lead to enhance dephasing and shorter coherence times [57].

However, we have taken advantage of otherwise undesirable enhanced dephasing due to exciton-exciton, exciton-carrier induce dephasing in our favor. We have overcome the shortcomings of the EIT-based approach by exploiting unique incoherent nonlinear optical process in semiconductors. In this approach, a control beam injects free carriers above the bandgap of a GaAs QW. In semiconductor absorbers, coherent and incoherent many-body processes due to carrier-carrier Coulomb interactions (Coulomb correlations) can significantly modify the optical properties such as the dephasing of induced optical polarization, which is the source for the propagating light fields. Strong Coulomb interactions between excitons and free carriers lead to
highly efficient broadening and bleaching of the exciton absorption resonance, effectively modifying the dispersion and hence the group velocity of a signal pulse propagating near the exciton resonance. Fractional delay of more than 200% has been obtained for an 8 ps optical pulse tuned near the heavy-hole exciton resonance [29]. Our approach provides an improvement of more than one order of magnitude in terms of both fractional delay and signal bandwidth for tunable optical delays in semiconductors.

Tunable optical delay in semiconductors based on either coherent or incoherent optical processes necessarily involves the propagation of light through dispersive medium of semiconductors. As mentioned before, pulse propagation through dispersive medium has been a topic of great interests over many decades [38-39, 117-120]. In the context of tunable slow light, pulse propagation in semiconductors has become more interesting and timely topic of research. Propagations of Off-resonant pulse and On-resonant pulse can be very different due to different dispersions for two regions. Moreover, both coherent [128-129] and incoherent optical processes can significantly affect the pulse propagation. Coherent effects such as self induced transparency (SIT) and incoherent effects such as saturation has been well studied in the context of pulse propagation [121-123, 100]. Still there are open questions about the effects of various physical mechanisms on pulse propagation and about the meaning of group velocity. In this dissertation, we will present both Off-resonant and On-resonant pulse propagation around excitonic transitions in GaAs QW. We have found that for strong absorption, both the group velocity and energy velocity definition of pulse propagation becomes inapplicable [118]. One input Gaussian pulse breaks up into two Gaussian pulses, and thus group velocity defined by the velocity of the peak of a pulse (input) can not be applied. One output Gaussian pulse precedes the input pulse, whereas, the other output Gaussian pulse follows the input pulse. We will also present our studies of the effects of incoherent optical processes on pulse propagation. Efficient carrier induced dephasing
due to optical injection of free carriers by a control beam has been used for this study. Effects of coherent optical processes such as SIT will be the topic of future work in our laboratory. As it is evident from the foregoing discussion, electron spin coherence and dipole (excitonic) coherence and dephasing form the basis of our two approaches to tunable delay light in semiconductors. In the next section, we will briefly discuss these two coherences. Details of coherences will be discussed in later chapters.

**Coherences in Semiconductors**

Coherence means definite relation. For quantum systems, quantum coherence can be understood as a well-defined phase relationship between the probability amplitudes of two eigenstates in a superposition state. Under resonant optical excitations, individual optical transitions in semiconductors can be approximated to involve only two levels. Figure 3 shows basic schemes of generating dipole coherence (excitonic coherence) in a two-level system and electron spin coherence in three-level systems. A phenomenological model of relaxation dynamics of a collection of two-level systems was first introduced by Bloch in the context of magnetic resonance of spin-1/2 particles was later applied to resonant optical excitations of atoms (Optical Bloch Equation or OBE). OBE was further modified to include many-body Coulomb interaction in semiconductors. The resulting equation, known as Semiconductor Bloch Equations (SBE), reduce to OBE for independent two-level systems when Coulomb interaction between excitons (bound electron-hole pair in semiconductors) or the electron-hole pairs is set to zero. Two observables characterize the relaxation dynamics of a quantum mechanical two-level system: 1) population difference between two states and 2) coherence or phase relationship between the two states. Population or energy relaxation is characterized by the longitudinal decay time or population relaxation time $T_1$. Loss of coherence between two states is characterized by the transverse decay time or dephasing time $T_2$. Coherences can significantly modify optical
responses of semiconductors and can lead to new phenomena due quantum interference arising from quantum coherence, and can be studied using optical spectroscopy. Coherences can be destroyed by various dephasing mechanisms. Excitonic coherences can be dephased due to various mechanisms such as carrier-carrier interactions, carrier-exciton interactions, and exchange interactions [47, 57]. For electron spin decoherence, four mechanisms are primarily relevant for semiconductors: the Elliott-Yafet, D’yakonov-Perel’, Bir-Aronov-Pikus, and hyperfine-interaction mechanisms [58].

\[ |\psi(t)\rangle = c_0(t)|0\rangle + c_1(t)|1\rangle \]

**Population:** \[|c_1(t)|^2 - |c_0(t)|^2 \sim e^{-t/T_1} \]

**Coherence:** \[c_0(t)c_1^*(t) \sim e^{-t/T_2} \]

**Nonradiative or Raman Coherence:**
\[c_1(t)c_2^*(t) \sim e^{-t/T_2^R} \]

**FIGURE 3.** Exciton spin coherence and electron spin coherence. Generic two- and three- level systems. The arrows indicate dipole allowed optical transitions. Resonant optical interaction can generate dipole coherence in the two-level system. The coupling of the two upper states to a common ground state in the three-level system allows for optical control of the non-radiative coherence between states \(|1\rangle\) and \(|2\rangle\) that can be two electron spin states.

In contrast to hole spin and exciton spin which exhibit coherence time on the order of couple of picoseconds, electron spin coherence has coherence time on the order of the recombination time
and can last for nanoseconds \[65\]. Electron spin coherence times up to 100 ns have been observed in n-doped semiconductors \[61\]. Weaker electron-phonon interactions compared to hole-phonon and exciton-phonon interactions lead to robust electron spin coherence in semiconductors even at room temperature. There is another compelling reason for studying electron spin coherence. Already well developed electronics based on electron charge can be easily integrated with results and ideas based on electron spin \[58\].

\[
\begin{align*}
B_z &= 0: \\
J_z &= 3/2, s_z = 1/2, \sigma^+ \\
J_z &= -3/2, s_z = -1/2, \sigma^-
\end{align*}
\]

\[
B_z \neq 0:
\]

![Diagram](image)

**FIGURE 4.** Optical selection rules between HH and conduction band. Top: Without magnetic field. Bottom: Effects of in-plane magnetic fields on the energy level structure. Magnetic field changes the optical selection rules also. Two V-type systems result suitable for optical control of the non-radiative electron spin coherence.

The key to optically address spin coherences in semiconductors is to use optically allowed transitions (optical selection rules). One way of coherent spin manipulation is applying magnetic
fields. To study electron spin coherence, semiconductor quantum wells placed in a transverse magnetic field (Voigt geometry) are commonly used. For example, in GaAs quantum well (QW) the magnetic field can induce a Zeeman splitting between the two conduction band electron spin states. Coherent superposition of these spin states leads to a Larmor precession of the electron spin, which can be probed with transient optical techniques.

Semiconductor QWs are particularly well suited because confinement lifts the degeneracy between HH and LH. Moreover, growth direction defines a preferred axis in QW systems. Modifications of the energy level structure (thereby, possibly changing the optical selection rules) by applying magnetic fields depend therefore on the orientation of the magnetic field with respect to growth direction, allowing the study of different kinds of spin coherences. To probe electron spin coherence in QWs, an external magnetic field is applied in the plane of QW which modifies the band structure so that both conduction band electron spin states are optically coupled to the same valence band state. As a result, the non-radiative or Raman coherence between two electron spin states in the conduction band can be created optically in a V-type energy level scheme as shown in Figure 4. Coherence between two spin states can lead to interference between two pathways to common ground state which, in turn, can lead to quantum beats. These quantum beats combined with spin relaxation and dephasing processes lead to oscillating decay phenomena of macro observables, such as rotation of plane of polarization or intensity of photoluminescence, absorption. This quantum coherence of electron spins can be understood in terms of spin precession. The initial coherent superposition of spin states corresponds to a net spin along the direction of light propagation and perpendicular to applied field. This net spin or magnetization will then precess around the magnetic field \( \mathbf{B} \). The amplitude of beats is proportional to the optical magnetization, in other words, proportional to the intensity of the polarizing light intensity.
Manipulation of electron spin coherence without magnetic fields just by exploiting optical selection rules has not been explored so far. An interesting question of both conceptual and practical importance is whether coherent electron spin manipulation in semiconductors can be carried out without either external or internal magnetic fields. By taking advantage of spin-orbit coupling in GaAs QW, we have shown in this dissertation that just by using optical selection rules one can induce electron spin coherence between two spin states in the conduction band, as shown in Figure 5. There is no spin precession in our approach; therefore, time domain techniques are no longer applicable for experimental study.

FIGURE 5. Optical selection rules between LH and conduction band. Top: Without magnetic field. Bottom: Effects of magnetic field perpendicular to QW plane on the energy level structure. Magnetic field does not change the optical selection rules also. Two V-type systems result suitable for optical control of the non-radiative electron spin coherence.
Instead we have used a spectral domain optical technique measuring differential transmission (transmission with control beam ON minus transmission with control beam OFF). Destructive interference between two pathways to the common state shows up as a transmission peak in differential transmission. Because of the unique optical selection rules used, we need to do the experiment in waveguide geometry.

**Dissertation Objective and Organization**

As mentioned before, two different approaches will be presented in pursuit of tunable all-optical delay in semiconductors. One approach is based on EIT arising from electron spin coherence. We will present a new way of inducing and manipulating electron spin coherence without magnetic fields in GaAs QW waveguides. Techniques of nonlinear pump-probe spectroscopy in semiconductors waveguides from 4 K to room temperature have been developed for studying electron spin coherence and excitonic coherence in GaAs QW waveguides. This shows the feasibility of observing EIT using electron spin coherence in semiconductors. Moreover, observed signature of Rabi oscillations arising from excitonic coherence makes waveguides promising for coherent manipulation with added advantage of long interaction lengths.

The other approach takes advantage of incoherent carrier induced exciton dephasing in GaAs QW. We report delay of more than two pulse-widths for an 8 ps optical pulse. Our method provides higher bandwidth, less absorption of the control beam, stability against variations of parameters of the control beam, and as good delays as compared to other approaches. We will also present pulse propagation through different dispersive regions and effects of nonlinear optical processes on pulse propagation in GaAs QW.

The organization of this dissertation is as follows. Chapter 2 contains a general discussion about optical processes in semiconductors, particularly in direct bandgap GaAs. We will discuss how coherences, many-body Coulomb correlations affect coherent optical responses in
semiconductors and how these can be studied using pump-probe spectroscopy. Chapter 3 discusses a particularly important nonlinear optical phenomenon called EIT. EIT is a process in which an otherwise opaque (absorbing) medium is made transparent as a consequence of destructive interference due to quantum coherence. We will discuss both exciton (dipole) coherence and electron spin coherence in the context of EIT. We will show that a chirped pulse can be used to cancel the effect of blue shift due to band renormalization in semiconductors. This will help satisfying the two-photon resonance condition, which is one condition for optimum EIT. In chapter 4, we present various experimental techniques used in our research. Experimental results presented include transmission spectra, saturation effects, coherent effects, propagation effects etc. Electron spin coherence without magnetic field in waveguide and signature of Rabi oscillations in waveguides deserve special attention and will be discussed in two separate chapters later. Chapter 5 presents a general discussion on electron spin coherence because of its importance not only for slow light, but also for spin-based “spintronics” device applications. Electron spin coherence without magnetic field in GaAs QW waveguide will be analyzed theoretically in detail. Chapter 6 contains experimental results proving that electron spin coherence can be created optically without magnetic field in GaAs QW waveguide. Chapter 7 discusses observed signature of Rabi oscillations in GaAs QW waveguide. Observation of Rabi oscillations confirms that QW waveguide can be used for quantum coherent manipulation. Waveguides have optical interaction length given by their length in contrast to normal geometry where optical interaction length is given by the quantum well width. Chapter 8 contains our approach of tunable slow light based on incoherent saturations and bleaching of excitonic absorption. Chapter 9 contains our studies of both off-resonant and on-resonant pulse propagation around excitonic transitions in GaAs QW. We will also discuss how a control beam affects pulse propagation. Finally, Chapter 10 gives a summary of our research and future outlook.
In this chapter, we will discuss basic concepts of optical processes in semiconductors to facilitate the understanding of later chapters. Topics discussed include optical excitations in semiconductors [50], band structure of GaAs [45-46], semiconductor optics in the two-level approximation [44], excitonic transitions in GaAs QWs [51-54], pump-probe spectroscopy [47, 55].

Optical excitations in semiconductors

Optical excitations in semiconductors can often be understood qualitatively in analogy to atomic systems. The optical process of exciting an electron from valence band to conduction band leaves behind a positively charged hole in the valence band that behaves as a pseudo particle with an effective mass. This process can be approximated as an electric dipole transition. Close to the band edge (crystal momentum \( k = 0 \)) of a direct bandgap semiconductor such as GaAs, the optical transitions are dominated by excitonic effects. An electron (negatively charged) in the conduction band and a hole (positively charged) in the valence band can form an exciton (X) due to Coulomb interaction if the thermal energy (\( k_B T \)) is less than the exciton binding energy. In this regard, an exciton is similar to a hydrogen atom with an electron and proton, and it is not surprising that excitonic transitions share many similarities with atomic transitions. Excitons are classified into two types: Frenkel excitons (smaller radii) for which the electron-hole pair is very tightly bound, and Wannier excitons (larger radii) where electron orbits around a hole with larger
radius. Excitons in semiconductors are usually Wannier excitons with radii (typical value 10 nm). The dynamics of excitons is influenced by their interactions with surrounding crystal environments. These interactions include scattering with acoustic and optical phonons, scattering with other excitons and free carriers, and scattering from defects, impurities, interfaces and surfaces in the crystal.

Regardless of the similarities between excitons and atoms, excitons are different from atoms due to the effects and interactions simply not present in atomic systems. Whereas atomic transitions mean moving an electron from one atomic state to another, excitonic transitions refer to creating an exciton not making transitions within an existing exciton. In atomic ensembles, the atomic density is fixed and independent of laser excitations. However, density of excitons in semiconductors depends necessarily on laser excitations which create them in the first place. Increasing exciton density can cause exciton-exciton scattering, leading to exciton induced dephasing (EID) of dipole coherence.

Exciton-exciton interactions can also lead to formation of many-exciton states. For example, bound and unbound biexciton states can be formed due to Coulomb interactions similar to molecule formation from atoms. Biexcitons can have dramatic effects on optical properties of semiconductors. Even though many-exciton states comprising more than two excitons are possible, biexcitons have been found to have the most significant effect on optical properties. Charged excitons or trions can also form and have significant effect on optical response of semiconductors.

While quantitative theory of optical properties of semiconductors requires models that includes many-body interactions, in most cases optical interactions with excitons in semiconductors can be qualitatively described using simpler optical Bloch equations (OBE). Corrections such as local field effects, EID can be included in OBE and resulting model can be used quite effectively to
understand nonlinear optical properties of excitons. These models have been quite successful in explaining effects such as incoherent effects (e.g. bleaching, spectral hole burning), coherent effects (e.g. EIT [91-95], Population pulsation [49]), Rabi splitting [109-110], Mollow triplets [105-106]), four-wave mixing (FWM), quantum beats [67], photon echo [100] in semiconductors. Rigorous theoretical model of semiconductor optics including exciton-exciton interactions is challenging, but significant progress has been made. Semiconductor Bloch equations (SBE) within Hartree-Fock (HF) or mean-field limit takes electron-hole Coulomb interaction and exchange interactions into account and are highly effective in understanding a wide variety of phenomena in semiconductor optics. However, SBE within HF limit does not go beyond the mean-field limit and does not consider Coulomb correlations. As effects of Coulomb correlations such as in biexcitons have been explored experimentally, there is a need for sophisticated theoretical model. The Dynamics Controlled Truncation Scheme (DCTS) [56] truncates the infinite series of correlation terms by limiting the field up to certain order. DCTS have been used to explain optical properties of semiconductors, including biexcitonic effects, up to third order and in some cases up to fifth order of the applied field.

It should be noted that perturbative approach does not apply for phenomenon such as Rabi oscillations. It is well known that for resonantly excited two-level systems the populations of levels oscillate with Rabi frequency given by the electric field amplitude and the electric dipole moment. Rabi oscillations have been observed in semiconductors both in time domain and frequency domain experiments. This shows that even highly nonlinear phenomena in atomic systems have analogs in excitonic systems in semiconductors; however, full effects of Coulomb correlation on excitonic Rabi oscillations are not fully understood yet. EIT in semiconductors was also motivated by observation of EIT in atomic systems.
While phenomena in atomic systems can motivate experiments in semiconductors because of similarities observed, it is the observed differences that make semiconductor optics interesting.

**Band structure of GaAs**

Gallium arsenide (GaAs) is a noncentrosymmetric (i.e. do not have a center of inversion) semiconducting material with filled valence band and empty conduction band in equilibrium, and possess a cubic lattice. GaAs crystallizes in zincblende structure which has crystal point group $\bar{4}2m$. As a material of $\bar{4}2m$ crystal class GaAs possess large second-order nonlinear susceptibility. However, as it also possess cubic lattice, GaAs does not have birefringence (dependence of refractive index on the direction of polarization), and can not normally participate in phase-matched (so that emission by dipoles add coherently in the forward direction) second-order interactions.

To understand the optical properties of the semiconductors, it is important to know band structure including the energy band and the corresponding wave function. Energies (band structure) that an electron is allowed to have according to quantum mechanics is determined by combined effects of crystal potential, confinement potential and Coulomb interaction, but to treat them all at the same is theoretically challenging. For a periodic potential, the band structure and the wave function can be obtained from the Hamiltonian that satisfies the symmetry of the crystal. The general theory follows the Bloch theorem. There are many numerical methods of band structures calculations.

For direct bandgap semiconductors such as GaAs, the $\bar{k}\cdot\bar{p}$ method with spin-orbit coupling taken into account, is quite useful for calculating band structure near an extremum (for GaAs zone center at $k = 0$). In Kane’s model only a conduction band, a heavy-hole, a light-hole, and a spin-orbit split-off band, all doubly degenerate, are considered. All other higher and lower bands are discarded. In Luttinger-Kohn’s model the heavy-hole, light-hole, and split-off bands with double degeneracy are of interest and are called class A. All other bands are denoted as class B.
The effects of class B on class A is taken into account in this model. Quantized energy due to confinement in QWs is inversely proportional to mass. Due to their different effective masses, LH and HH degeneracy is lifted due to the confinement at the $\Gamma$-point (Brillouin zone center). Away from the $\Gamma$-point HH-LH band mixing becomes significant and HH and LH bands cross each other at some finite $k$. Figure 6 shows the band structure within the effective mass approximation near $k = 0$ for both bulk GaAs and GaAs QW.

**FIGURE 6.** Band structure of (a) bulk GaAs and (b) GaAs QW. In quantum wells degeneracy between HH and LH is lifted due to quantum confinement effect. For direct bandgap material such as GaAs band extrema occurs at $k=0$. Optical transitions in GaAs more efficient than that in indirect bandgap Si.

Bloch states are denoted according to the total angular momentum $J$ and its projection along positive z-axis $m_j : \langle J,m_j \rangle$. For GaAs, conduction band is s-like and valence band is p-like.
Expressing the wave functions with symmetry of $s, p_x, p_y, p_z$ orbitals as $|S\rangle, |X\rangle, |Y\rangle, |Z\rangle$, respectively, the band wavefunctions can be written as:

$$E \begin{array}{c}
\begin{array}{c}
\text{Al}_{0.3}\text{Ga}_{0.7}\text{As} \\
\text{GaAs} \\
\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}
\end{array}
\end{array}
\begin{array}{c}
\text{10-20 nm}
\end{array}
\begin{array}{c}
|e\rangle
\end{array}
\begin{array}{c}
|h\rangle
\end{array}
\begin{array}{c}
z \text{ (growth direction)}
\end{array}
$$

$|J, J_z\rangle = \begin{cases} 
\frac{1}{2} & \text{conduction band} \\
-\frac{1}{2} & \text{HH} \\
\end{cases}$

$|3/2, -3/2\rangle$  $|1/2, -1/2\rangle$

Figure 7. Type-I GaAs QW structure and optically allowed transitions. Materials with different bandgaps are grown epitaxially to create an effective QW.

Conduction band: $|1/2, 1/2\rangle = |S \uparrow\rangle$

$|1/2, -1/2\rangle = |S \downarrow\rangle$
Valence band

Heavy-hole

\[ |3/2, 3/2\rangle = \left(1/2\right)^{3/2} (X + iY) \uparrow \]
\[ |3/2, -3/2\rangle = \left(1/2\right)^{3/2} (X - iY) \downarrow \]

Light-hole:

\[ |3/2, 1/2\rangle = \left(1/6\right)^{3/2} \left[ (X + iY) \downarrow + 2Z \uparrow \right] \]
\[ |3/2, -1/2\rangle = -\left(1/6\right)^{3/2} \left[ (X - iY) \uparrow - 2Z \downarrow \right] \]

Figure 7 shows a schematic of QW structure (top) and optically allowed transitions in GaAs QW. In GaAs QW HH transitions are three times stronger than LH transitions as can be seen from the dipole matrix elements calculated using above band-edge wavefunctions. Level structure including the effects of Coulomb interaction is shown in Figure 8. The exciton states are conveniently expressed in terms of total angular momentum and its projection along z-axis: \( |F_z, F_z\rangle \), where \( F_z = J_z^\text{hole} + S_z^\text{electron} = \pm 1, \pm 2 \). In the absence of electron-hole exchange interaction, all four HH1 states are degenerate. As shown, exchange interaction removes this degeneracy partially and leads to bright states \(|\pm 1\rangle\) and dark states \(|\pm 2\rangle\).

In a quantum mechanical description of interacting electrons, we have Coulomb interaction among charge clouds associated with the electronic states as well as exchange interaction resulting from required anti-symmetry property of the many-electron wavefunction. Electron-hole exchange interaction that results from overlap of the electron and the hole wavefunctions, are generally enhanced as the dimension of the semiconductor structure decreases. Whereas direct Coulomb interaction dominate in the bulk (3D) and QWs (2D), effects of electron-hole exchange interaction dominates the band-edge exciton fine structure in QDs subjected to strong confinement.
**FIGURE 8.** Exciton states with Coulomb interaction. Exciton states can be expressed in terms of total angular momentum and its projection along z-axis. In the absence of exchange interactions all four HH1 states are degenerate. Exchange interactions remove this degeneracy partially and lead to bright $|\pm 1\rangle$ and dark states $|\pm 2\rangle$.

Semiconductor optics in the two-level approximation

For parametric processes (nonresonant excitation, i.e., $\hbar \omega < E_g$) the initial and final states are identical, and population can be lifted to a virtual level for a brief period of time of the order of $\hbar / \delta E$, where $\delta E$ is the energy difference between the virtual level and the nearest real level. Parametric processes are extremely fast (~ 0.1 fs) and can be described by a real susceptibility. Two-photon processes are usually much larger than one-photon processes except for photon energies $\hbar \omega$ approaching bandgap energy $E_g$. For parametric processes narrow bandgap semiconductors produce much larger nonlinear response than large bandgap semiconductors.

On the other hand, nonparametric processes ($\hbar \omega > E_g$) such as absorption involve transfer of population from one real level to another. For all but shortest laser pulses, nonlinear response can be described in terms of conduction band electron population $N_c$ that obeys the rate equation:

$$\frac{dN_c}{dt} = \frac{\alpha I}{\hbar \omega} \left( N_c - N_c^0 \right) T_1$$
where $\alpha$ is the absorption coefficient at the applied frequency, $N_c^0$ is the conduction band electron population in thermal equilibrium, and $T_1$ is the response time or the electron-hole recombination or radiative lifetime. If we use laser pulse with duration shorter than $T_1$, the $N_c$ increases monotonically throughout the duration of the pulse. $N_c$ changes the optical response of the semiconductors via several mechanisms. Free electron response: conduction band electrons can respond freely to an applied electric field with response time much faster than $T_1$. This mechanism becomes significant for longer wavelengths. Plasma screening effect: conduction band electrons make semiconductor slightly conducting. As a result, charges flow to screen any unbalanced free charges, and the Coulomb interaction between charged particles is effectively weakened. One consequence of weakened Coulomb interaction is the disappearance of excitonic features at high electron densities. Band-filling effects: the absorption coefficient $\alpha$ of a semiconductor depends on the population difference between the valence band and conduction band, and so $\alpha$ should decrease as electrons are excited from the valence band to the conduction band. This is analogous to saturation effects in atomic systems, with additional complexity because now the electrons must be Pauli exclusion principle and thus must occupy a range of energies in the conduction band. For frequencies below (above), this effect lowers (raises) the refractive index. Bandgap renormalization: due to exchange interactions and Coulomb correlations, the bandgap of most semiconductors decreases at high concentrations of conduction band electrons, thereby, changing the optical response of the semiconductors.

For nonresonant excitation, one can use power series expansions to express optical response of the material in terms of powers of the applied field. For resonant excitation such power series expansion does not converge for laser excitations above saturation intensity, and so can not in general be used. However, under resonant excitations it is often sufficient to consider two levels
that are connected by the optical excitation, and it is not necessary to sum over all possible states that appear in general quantum mechanical perturbative calculations. In the next section, we shall discuss interaction of a monochromatic beam with a collection of two-level systems (for example, an ensemble of excitons in semiconductors).

For most of the cases a semiclassical treatment, where we consider matter states quantum mechanically and light classically, is enough. Consider the case where a monochromatic light of frequency $\omega$ interacts with a two quantum mechanical levels $|g\rangle$ and $|e\rangle$ of a system, and creates a superposition states. Hamiltonian of an electron in this case can be written as

$$H_{\text{total}} = H_0 + H_{\text{light-matter interaction}} + H_{\text{others}}$$

Light-matter interaction in the dipole approximation can be written as

$$\hat{H}_m = -\mathbf{e} \mathbf{r} \cdot \mathbf{E} = \begin{pmatrix} 0 & -\mu_{eg} \cdot \mathbf{E} \\ -\mu_{ge} \cdot \mathbf{E} & 0 \end{pmatrix}$$

where $\mu_{eg}$ is the dipole moment given by $\mu_{eg} = \langle e | \mathbf{r} | g \rangle$. If the dipole moment is nonzero, electron can be transferred optically from state $|g\rangle$ to $|e\rangle$, and it is called optically allowed transition or optical selection rule. Once we have $\bar{\mu}$, we can calculate macroscopic polarization using $P = \varepsilon_0 \chi \mathbf{E} = N \langle \bar{\mu} \rangle = \bar{\mu}_{eg} c_g(t) c_e^\dagger(t) + \text{c.c.}$. Real part of susceptibility $\chi$ gives refractive index, whereas imaginary part gives absorption coefficient of the material.

Usually we do not possess the full quantum mechanical knowledge about system (particularly solid state environment of semiconductors) because of decay mechanisms such as upper to lower level decay and more rapid decay of dipole coherence. Rather we know the classical probabilities for having various possible quantum mechanical states. In such cases density matrix approach is
quite effective in calculating material response. For example, a state vector
\[ |\psi\rangle = c_e(t)|g\rangle + c_c(t)|e\rangle \]
can be expressed in density matrix form as
\[
\hat{\rho} = |\psi\rangle \langle \psi| = \begin{pmatrix}
|c_e|^2 & c_c^* c_g \\
 c_g c_e^* & |c_g|^2
\end{pmatrix}
\]
where diagonal elements gives population of states and off diagonal elements represents coherence between states. Time evolution of the density matrix is given by
\[
\frac{\partial}{\partial t} \hat{\rho} = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}]
\]
With the condition \( \rho_{ee} + \rho_{gg} = 1 \) to normalize population, we can derive equations of motions for individual density matrix elements:
\[
\dot{\rho}_{eg} = (-i\omega - \gamma) \rho_{eg} - \frac{i\mu E}{\hbar}[\rho_{ee} - \rho_{gg}]
\]
\[
\dot{\rho}_{ee} = -\Gamma \rho_{ee} - \frac{i\mu}{\hbar}[\rho_{eg} E^* - c.c.]
\]
where we have introduced phenomenological dephasing rate \( \gamma \) and population relaxation rate \( \Gamma \).

From the equations of motions above, under steady state condition, for an applied field \( E = \frac{1}{2}(Ee^{-i\nu} + c.c.) \), we can derive real and imaginary part of the susceptibility \( \chi \):

\[
\text{Refractive index } n = \text{Re}(\chi) = \chi = a \frac{\Delta T_2}{1 + \Omega^2 T_1 T_2 + \Delta^2 T_2^2}
\]
\[
\text{Abs. coefficient } \alpha = \text{Im}(\chi) = \chi' = a \frac{1}{1 + \Omega^2 T_1 T_2 + \Delta^2 T_2^2}
\]
where “a” is a constant, Rabi frequency \( \Omega = \frac{\mu E}{\hbar} \), pump detuning \( \Delta = \omega - \nu \), dipole coherence decay rate \( \gamma = \frac{1}{T_2} \), radiative decay rate \( \Gamma = \frac{1}{T_1} \). Figure 9 shows the theoretical plot of the
absorption and refractive index of a two-level system. For a homogeneously broadened transition every two-level system of the ensemble has the same center frequency and lineshape.

**FIGURE 9.** Absorption and refractive index of a two-level system as a function of frequency of the applied field. Region where refractive index increases with frequency is called normal dispersion region. On the other hand, on resonance refractive index decreases with frequency and hence called anomalous dispersion region. Propagation of light shows different behaviour in normal and anomalous dispersion region.

Under the influence of a strong beam centered anywhere within the linewidth, susceptibilities will saturate uniformly across the entire linewidth for a homogeneously broadened transition, as shown in Figure 10. In contrast, for inhomogeneously broadened medium a strong beam at
frequency $\omega_1$ will saturate will for the subgroup of two-level systems whose frequencies are in resonance or nearly in resonance with the applied frequency $\omega_1$. As a result, for an inhomogeneously broadened medium, strong saturating beam will burn a hole (often called spectral hole burning or SHB) in the absorption line that can be observed by a weak probe beam, as shown in Figure 11.

**FIGURE 10.** Saturation in homogeneously broadened transition. For homogeneously broadened transition, intensity dependence of the (a) real (b) imaginary part of the susceptibility show uniform saturation.

For a sufficiently strong beam one might think that it will only decrease the overall response of the medium. However, it leads to something else also. Stark shifts due to optical field modify the energy level structure and give rise to new resonances in the optical susceptibility. In the absence of dephasing, population of a strongly driven (with detuning $\Delta$) two-level system oscillates (Rabi
oscillations) at the generalized Rabi frequency $\Omega_{\text{gen}} = \left(\Omega^2 + \Delta^2\right)^{1/2}$. Induced dipole moment oscillates not only at the applied frequency $\omega$, but also at the Rabi sidebands $\omega \pm \Omega_{\text{gen}}$.

**FIGURE 11.** Spectral hole burning in inhomogeneously broadened transition. For inhomogeneously broadened transition, overall lineshape (solid line) in (a) is a result of individual transitions with different transition frequency. $\Delta$ is the detuning of the light and $T_2$ is the dipole coherence time. In such cases, strong saturating beam will burn a hole (often called spectral hole burning or SHB) in the absorption line as shown in (b).

In the presence of dephasing, Rabi oscillations die out in a time given by the dipole dephasing time $T_2'$, and hence, Rabi oscillations are not present in the steady state. However, in the presence of two beams (such as in pump-probe spectroscopy) with frequency $\delta$, Rabi oscillations can be driven when $\delta = \Omega_{\text{gen}}$. In this case, the population oscillates at beat frequency $\delta$ and the induced dipole moment oscillates at $\omega$ and $\omega \pm \delta$. 
Excitonic transitions in GaAs QWs

An electron can be transferred from valence band to conduction band in semiconductors by optical excitations. As mentioned before, an electron (negatively charged) in the conduction band and a hole (positively charged) in the valence band can form an exciton (X) due to Coulomb interaction if the thermal energy ($k_B T$) is less than the exciton binding energy. Exciton energy and oscillator strength for 2D (QW) and 3D are given by

\[
E_n^{2D} = -\frac{E_B}{(n-1/2)^2} f_n^{2D} \sim \frac{P^2}{(n-1/2)^3}
\]

\[
E_n^{3D} = -\frac{E_B}{n^2} f_n^{3D} \sim \frac{P^2}{n^5}
\]

**FIGURE 12.** Absorption spectrum of a GaAs QW with 17.5 nm well width and multiple (50X) quantum well (MQW) at $T=20$ K. HH and LH excitonic transitions for $n=1$ and $n=2$ can be seen clearly.

Above equations show that the exciton binding energy increases by a factor of 4 for 2D compared to 3D. This enhancement of binding energy results from the larger overlap of the electron and
hole wavefunction in 2D, despite possible penetration of the electron/hole wavefunction into the barrier region. This increased binding energy, typically around 10 meV depending on quantum well width, allows the excitonic transitions separately from the continuum states. Figure 12 shows the absorption spectrum of a multiple quantum well (MQW) of GaAs with 50 wells of width 17.5 nm each. Both heavy-hole and light-hole excitonic peaks can be clearly seen for n=1 and n=2.

Pump-probe spectroscopy

Nonlinear phenomena can be used in two general ways. One can use them to applications such as second-harmonic generation, lasers, phase conjugation, optical bistability etc.

\[
\alpha(\nu) L = -\ln \left( \frac{I(\nu)}{I_0(\nu)} \right)
\]

FIGURE 13. A Schematic of pump-probe spectroscopy setup. Together pump and probe can create population pulsation that leads to coherent effects such as ac Stark shift (off-resonant pump), dynamic Stark splitting (on-resonant pump). Overall lineshape of the probe transmission depend both on dipole coherence time \( T_2 \) and electron-hole recombination time \( T_1 \).
On the other hand, nonlinear phenomena can be used as a spectroscopic tool to study the properties of the medium that generates them. In the simplest case, a single beam itself is of substantial use in various spectroscopic techniques, laser theory and optical bistability. Even though interesting spectroscopic studies are possible with more than two beams, typically two-beam pump-probe spectroscopy is quite useful particularly as a spectroscopic tool for coherent optical phenomena. In pump-probe experiment, one measures how the response of the medium to a (typically weak) probe is modified the presence of a (typically strong) pump beam. We consider a medium subjected to a strong saturating pump beam of frequency $\nu_1$ and a weak nonsaturating probe beam of frequency $\nu_2$, as shown in Figure 13.

Overall response of the medium along probe direction $\tilde{k}_2$ contains both incoherent and coherent parts. For cw laser excitations with a weak probe and strong pump, an analytical expression for the absorption coefficient $\alpha$ (along $\tilde{k}_2$) can be calculated [49] and is given by

$$\alpha(\text{probe}) = \alpha_{\text{inc}} + \alpha_{\text{coherent}} \sim \rho_{eg} = \frac{i \Omega_{\text{probe}} d_0 + \Omega_{\text{pump}} d_{-1}}{2 \gamma i + \Delta_{\text{probe}}}$$

$$d_0 = \frac{1}{1 + \left(\frac{\Omega_{\text{pump}}}{\Gamma_c}\right)^2 \gamma^2 + \Delta_{\text{pump}}^2}$$

$$d_{-1} = -\frac{\Omega_{\text{pump}} \Omega_{\text{probe}}}{2} F(\Delta) \frac{D_{\text{probe}} + D_{\text{pump}}^*}{1 + \frac{\Omega_{\text{pump}}^2}{2} (D_{\text{probe}} + D_{\text{pump}}^*)} d_0$$

where $F(\Delta) = \left(\frac{1}{\Gamma_c + i\Delta} + \frac{1}{i\Delta}\right), D_j = \frac{1}{\gamma + i\Delta_j}, \Delta = \nu_{\text{pump}} - \nu_{\text{probe}}, \Delta_j = \Delta_{\text{pump}} - \Delta$. Overall features of transmission spectra depend strongly on relative time scales of $T_1$ and $T_2$, pump
intensity and detuning, and can be used as a spectroscopic tool for studying coherent phenomena and their time scales. Both cw and pulse lasers can be used for pump-probe spectroscopy revealing wealth of information. Similar expression can be obtained for coherent four-wave mixing (FWM) signal along $2\vec{k}_1 - \vec{k}_2$ and $2\vec{k}_2 - \vec{k}_1$ directions. Pump-probe spectroscopy has been used extensively both in the transmission and FWM geometries to study nonlinear optical properties of semiconductors.

**Summary**

In this chapter, we have discussed the similarities and differences of excitons (bound electron-hole pairs) created by optical excitation with respect to atomic systems. Even though excitons share many features similar to atomic systems, many-body interactions such as exciton induced dephasing (EID), carrier-carrier scattering etc. affect the optical response of the semiconductors [48]. Band structure and optical selection rules of GaAs QWs [45-46] have been discussed. We have discussed light-matter interaction in semiconductors considering excitons as two-level systems [44]. In the end, pump-probe spectroscopy as a tool of studying nonlinear optical processes has been mentioned briefly.
CHAPTER III

ELECTROMAGNETICALLY INDUCED TRANSPARENCY

In this chapter, we will discuss the nonlinear optical phenomenon called electromagnetically induced transparency (EIT). EIT has already been used for dramatic dynamic control of group velocity of light [12], and EIT is one of the approaches pursued in our research.

Introduction

Since EIT was first reported in 1991 [86], electromagnetically induced transparency (EIT) has generated profound interests in the areas of atomic dynamics, pulse propagation, enhanced frequency conversion, quantum information processing [80-83]. Interference between alternative pathways in quantum-mechanical processes leads to many interesting effects one of which is EIT. By EIT we mean a process where optically induced quantum interference makes a material transparent to a wavelength which the material would otherwise absorb. EIT may be observed in three-level system, such as the Λ -type system shown Figure 14. Two optically allowed transitions (dipole transitions) $|a\rangle \rightarrow |e\rangle$ and $|b\rangle \rightarrow |e\rangle$ share a common state. Probability amplitudes of two transition pathways to the common state $|e\rangle$ can interfere and lead to a vanishing net transition amplitude. When probe acts alone material absorbs the probe beam, but when probe acts together with the pump beam sharing a common state material becomes transparent due to quantum interference, as shown in Figure 14. The destructive quantum interference in EIT is the direct consequence of the nonradiative (Raman) coherence induced
between $|a\rangle$ and $|b\rangle$. Many quantum interference effects related to nonradiative coherence and EIT have been studied in atomic systems. As shown in Figure 14 (b), we should also note that large changes in the absorption spectrum due to EIT are accompanied by corresponding changes in refractive index.

**FIGURE 14.** Effect of EIT in three-level systems. (a) Response of the material when probe acts alone showing usual absorption and refractive index as a function of frequency. (b) Response of the material when probe acts together with pump sharing a common state. Quantum interference between two pathways to excited state leads to transmission dip in the absorption spectrum.
In the presence of EIT refractive index changes sign of its slope and can become very steep depending on the width of the EIT window. This has been used to slow down the group velocity of light down to few meters per second. EIT has been used to stop, store and retrieve light in atomic systems. Related phenomenon of adiabatic population transfer can be used to transfer population between two states that are not optically coupled (zero dipole moment). There are studies on lasing without inversion based on the generation of nonradiative coherence.

Nevertheless, EIT and related phenomena are distinct from other quantum interference based phenomena such as coherent population trapping (CPT) [84] because EIT occur in optically dense medium ($\alpha L \geq 1$). In contrast to mainly spectroscopic tools such as CPT occurring in optically thin medium ($\alpha L \ll 1$) where only modification of states of matter happens, EIT is associated with modifications of both the states of matter and the optical fields used. Modifications of both states of matter and optical fields due to EIT makes it a new means to change the optical characteristics such as absorption, refraction, group velocity etc. and so provides a better way to manipulate propagation of optical fields and to enhance the generation of new optical fields.

Observations of EIT and related phenomena in atomic systems motivate us to extend those studies in semiconductors. On one hand, excitonic transitions in semiconductors have similarities with two-level atomic systems. On the other hand, various interactions such as many-body interactions in solid state environment of semiconductors lead to more complexities and differences as compare to atomic systems. While similarities with atomic systems have guided us towards possible studies, it is particularly the complexities and differences due to interactions such as many-body effects that we have used to our advantage. Semiconductors provide an excellent platform to study many-body physics. In this regard, EIT can use as a spectroscopic tool to investigate many-body effects. EIT in semiconductors using excitonic coherence has already been observed and used as a spectroscopic tool to study various types of coherences and many-
body effects. Excitonic coherence is, however, quite fragile and leads to limited degree of EIT. Moreover, excitonic coherence is almost negligible at room temperature at which most of the device applications (e.g., tunable delay based all-optical buffer) work. This led to our research of using more robust electron spin coherence in semiconductors to observe EIT. Not only electron spin coherence persists even up to room temperature, robust electron spin coherence would manifest itself as a narrow transmission (EIT) window that can used to dramatically control group velocity of light. We have taken advantage of the spin-orbit coupling in the valence band to couple two electron spin states to a common light-hole valence band state in a GaAs QW waveguide. Induced transparency due to electron spin coherence in optical absorption has been demonstrated by investigating both the polarization and magnetic field dependence of the induced transparency (Chapter 6). In the next section, we will present a general discussion of EIT and its relation to other coherent phenomena.

**FIGURE 15.** Quantum interference of transition pathways. Interferences of pathways into continuum: (a) single autoionizing state, (b) two autoionizing states.
EIT and related coherent phenomena

Interference between the excitation channels to the continuum can be constructive or destructive and thus leads to frequency dependent enhancement or suppression of the photoionization. These are called Fano interferences. Figure 15 shows Fano interferences involving one and two autoionizing states.

For the second case, for significant interference two states should be closely spaced compared to their widths. Interference between two closely spaced lifetime-limited resonances decaying to the same continuum, can lead to lasing without inversion. In this case interference between two channels eliminates absorption while leaving stimulated emission from two states unchanged. A significant realization was that two closely spaced lifetime-limited resonances were equivalent to dressed states created by coupling two well separated atomic levels with a resonant laser. Their work showed that required energy level structure for quantum interference can be engineered optically. We must mention that energy level structure can also be engineered using static magnetic fields (Zeeman effect) or static electric fields (Stark effect) and there have been extensive studies. Using their proposal they reported highly enhanced frequency conversion in four-wave mixing. While linear susceptibility is cancelled, nonlinear susceptibility is enhance due to quantum interference and thus leads to enhanced frequency conversion. In this context the term EIT was first introduced. First experimental observation of EIT was reported in Sr vapor by Boller et al. in 1991 [86].

Three types of level structures can be used to study EIT: $\Lambda$, cascade and $V$ systems. $\Lambda$-type system is the best EIT while $V$ – type system is the worst [89]. It is instructive to explain EIT from different point of views to elucidate the differences and similarities with other quantum interference phenomena. There are several informative ways: interference between dressed states, multiple routes to excitation, coherent population trapping and so on. Boller et al. [86] presented
two viewpoints to explain EIT. First viewpoint is in terms of dressed states. As shown in Figure 16, dressing laser creates two closely spaced states decaying to the same continuum. If we tune probe exactly on zero-field resonance, then contributions of two dressed states to linear susceptibility are equal and opposite and thus cancel each other giving rise to EIT.

Another viewpoint is in terms of bare states instead of dressed states. Here EIT can be viewed as arising from different pathways between bare states. The probe absorption given by transition amplitude to \( |3\rangle \), is driven by two pathways- \(|1\rangle - |3\rangle\) and \(|1\rangle - |3\rangle - |2\rangle - |3\rangle\). Probability amplitudes for both direct and indirect pathways can be equal and amplitude on resonance because control is much more intense than probe.

EIT can also be explained in the context of dark states and coherent population trapping (CPT). CPT was first experimentally observed by Alzetta et al. Their experimental results were
theoretically explained considering a $\Lambda$-system and using the concept of CPT in dark eigenstate of the Hamiltonian $H = H_0 + H_{\text{int}}$, where $H_{\text{int}}$ is the matter-optical field interaction.

Hamiltonian of a three-level system interacting with probe and control fields in a rotating frame and under rotating wave approximation (RWA) can be written as

$$H_{\text{int}} = -\frac{\hbar}{2} \begin{pmatrix} 0 & 0 & \Omega_p \\ 0 & -2(\Delta_p - \Delta_c) & \Omega_c \\ \Omega_p & \Omega_c & -2\Delta_p \end{pmatrix}$$

where $\Delta_p$ and $\Delta_c$ are the probe and control detunings respectively. Eigenstates of this Hamiltonian can be written in terms of bare states as follows:

$$|a^+\rangle = \sin \theta \sin \varphi |1\rangle + \cos \varphi |3\rangle + \cos \theta \sin \varphi |2\rangle$$

$$|a^0\rangle = \cos \theta |1\rangle - \sin \theta |2\rangle$$

$$|a^-\rangle = \sin \theta \cos \varphi |1\rangle - \sin \varphi |3\rangle + \cos \theta \cos \varphi |2\rangle$$

where $\theta$ and $\varphi$ are mixing angles. For two-photon resonance ($\Delta_p = \Delta_c$), mixing angles are given by:

$$\tan \theta = \frac{\Omega_p}{\Omega_c}$$

$$\tan 2\varphi = \frac{\sqrt{\Omega_p^2 + \Omega_c^2}}{\Delta_p}$$

Note that the state $|a^0\rangle$ does not contain $|3\rangle$ and therefore, if the system is in this state there is no possibility of optical transition to $|3\rangle$ and subsequent spontaneous decay. State $|a^0\rangle$ is called dark state. 3-level system can be in one particular or in a superposition of above three eigenstates. Optical transition to $|3\rangle$ and subsequent spontaneous decay from $|3\rangle$ to the dark state $|a^0\rangle$ is one way to trap population in the dark state. The time to optically pump population to dark state this
way is given by radiative lifetime and so time is not controllable at will. EIT is an alternative way
to trap population in a dark state within a rapid and controllable time in an adiabatic manner.

To relate EIT with dark state above in the dressed state picture above, we consider a weak probe
such that $\Omega_p \ll \Omega_c$. In this case $\sin \theta \to 0; \cos \theta \to 1$ and we have,

$$\begin{align*}
|a^+\rangle &= \cos \varphi |3\rangle + \sin \varphi |2\rangle \\
|a^0\rangle &= |1\rangle \\
|a^-\rangle &= -\sin \varphi |3\rangle + \cos \varphi |2\rangle
\end{align*}$$

Interestingly the ground state itself is now the dark state from which there would be no transition
to excited state. For resonant probe i.e. $\Delta_p = 0$, we have usual normalized dressed states

$$\begin{align*}
|a^+\rangle &= \frac{1}{\sqrt{2}} (|3\rangle + |2\rangle) \\
|a^-\rangle &= \frac{1}{\sqrt{2}} (-|3\rangle + |2\rangle)
\end{align*}$$

discussed earlier relevant to EIT. Dark state picture also shows that the EIT interference can be observed even when both probe and control are strong and is useful for creating maximal coherence.

To ensure formation of dark states adiabatic turn on of the fields can be used, for example, using
so called counterintuitive pulse sequence. Starting with the whole population in ground state $|1\rangle$
control field $\Omega_c$ is applied first. This is similar to EIT situation because $\Omega_p \ll \Omega_c$ and dark state
 corresponds to $|1\rangle$ with negligible or no population in $|2\rangle$ and $|3\rangle$. Now probe field $\Omega_p$ is
gradually (i.e. adiabatically) increased and at the same time control field $\Omega_c$ is adiabatically
decreased; thus eventually satisfying $\Omega_c \ll \Omega_p$ and the dark state becomes $|a^0\rangle = -|2\rangle$. This
process is sometimes called stimulated Raman adiabatic passage (STIRAP). Thus population has
been transferred from $|1\rangle$ to $|2\rangle$ via maximally coherent (corresponding to the point of time
when $\Omega_c = \Omega_p$) dark state.
EIT based on the effects of the nonradiative coherence can also be conveniently studied using OBE. Let us consider a Λ-type three-level system as shown in Figure 17. Similar treatment can be applied to V-type and cascade type three-level systems. Apply a weak probe beam \( \Omega_a \) and a strong pump beam \( \Omega_b \). Assume that initially all the population is in state \( |a\rangle \). Using density matrix approach, we can derive the equations of motions of for dipole coherence and nonradiative coherence as follows:

\[
\begin{align*}
\text{Dipole coherence: } \dot{\rho}_{\text{e}}^{(l)}(t) &= (i\delta_a - \gamma_a)\rho_{\text{e}}^{(l)}(t) + \frac{i\Omega_a}{2} + \frac{i\Omega_b}{2} \rho_{ba}^{(l)}(t) \\
\text{Nonradiative coherence: } \dot{\rho}_{ba}^{(l)}(t) &= i(\delta_a - \delta_b)\rho_{ba}^{(l)}(t) + \frac{i\Omega_a}{2} \rho_{ba}^{(l)}(t)
\end{align*}
\]

As evident from above equations, to observe EIT we need to generate nonradiative coherence \( \rho_{ba}^{(l)} \) in the system which in turn requires that both pump and probe act on the system. Solving...
above equations to the first order in probe and to the all order of the pump, under steady state condition, we have:

\[
\rho_{aa}^{(l)} = \frac{-\alpha_0}{\Omega_0^2 \left[ i(\delta_a - \delta_b) - \gamma_{ba} \right] + \frac{(i\delta_a - \gamma_a)(i(\delta_a - \delta_b) - \gamma_{ba})}{4}}
\]

Absorption coefficient \((\alpha)\) as a function of probe detuning \(\delta_a\) is given by

\[
\alpha(\delta_a) \propto \text{Im} \left[ \frac{\rho_{aa}^{(l)}}{\Omega_0} \right]
\]

Depth and width of the transparency window are given by

\[
\text{EIT(depth)} \rightarrow \alpha(0)/\alpha_0 = \frac{1}{1 + \frac{\Omega_0^2}{4\gamma_a\gamma_{ba}}}
\]

\[
\text{EIT(width)} \rightarrow \Delta \omega_{\text{FWHM}} = \frac{1}{4} \frac{\Omega_0^2 + 4\gamma_a\gamma_{ba}}{\gamma_a + \gamma_{ba}}
\]

Efficiency of EIT depends on the intensity of the pump beam. Figure 18 shows EIT in a \(\Lambda\)-type three-level system for different intensities of the pump beam. EIT dip is strongly dependent on the decoherence rate \((\gamma_{ba})\) for the nonradiative coherence generated. Magnitude of nonradiative coherence is maximum when pump detuning and probe detuning are equal in magnitude because then two photon resonance condition between nonradiatively coupled transitions is satisfied. In order to achieve more than 50% reduction in absorption, we need \(|\Omega_{\text{pump}}| > 2\sqrt{\gamma_{\text{dipole}}\gamma_{\text{nonradiative}}}
\).

Absorption spectra modified by EIT reflect the destructive interference caused by the nonradiative coherence; it is not the sum of two Lorentzian peaks. For a given pump intensity, width of the EIT window is given by the larger of nonradiative decoherence rate and pump linewidth. EIT experiments pulsed lasers can be formulated as is done above for experiments in steady state using CW lasers, but have to be solved numerically in general.
FIGURE 18. EIT in a Λ-type three-level system for different intensities of the control (pump) beam. Corresponding Rabi frequencies are $\Omega_{\text{pump}} = 0$ (dash), $\Omega_{\text{pump}} = 0.2 \gamma_{\text{dipole}}$ (solid), $\Omega_{\text{pump}} = 2 \gamma_{\text{dipole}}$ (open circle), $\Omega_{\text{pump}} = 4 \gamma_{\text{dipole}}$ (close circle).

EIT using excitonic coherences have been experimentally observed using pump pulses of a few ps duration and probe pulses of ~ 200 fs duration. For better EIT duration of pump pulse should be long compared to dipole decoherence time, but still short enough to induce Rabi frequency greater than dipole decoherence rate. The depth of EIT dip strongly depends on pump-probe delay and is not symmetric around zero delay. EIT dip is deepest when the probe comes slightly before pump. EIT signatures using electron spin coherence in semiconductor using CW excitations have also been observed and presented in Chapter VII.

**EIT and blue shift: effect of chirped pulse**

Blue shift of excitonic transitions due to laser excitation is well known. Even if laser is on resonance initially, blue shift makes it off resonance, and so the efficiency of EIT goes down.
Idea is to introduce chirp in the laser pulse so that the red comes first and then the blue in such a way that excitation is always on resonance.

For a cascade type three-level system (where a, e and b are the lower, middle and upper levels respectively) with two applied fields (probe between a and e states and pump between e and b states), total Hamiltonian can be written as

\[
H = \begin{pmatrix}
\hbar \omega_a & 0 & -\mu_a E_a \\
0 & \hbar \omega_e & -\mu_e E_e \\
-\mu_a E^*_a & -\mu_e E^*_e & \hbar \omega_e
\end{pmatrix}
\]

where \( \mu_a \) and \( \mu_b \) are the dipole matrix elements given by \( \overline{\mu_a} = \langle a | e r | e \rangle \) and \( \overline{\mu_b} = \langle b | e r | e \rangle \) along the electric fields \( \overline{E}_a \) (probe) and \( \overline{E}_b \) (pump) respectively. The applied electric fields are given by

\[
E_a(t) = \frac{1}{2} E_a \left( e^{i \nu_a t} + e^{-i \nu_a t} \right)
\]

\[
E_b(t) = \frac{1}{2} E_b \left( e^{i (\nu_b t + \phi(t))} + e^{-i (\nu_b t + \phi(t))} \right)
\]

where \( E_a(t) \) and \( E_b(t) \) are time dependent amplitudes of probe and pump beam respectively, \( \nu_b \) is the center frequency of the pump, and a linear chirp is introduced via \( \phi(t) = bt^2 \). A positive chirp parameter \( b \) means higher frequencies (blue) come later. We will refer to the electric fields in terms of their Rabi frequencies

\[
\Omega_a(t) = \frac{\mu_a E_a(t)}{\hbar}
\]

\[
\Omega_b(t) = \frac{\mu_b E_b(t)}{\hbar}
\]
Evolution of the three-level system can be represented by the evolution of the density matrix of the form:

$$\rho = \begin{pmatrix} \rho_{ee} & \rho_{ea} & \rho_{eb} \\ \rho_{ae} & \rho_{aa} & \rho_{ab} \\ \rho_{be} & \rho_{ba} & \rho_{bb} \end{pmatrix}$$

The diagonal elements give the populations of respective levels. “Dipole Coherence” is given by the matrix elements $\rho_{ea} = \rho_{ae}^*$ and $\rho_{eb} = \rho_{be}^*$, whereas “Nonradiative Coherence” is given by $\rho_{ba} = \rho_{ab}^*$. The equations of motion for the density matrix elements are given by

$$\dot{\rho}_{ij} = -\frac{i}{\hbar} \sum_k (H_{ik} \rho_{kj} - \rho_{ik} H_{kj}); \ k = a, b, e$$

Using the constraints $\rho_{aa} + \rho_{bb} + \rho_{ee} = 1; \rho_{ab} = \rho_{ba}^*; \rho_{ae} = \rho_{ea}^*; \rho_{be} = \rho_{eb}^*$ and making following assumptions to factor out the rapidly varying components which oscillates at the optical frequencies:

$$\rho_{ea} = \rho_{ae} e^{-i\eta_{ad}}$$
$$\rho_{be} = \rho_{be} e^{-i(\nu_a \tau + \phi(t))}$$
$$\rho_{ba} = \rho_{ba} e^{-i((\nu_a + \nu_b) \tau + \phi(t))}$$

For convenience, we also make some additional replacements: $\rho_{aa} = n_a; \rho_{bb} = n_b; \rho_{ee} = n_e$.

Using the Hamiltonian and the above substitutions for the density matrix elements we can get the OBE for cascade type level structures. Under rotating wave approximation we drop rapidly varying terms. We set $n_a + n_b + n_e = 1$ to normalize the total populations to one. Finally, we take into account various decay mechanisms of populations and coherences by introducing phenomenological decay constants.
We consider the case where probe is weak and pump is strong, and we solve OBE up to the first order in probe and to the all order of pump. To zeroth order in probe \( n_a^{(0)} = 1 \), and all other terms are zero. Note that this is a direct consequence of upper two states being unoccupied in the absence of probe, so that the pump is acting on an “empty” transition. This would not be the case for a V system.

To the first order in probe and all orders in pump, including chirp in the pump and blue shift of the transition (due to band renormalization), we have the equations of motion for relevant matrix elements as follows:

\[
\begin{align*}
\text{Dipole Coherence:} & \quad \dot{\rho}_{aa}^{(1)}(t) = (i\delta_a - \gamma_a)\rho_{aa}^{(1)}(t) + \frac{i\Omega_a}{2} + \frac{i\Omega_a}{2}\rho_{rr}^{(1)}(t) \\
\text{Nonradiative Coherence:} & \quad \dot{\rho}_{rr}^{(1)}(t) = \left\{ i(\delta_a + \delta_b + \dot{\phi}(t) - \beta|\Omega_b(t)|^2) - \gamma_r \right\}\rho_{rr}^{(1)}(t) + \frac{i\Omega_b}{2}\rho_{aa}^{(1)}(t)
\end{align*}
\]

where \( \delta_a = \nu_a - \omega_a \) (probe detuning); \( \delta_b = \nu_b - \omega_b \) (pump detuning); \( \dot{\phi}(t) = 2bt \) (for linear chirp); \( \gamma_a \) is the dipole decay time (a-e transition); \( \gamma_r \) is the nonradiative or Raman coherence decay time (a-b transition); probe Rabi frequency \( \Omega_a = \Omega_{0a} e^{-\nu_a t} \); pump Rabi frequency \( \Omega_b = \Omega_{0b} e^{-\nu_b t} \); \( t \) is the pump-probe delay.

Figure 19 shows the numerical calculation for conditions (in the units of dipole coherence time \( \gamma_a \) ) given below:

\[
\begin{align*}
\gamma_a = 1, \gamma_r = 0.002, \text{probe pulse width} = 1, \text{pump pulse width} = 40, \\
\Omega_{\text{probe}} = 0.05, \Omega_{\text{pump}} = 3, \delta_{\text{pump}} = 0, t_{\text{pump-probe delay}} = 0,
\end{align*}
\]

chirp parameter \( b = 0.01 \), renormalization \( \beta = 0.3 \)

From the numerical calculations using parameter given above, it is clear that the blue shift due to band edge renormalization arising from laser excitation can be eliminated using a linearly chirped pump beam. But chirped pump beam does not improve EIT effect (i.e. more transparency).
To introduce a linear chirp experimentally, it can be sufficient to insert a glass wedge (~1°) into the spectral filter setup that we have used to derive 6 ps pump from 150 fs laser output. Considering $\alpha \approx 0.01 cm^{-1}$ for glass, we see that glass wedge will introduce almost no amplitude modulation. This section not only motivates the use of chirped pulse for experimental studies of EIT and blue shift, but also provides a very simple way to generate chirped pulse.

**FIGURE 19.** Effect of chirped pump and blue shift on absorption spectrum. Dashed curve is the linear absorption in the absence of the pump. (a) Absorption spectrum when both blue shift and chirp are absent. EIT dip is at zero detuning. (b) Band edge renormalization due to laser excitation shifts resonance to blue side, thereby, shifting the EIT dip to blue. (c) Effect of renormalization can be eliminated using a chirped pump, even though efficiency of EIT does not improve.
Summary

In this chapter, we have discussed EIT [80, 85]. Related topics such as coherent population trapping [84], dark states [85, 87], and adiabatic population transfer [88] have been discussed in the context of EIT. Theoretical analysis of EIT in a $\Lambda$-type three-level system has been given. One issue in semiconductors is blue shift of the resonances that make initially on-resonant pump become off-resonant. We have analyzed how this can be cancelled using chirped pulses.
CHAPTER IV

EXPERIMENTAL TECHNIQUES

In this chapter, we will discuss some important features of the experimental techniques used. We will also discuss general experimental techniques developed in our laboratory for doing optical spectroscopy in semiconductor waveguides. Our studies of EIT arising from electron spin coherence and Rabi oscillations arising from heavy-hole excitonic coherence will be discussed in detail in later chapters.

Laser systems and pulse shaper

For all-optical tunable delay experiments two pulsed lasers were used. Signal pulses and reference pulse was derived from a 150 fs mode-locked pulsed Ti: sapphire laser (Spectra-Physics Tsunami) pumped by Coherent Verdi V10 at 532 nm. Pump power of 7 W from Verdi generates around 1.2 W mode-locked pulsed average power output at 800 nm. Control beam is derived from a 60 ps mode-locked pulsed Ti:sapphire laser (Spectra-Physics Tsunami) pumped by Spectra-Physics Millennia Pro at 532 nm. Pump power of 5 W from Millennia generates around 1.2 W mode-locked pulsed output at 800 nm. For our experiments both the pulsed lasers were synchronized at a repetition rate of 80 MHz.

For electron spin coherence studies in waveguides two CW lasers were used. New Focus Velocity Tunable Diode Laser (Model 6317) with resolution 10 MHz and linewidth less than 300 kHz was used probe. Typical output 5-7 mW. A tunable Coherent Ti:Sapphire laser (Model 899) pumped by Coherent Verdi-5W at 532 nm was used as pump. Typically output for single mode operation
of Coherent 899 is 350 mW with 5 W pump power with both resolution and linewidth around 10 MHz.

Signal pulses of 6-8 ps duration were derived from 150 fs pulses coming out of Tsunami by using a pulse shaper (spectral filter). Idea is to cut out frequency components so that pulses are broadened in time-domain according to transform-limited or energy-time uncertainty relation. The pulse shaper consists of a diffraction grating (Spectrogon gold coated blazed grating at near infrared with 1800 lines/mm), a high power biconvex lens (focal length 30 cm), a slit (with both width and position adjustments), and a mirror as shown in Figure 20. The input beam is diffracted by the grating and undergoes through angular separation of frequency components. Angle of incidence measured with respect to grating normal should be more than 60 degree, so that the input beam covers more grating lines and get more dispersion. For an input of 1W (150 fs 80 MHz Tsunami) with horizontal polarization, grating output in the dispersive direction is more than 650 mW. Some power is lost in the direction of specular reflection. Angular separation of the frequencies is made spatial separation by a putting the lens one focal length away. On the

**FIGURE 20.** A schematic of the pulse shaper used in our experiments.
other side of the lens, we put a mirror one focal length away so that each frequency was focused to a point in the image plane within the resolution limit. Slit was made with two razor blade (each one in separate 1-d stages) and placed closed to the mirror around 1 cm away to narrow down the spectral bandwidth of the pulses. Remaining spectral components reflects from the mirror and follows the reverse path to be recombined by the grating again. By introducing a small vertical tilt of the mirror, the incident and the filtered beam could be separated spatially.

Relative distances of grating, lens, slit and mirror are very important to avoid pulse broadening due to uncompensated dispersion. For initial adjustment an autocorrelator was used to set the distances. To check if the pulses coming out of the spectral filter are transform-limited, pulses were measured both in the frequency domain (Monochromator TRIAX 320 and PMT R928) and in time-domain by cross-correlation with 150 fs pulses (BBO SHG crystal). Less than 10% deviation from the theoretical value was obtained assuming Gaussian shape of the pulses. The minimum spectral bandwidth obtained was approximately 0.2 nm in frequency domain and 6 ps in the time-domain. Closing the slit further would cause diffraction due to slit.

**Time-domain pump-probe spectroscopy**

Schematic of the experimental setup is shown in Figure 21. The output from the 150 fs Ti:sapphire pulsed laser was split into two parts. One part is used as 150 fs probe; the other part is fed to spectral filter to get pump pulses of 6-8 ps duration. One of the beams goes through retroreflective mirror put on a delay stage with 1 μm resolution. The zero-delay position between pump and probe is determined by cross-correlation using a BBO second harmonic generation (SHG) crystal to generate a sum-frequency signal.

Desired polarizations of the pump and probe are set by using polarization optics. Variations of intensity for circular polarizations are less than 5%, and for linear polarizations maximum to minimum (extinction ratio) ratio are at least 1000:1. The pump and probe are focused on the
sample to spot diameter typically around 40-100 \( \mu m \) using plano-convex lenses with focal lengths around 7.5-20 cm. Probe is usually focused to smaller diameter than pump so that probe samples only the central uniform region of intensity of the pump.

We used epitaxially grown GaAs QW samples. For transmission measurements samples were mounted on c-axis normal sapphire disc and substrates of QW were removed by first lapping and then using selective chemical etching. Experiments were typically done at low temperatures with liquid Helium using Janis ST-300 cryostat. The probe beam transmitted through the sample was spectrally resolved using a monochromator (TRIAX 320) and detected using a photomultiplier tube (PMT Hamamatsu R928). Absorption spectra with or without the pumps were calculated using:

\[
\alpha(\lambda) L = -\ln \left[ \frac{I(\lambda)}{I_0(\lambda)} \right]
\]

where \( \alpha(\lambda) \) is the absorption coefficient as a function of the wavelength \( \lambda \), \( L \) is the thickness of the sample, \( I(\lambda) \) is the probe transmission measured after passing through the sample, \( I_0(\lambda) \) is

![FIGURE 21. A schematic of the pump-probe experiments in time-domain using pulsed lasers.](image-url)
the probe intensity before passing through the sample, approximated by measuring the probe transmission only through the sapphire disc. Absorption/absorbance in this dissertation refers to the quantity $\alpha(\lambda)L$.

**Optical spectroscopy in GaAs QW waveguide**

GaAs quantum well waveguides have two exclusive features: unique optical selection rules accessible only in the waveguide configuration when light propagates perpendicular to the growth direction and variable optical interaction length determined by waveguide length. These features are not available in usual normal configuration experiments where light propagates along growth direction. Unique optical selection rules can be used to induce electron spin coherence without magnetic field, whereas, long optical interaction length can be used to slow, store and retrieve light among other potential applications of waveguide configuration.

First measurements of linear and nonlinear absorption of light propagating along the plane of a single quantum well in waveguide configuration were reported two decades ago in 1985 [96-97]. However, due to experimental difficulties nonlinear spectroscopic studies of coherent effects in semiconductor waveguides particularly at low temperature were not reported since then. Some studies on GaAs/AlGaAs [98] and wide bandgap GaN/AlGaN [99] QW waveguides have been reported, but most of them are at room temperature and do not address coherent nonlinear effects. Moreover, due to strong dephasing in solid state environment of semiconductors it is more convenient to work at low temperature.

We have developed new experimental techniques for nonlinear optical pump-probe spectroscopic studies of coherences in semiconductor waveguides at wide range of temperatures (4.2 K to room temperature). We have already been able to observe Rabi splitting which is a precursor to further quantum coherent control studies and to induce electron spin coherence without magnetic fields. In this chapter we will discuss various studies on semiconductor waveguides.
Making waveguides and coupling light

Experimental studies are done on slab waveguides made from an MBE grown structure consisting of a single quantum well (SQW). To make waveguides, samples are thinned down to ~ 100 μm on the substrate side using aluminum oxide 12 μm polish film. Thinned down samples are then cleaved into waveguide pieces of desired lengths using surgical blade. Waveguides are mounted on brass mounts using silver paint. New Focus Velocity 6317 Tunable Diode Laser and Coherent 899 Ti: Sapphire Tunable solid state laser is used as probe and pumps lasers respectively.
FIGURE 23. Absorption spectrum of a waveguide made from 17.5 nm single QW. (a) Optically allowed transitions in GaAs QWs. $z$ is the growth direction. For $z$-polarization, light has to propagate in the QW plane perpendicular to $z$-axis. (b) and (c) Absorption spectra for a slab waveguide of length ~ 100 µm made from QW sample with single QW of width 17.5 nm. Note that for TM polarization HH transition is negligible.

Basic idea of coupling light through the slab waveguides made from QW sample is shown in Figure 22 (c). Light is focused by Mitutoyo M Plan Apo 20X objective to ~ 1 µm beam radius on the front facet of the waveguide. Output is collected and collimated by Geltech 350110-B Molded Glass aspheric lens placed one working distance away from the back facet of the
waveguide. Thus, we can separate the guided output from scattered light and observe a single bright line ~ 1 m away from the output lens corresponding to the lowest order mode of the leaky slab waveguide. A continuous flow, liquid-He cryostat (Janis ST-300) is used to control the sample temperature. Waveguide mount and output lens with translational stage are assembled on a single mount and attached to the cold finger of the cryostat.

![Graph showing absorption spectra](image)

**FIGURE 24.** Saturation of excitonic transition in waveguide. Absorption spectrum for input of average intensity less than 1 W/cm² (solid circle) shows LH excitonic transition clearly, whereas, for an input of average intensity around 1 kW/cm² (open circle) almost bleaches the transitions. Saturation of LH transition is shown for 100 µm waveguide at room temperature.

Absorption spectra of GaAs QW waveguide

Width of the MBE grown quantum well sample used is 17.5 nm. Sample structure is shown in Figure 22 (a). Absorption spectrum of a single quantum well of width 17.5 nm when light propagates along growth direction is shown in Figure 22 (b). Note that αL is less than 1,
linewidths of the excitonic resonances are around 0.3 nm. Absorption spectrum of the same sample when light propagates in the QW plane (perpendicular to growth direction) is shown in Figure 23. In this geometry, the sample acts like a waveguide. We used slab waveguides from QW sample. In the context of waveguide, horizontal $z$-polarized light ($z$ is the growth direction) is called TM and vertically polarized light is called TE. Even though absorption coefficient $\alpha$ is small, $L$ for a waveguide is much higher compared to quantum well width of 17.5 nm. In the Figure 23 data are shown for a waveguide of length 100 $\mu m$. According to Beer’s law $I_{\text{out}}(\lambda) = I_{\text{input}}(\lambda) e^{-\alpha(\lambda)L}$, larger $L$ leads to stronger absorption, and hence waveguide absorption is not a true measure of absorption linewidth (in other words, dipole dephasing time). Optical selection rules in GaAs QW suggests that for TM polarized ($z$-polarized) light only LH transition can be excited, whereas TE polarized light (can also be thought of as mixture of left- and right- circularly polarized lights) can excite both HH and LH transitions. As shown in Figure 23, for TM polarized light HH transition is negligible. This confirms the optical selection rules. For small dimensions of waveguides in our experiment (100 $\mu m$ long unless otherwise stated and 1 $\mu m$ wide), it is difficult to measure $I_{\text{input}}(\lambda)$ and $I_{\text{output}}(\lambda)$ in Beer’s law because of scattering and to get true value of $\alpha L$. Moreover, our slab waveguides are leaky. Absorption data presented in this chapter are obtained using a pulsed laser (Spectra-Physics Tsunami) and then measuring the transmission by spectrally resolving the output using a monochromator (TRIAX 320) and PMT (Hamamatsu R928). Absorption spectra using cw laser is presented in chapter VII. Even though our waveguides can support Gaussian mode peaked at QW layer, TM mode is supported better than TE mode. After propagating through waveguide (around 100 $\mu m$) at room temperature, the extinction ratio of linear TE polarization (vertical) is degraded by a factor of
around 5. Optical confinement factor ($\Gamma = 0.021$) for waveguides made of 17.5 nm SQW considering refractive index $n(GaAs) = 3.7$ and $n(Al_{0.3}Ga_{0.7}As) = 3.4$.

**FIGURE 25.** Coherent signal from LH-HH excitonic coherence at $T = 10$ K. Increasing the pump power increases the coherent signal that lasts for around 1 ps.

**Saturation and broadening in GaAs QW waveguide**

Typically in two-beam pump-probe spectroscopy one beam is weak (probe) and another beam is strong (pump or saturator). It is quite useful to know the saturation intensities for excitonic transitions. Figure 24 shows the saturation of LH transition at room temperature in a waveguide made of 17.5 nm QW. For average intensities less than $1/W/cm^2$, absorption is linear. At higher intensities increased carrier concentration leads to enhanced carrier-carrier interactions. Increased carrier concentration in the conduction leads to lower value of absorption coefficient due to saturation, whereas, increased carrier-carrier interactions lead to linewidth broadening of transition due to enhanced dephasing. As shown in Figure 24, average intensities around $1 kW/cm^2$ almost bleach the transition at room temperature. At room temperature, increased
carrier-phonon interaction provides efficient broadening of the transition. Higher intensities (more than 10 times) are required to broaden excitonic transitions at low temperature.

**Coherent Effects in GaAs QW Waveguide**

Using nonlinear optical spectroscopy, we have created and studied various types of quantum coherences in GaAs QW waveguides. Coherent optical studies are guided by the results of optical studies in normal geometry where light propagates along growth. Coherent effects such as Rabi oscillation from HH excitonic coherence, Transient coherent signal from LH-HH excitonic coherence are common to both the normal and the waveguide geometries. However, electron spin coherence without magnetic fields is created by taking advantage of spin-orbit coupling in the valence band and can be accessed only in the waveguide geometry. Detailed studies on electron spin coherence are presented in chapter 6 and chapter 7, whereas, studies on Rabi oscillations from HH excitonic coherence are presented in chapter 8. Figure 25 shows the coherent signal arising from the coherence between LH and HH excitons. Coherence in the time-domain is seen by measuring the probe transmission as the delay between the pump beam and the probe beam is varied. LH-HH coherence time is estimated to be around 1 ps.

**Propagation effects in GaAs QW waveguides**

We have studied propagation of pulse through GaAs QW waveguide. Figure 26 shows the waveguide output as a function of the input intensity. Both the incoherent saturation effect and the coherent self induced transparency (SIT) can give rise to qualitatively similar behavior. However, oscillations for lower power are more likely to be SIT. These oscillations along with our pulse propagation studies in normal geometry (chapter 10) motivate similar studies both in the waveguide and the normal geometries in future. Slow light experiments by measuring the phase change of the beat signal between two optical fields with different frequencies have been done. We have also studied beat propagation through
GaAs QW waveguides. Two mutually orthogonal polarized beams have been derived from 0th and 1st order of an acousto-optic modulator (AOM). 1st order beam is upshifted by an amount given by the frequency of the RF field applied to the AOM. 0th and 1st order beams have been made to copropagate along the two axes of a single mode polarization preserving fiber.

**FIGURE 26.** Propagation of light pulses through waveguide centered around HH resonance through GaAs QW waveguides at $T = 10$ K. Waveguide output as function of the input intensity is plotted for different spectral bandwidth of the pulses.

Copropagating beams then go through waveguide after which beams are made to beat using a polarizer. Beat frequency is given by the frequency of the RF field applied to the AOM; Changes of the phase of this beat signal are studied on and off resonance. Idea was to see the effect of narrow transparency window due to electron spin coherence on the phase of the beat. Experimental difficulties due to not being able to change RF frequency (this would change the direction of 1st order beam and hence coupling to the single mode fiber) during wavelength scan and also less than 10% transparency seems to be insufficient for making any definite conclusion.
However, an interesting nonlinear change of phase has been observed as we scan through resonance, as shown in Figure 27. Further studies need to be done to investigate this.

![Figure 27](image)

**FIGURE 27.** Propagation of optical beat through GaAs QW waveguide at T= 10 K. (a) Oscilloscope screen shots of the traces of RF reference and the beat signal. Beat frequency is given by the RF frequency. (b) Corresponding data plotted where dashed curve is RF reference and solid curve is beat signal. (c) Relative phase difference between the RF and the beat signal as the wavelength is scanned through the resonances.

**Summary**

In this chapter, we have presented the laser systems used in our experiments. Pulse shaper being an important part of our experiments has been discussed. A general schematic of pump-probe spectroscopy is given. We have also presented experimental techniques of optical spectroscopy in semiconductor waveguides. Studies on absorption, saturation, coherent effects and propagation
effects have been presented. Two coherent effects, namely, EIT from electron spin coherence and Rabi oscillations from exciton spin coherence will be discussed in next three chapters.
As mentioned in previous chapters, electron spin coherence based EIT in semiconductors is one of the approaches pursued for our slow light applications. However, electron spin coherence in semiconductors is important not only for EIT and slow light, but also has tremendous application potentials in spin-based “spintronics” devices [59]. Over the past several years intense research has been done on electron spins [58]. Robust electron spin coherence even at room temperature has been reported [63]. Electron spin coherence is usually induced by applying a magnetic field. It is both conceptually and practically important to ask if electron spin coherence can be induced without any magnetic field. This chapter analyzes electron spin coherence without magnetic field theoretically, whereas, experimental demonstration will be presented in the next chapter. We will start with general motivation of studying electron spin in the context of spintronics, then a general discussion on decoherence mechanisms will be given, and finally, we will give theoretical analysis of electron spin coherence without magnetic field.

Spintronics: Motivation for studying spins
Spin degree of freedom in solid state systems has been the topic of great interests in current years. Spin-based electronics or spintronics is an emerging area where spin degree of freedom is either added to the conventional charge-based electronics or exploited alone for better performance and novel technologies. The term spin can stand for the spin of a single electron or nucleon that can be measured by its magnetic moment, the average spin of an ensemble of electrons or nuclei that
can be measured by net magnetization, or the spin of excitons (bound electron-hole pair) in semiconductors. Experimental and theoretical challenges include the optimization of spin coherence lifetimes, the detection and generation of spin coherence in nanoscale structures, transport of spin polarized carriers across relevant length scales and heterointerfaces, and the manipulation of spins on sufficiently fast time scales, development of magnetic semiconductors exhibiting room temperature ferromagnetism. Methods for spin injection, detection and control have exploited both optical manipulation of spin-photon interactions and electrical manipulation of spin-orbit coupling. Experiments have shown long spin coherence times in semiconductors approaching microseconds, injection of spin polarized currents into semiconductors, ultrafast coherent spin manipulation, as well as phase coherent spin transport over distances up to 100 µm. It is expected that the combination of electronics, photonics and magnetics will lead to spin-based multifunctional devices such as spin-FET (field effect transistor), spin-LED (light emitting diode), spin-RTD (resonant tunneling devices), optical switches for terahertz, modulators, encoders, decoders, and qubits for quantum computation and communication. Two main motivations for studying spins arise from the quest of quantum computation and better performance of present technology.

Present semiconductor technology is based on transport and storage of charge carriers (electrons and holes). Charge-based devices such as transistors can be either in ON or OFF states (classical bit), thus implementing binary logic. In contrast, spin being quantum mechanical system, will allow a spin-based transistor to be in many different states (quantum bit or “qubit”) because of the coherent superposition of states. This creates a fundamentally new way of data processing and leads to quantum computation. As of a matter of fact, spin qubits in semiconductors (e.g. GaAs, Si) are one of most promising avenues for eventual making of a quantum computer. There are
two compelling reasons for this: 1) electron spin coherence has very long lifetime, 2) scalability due to already well established semiconductor technology.

On the other hand, current performance improvement in semiconductor industry is mainly based on reduction of component size in line with Moore’s law. This approach is already facing daunting challenges like high leakage, high power dissipation and is ultimately limited by the size of a molecule. Further improvements require a new technology. Spintronics has the promise to be one such technology.

Research interests in spins fueled by motivations such as above, of course, can lead to better understanding of physics involved and new phenomena; that, in turn, lead to novel applications. In fact, spintronic devices such as hard drive for mass storage are already in use; in 2002 IBM announced data storage capability at approximately one trillion bits per square inch (1.5 Gbit/mm²) or roughly 1 TB on a single sided 3.5” diameter disc. The storage density of hard drives is rapidly increasing along an exponential growth curve known as Kryder's Law. The doubling period for the areal density of information storage is twelve months, much shorter than Moore's Law, which observes that the number of transistors in an integrated circuit doubles every eighteen months. Spin valves based on the effect giant magnetoresistance (GMR) have already replaced anisotropic magnetoresistance sensors in computer hard disk drive heads. The operation of MRAM or magnetic random access memory is also based on spintronic principles.

Three fundamental aspects of spin are important for spintronics: spin coherence, spin transport and spin entanglement. Manipulation of spin coherence is at the heart of spin-based electronics (spintronics) devices, and spin-based “quantum bits”. Potential applications based on coherent control are facing technological difficulties due to decoherence. Decoherence or dephasing is the loss of quantum coherence in course of time due to inevitable interactions of quantum systems to their environment. In the next section, we discuss some general aspects of coherence.
Spin relaxation and spin dephasing in semiconductors

Although relaxation and dephasing in an ensemble of spins are generally too complex to be described only two parameters, the longitudinal decay time or population relaxation time $T_1$ and the transverse decay time or dephasing time $T_2$ are nevertheless quite accurate and convenient measure for quantifying decoherence processes in many cases of interests. $T_1$ processes denote energy relaxation, energy has to be transferred from the spin system to the lattice, usually by phonons. Time $T_2$ is determined by both energy relaxation and pure dephasing (elastic processes).

Linewidth of a transition (given by $1/T_2$) depends on homogeneous and inhomogeneous broadening mechanisms. Homogeneous broadening (such as collision broadening) is an irreversible process, whereas inhomogeneous broadening (such as Doppler shifts, spatial homogeneities) is reversible process (e.g. photon echo). Degree of dependence of spin dephasing on various dephasing mechanisms depends on whether the electron is mobile or localized.

Four mechanisms for spin relaxation of conduction electrons are relevant for metals and semiconductors: the Elliott-Yafet, D’yakonov-Perel’, Bir-Aronov-Pikus, and hyperfine-interaction mechanisms. In semiconductors with magnetic impurities, magnetic scattering due to an exchange interaction between conduction electrons and magnetic impurities has to be considered.

Spin-orbit coupling induced by lattice ions causes Elliott-Yafet effect. In combination with momentum scattering caused by impurities and phonons, the two spin states can couple and lead to spin relaxation. This mechanism is considered significant for small bandgap semiconductors with large spin-orbit splitting and with high impurity scattering, but is considered less significant at higher temperatures. D’yakonov-Perel’ mechanism arises in solids without a center of symmetry where spin states are not degenerate at $k \neq 0$. This $k$-dependent effective magnetic
field arising from the lack of inversion symmetry and spin-orbit interaction. This effective magnetic field changes direction randomly every time electron scatters to a different momentum state. This effect is important in most cases and dominates spin relaxation for middle bandgap semiconductors at high temperature. Bir-Aronov-Pikus is important for p-doped semiconductors, in which electron-hole exchange interaction leads to fluctuating local magnetic fields flipping electron spins. Finally, hyperfine interaction between electron spins and nuclear moments dominates spin relaxation for heterostructures based on semiconductors with a nuclear magnetic moment.

**Electron spin coherence without magnetic field**

Various nonradiative coherences including exciton spin coherence, biexciton coherence, and intervalence band coherence have been used to realize EIT in semiconductors [92-95]. These coherences are extremely fragile due to carrier-carrier and carrier-phonon interactions. At higher temperature (desirably at room temperature) carrier-phonon interactions become so dominant that it becomes very difficult, if not impossible, to use these coherences to realize EIT. Experimental studies have shown that nonradiative coherence among electron spins can be extremely robust. Electron spin coherence can persist over remarkably long time and length scales even at room temperature [63]. Therefore, destructive quantum interference induced by robust electron spin coherence is a promising way to realize EIT in semiconductors at room temperature.

Spin of an electron is a two-level system. Usually coherent electron spin dynamics is studied by applying magnetic fields. Degeneracy of the two levels, viz, spin-up and spin-down can be lifted by using a magnetic field $B$. Splitting of the two levels is given by $\Delta E = g \mu B$, where $g$ is the gyromagnetic ratio of the electron. Electron spins line up along $B$ in equilibrium. To move electron spins out of equilibrium, optical field (laser) is applied to orient them perpendicular to $B$. 
As electron spin precesses around B, their dynamics is studied by sending a weak beam and measuring, for example, time resolved Faraday rotation (TRFR).

In our study, we take advantage of the spin-orbit coupling and use spectral domain instead of time domain to detect quantum interference induced by the electron spin coherence. To induce electron spin coherence, we exploit optical polarization selection rules for the lh transition in GaAs quantum well. In this way, there is no need to apply external magnetic field or to introduce strain in the sample for creating internal magnetic field.

\[
\begin{align*}
|J, J_z\rangle &= \left|\frac{1}{2}\right\rangle \equiv |d\rangle \\
|\frac{3}{2}, \frac{3}{2}\rangle &= |c\rangle \\
|\frac{3}{2}, \frac{1}{2}\rangle &= |b\rangle \\
\end{align*}
\]

\[\begin{align*}
\sigma^+ &\quad \sigma^- \\
{z} &\quad {z} \\
\hbar \omega_e &\quad 2\hbar \Delta_e \\
\hbar \omega_e &\quad 2\hbar \Delta_{lh}
\end{align*}\]

**FIGURE 28.** Optical selection rules with magnetic field along growth. Polarization selection rules for LH transitions do not change in the presence of a magnetic field applied along growth direction. Note that magnetic field along QW plane would change the polarization selection rules.

In this chapter, detailed theoretical analysis of the proposal will be presented. Spin-orbit coupling mixes spin-up and spin-down states for LH valence band in GaAs quantum wells. As shown in Fig. 28, the two electron spin states in the conduction band can now couple to a common LH valence band state. In analogy to atomic systems, we have two V-type three-level systems. To induce coherence, we need two optical fields, one of which has to be z-polarized (z is the growth direction of the sample). The other field can be either right- \( \sigma^+ \) or left-circularly ( \( \sigma^- \) )
polarized. Light polarized along z-axis has to propagate in the plane of the QW (xy plane), which requires to do the experiment in waveguide geometry, as shown in Figure 29.

![Waveguide diagram](image)

**FIGURE 29.** Polarization configuration for electron spin coherence without magnetic field. Both pump and probe propagate in the QW waveguide along x-axis.

Waveguide under consideration is a leaky slab waveguide whose fundamental mode is assumed to be a plane wave. Lateral thickness of the waveguide is considered to be much greater than both the quantum well width and the optical wavelength. To induce electron spin coherence, we use an x-polarized (TE) control beam and z-polarized (TM) probe beam. TE polarized control beam can be thought of as a mixture of both right- and left-circularly polarized light. In this way, we access both V-systems. Both beams propagate in the QW plane along y-axis.

Coherence between two LH valence band states is neglected because of the rapid decay hole spin coherence compare to electron spin coherence. Crossed V-type system considered in our case can differ significantly from ordinary V-type systems.
The electron spin coherence, characterized by $\rho_{ad}$, leads to EIT in particular V-system by interacting, for example, with a $\sigma^+$ control beam. However, $\rho_{ad}$ can also couple to a $\sigma^-$ control beam and the resulting coherence, $\rho_{dc}$ oscillates with frequency $2\nu - \nu_1$ ($\nu$ and $\nu_1$ are the frequencies of the control and the probe beam respectively). We will refer to this second process as coherent wave mixing process.

We consider an oversimplified 4-level system analogous to atomic system. In a realistic model we need to include various interactions like carrier-carrier and carrier-phonon. Nevertheless, the model is sufficient for an excellent qualitative analysis of the proposed scheme and is able to explain the experimental results presented in the next chapter.

Consider y-polarized (TE) pump (control) beam with frequency $\nu$ propagating along x-direction. Rabi frequencies corresponding to positive and negative frequencies of the applied pump field are $+\Omega$ and $-\Omega$. Probe beam is z-polarized (TM) with frequency $\nu_1$ and Rabi frequency $\Omega_1$ and also propagating along x-direction. We will define various energy level differences as follows:

\[
\begin{align*}
\hbar \omega_{ab} &\equiv \hbar \omega_a - \hbar \omega_b = \hbar \omega_a - \hbar (\Delta_e + \Delta_{ib}) \\
\hbar \omega_{ac} &\equiv \hbar \omega_a - \hbar (\Delta_e - \Delta_{ib}) \\
\hbar \omega_{ia} &\equiv 2\hbar \Delta_e \\
\hbar \omega_{ic} &\equiv 2\hbar \Delta_{ib} \\
\hbar \omega_{ib} &\equiv \hbar \omega_b + \hbar (\Delta_e - \Delta_{ib}) \\
\hbar \omega_{ic} &\equiv \hbar \omega_b + \hbar (\Delta_e + \Delta_{ib})
\end{align*}
\]

We will treat the light-matter interaction as the perturbation. We will calculate the transitions between eigenstates of the unperturbed crystal potential due to the light-matter interaction. Eigenstates of the unperturbed potential are given by the band structure.

The Hamiltonian for the double-V system, $H = H_0 + H_{ext}$, where $H_0$ is the unperturbed Hamiltonian and $H_{ext}$ is the perturbation term for the relevant dipole interaction within the
rotating wave approximation (RWA), can be written in the form:

\[
H_0 = \hbar \omega_a |a\rangle \langle a| + \hbar \omega_b |b\rangle \langle b| + \hbar \omega_c |c\rangle \langle c| + \hbar \omega_d |d\rangle \langle d|
\]

\[
H_{\text{ext}} = -\hbar \left[ \frac{\Omega_1}{2} e^{-i\nu t} |a\rangle \langle b| + \frac{\Omega_1}{2} e^{-i\nu t} |d\rangle \langle c| + \frac{\Omega}{2} e^{-i\nu t} |a\rangle \langle c| + \frac{\Omega}{2} e^{-i\nu t} |d\rangle \langle b| + \text{h.c.} \right]
\]

Assume \( \rho_{bc} = 0 \)

\[
i \dot{\rho}_{ab} = \omega_{ab} \rho_{ab} - \frac{\Omega_1}{2} (\rho_{bb} - \rho_{aa}) e^{-i\nu t} + \frac{\Omega}{2} e^{-i\nu t} \rho_{ad} - i\gamma \rho_{ab}
\]

\[
i \dot{\rho}_{dc} = \omega_{dc} \rho_{dc} - \frac{\Omega_1}{2} (\rho_{cc} - \rho_{dd}) e^{-i\nu t} + \frac{\Omega}{2} e^{-i\nu t} \rho_{da} - i\gamma \rho_{dc}
\]

\[
i \dot{\rho}_{ad} = \omega_{ad} \rho_{ad} - \frac{\Omega_1}{2} e^{-i\nu t} \rho_{bd} + \frac{\Omega}{2} e^{-i\nu t} \rho_{ac} - \frac{\Omega}{2} e^{-i\nu t} \rho_{cd} + \frac{\Omega}{2} e^{i\nu t} \rho_{ab} - i\gamma \rho_{ad}
\]

\[
i \dot{\rho}_{db} = \omega_{db} \rho_{db} - \frac{\Omega}{2} e^{-i\nu t} (\rho_{bb} - \rho_{dd}) + \frac{\Omega}{2} e^{-i\nu t} \rho_{da} - i\gamma \rho_{db}
\]

\[
i \dot{\rho}_{dc} = \omega_{dc} \rho_{dc} - \frac{\Omega_1}{2} e^{-i\nu t} (\rho_{cc} - \rho_{dd}) + \frac{\Omega}{2} e^{-i\nu t} \rho_{da} - i\gamma \rho_{dc}
\]

\[
i \dot{\rho}_{ac} = \omega_{ac} \rho_{ac} - \frac{\Omega}{2} e^{-i\nu t} (\rho_{cc} - \rho_{aa}) + \frac{\Omega}{2} e^{-i\nu t} \rho_{ad} - i\gamma \rho_{ac}
\]

\[
i \dot{\rho}_{aa} = \left[ \left( \frac{\Omega}{2} e^{-i\nu t} \rho_{ba} - \frac{\Omega}{2} e^{-i\nu t} \rho_{ca} \right) - \text{c.c.} \right] - i\Gamma (\rho_{aa} - \rho_{aa}^{\text{equl.}})
\]

\[
i \dot{\rho}_{bb} = \left[ \frac{\Omega}{2} e^{-i\nu t} \rho_{ba} + \frac{\Omega}{2} e^{-i\nu t} \rho_{bd} \right] - \text{c.c.} \] - i\Gamma (\rho_{bb} - \rho_{bb}^{\text{equl.}})
\]

\[
i \dot{\rho}_{cc} = \left[ \frac{\Omega}{2} e^{-i\nu t} \rho_{ca} + \frac{\Omega}{2} e^{-i\nu t} \rho_{cd} \right] - \text{c.c.} \] - i\Gamma (\rho_{cc} - \rho_{cc}^{\text{equl.}})
\]

\[
i \dot{\rho}_{dd} = \left[ \left( -\frac{\Omega}{2} e^{-i\nu t} \rho_{bd} - \frac{\Omega}{2} e^{-i\nu t} \rho_{cd} \right) - \text{c.c.} \right] - i\Gamma (\rho_{dd} - \rho_{dd}^{\text{equl.}})
\]

To include the coherent wave mixing process, we write the relevant density matrix elements, up to the first order in the probe field, as follows:
\[ \rho_{ab} = A e^{-i\nu_1 t} + B e^{-(2\nu_1 - \nu_2) t} + O(\Omega_1^3) \]
\[ \rho_{ad} = X e^{i(\nu_1 - \nu_2) t} + Y e^{-(2\nu_1 - \nu_2) t} + O(\Omega_1^3) \]
\[ \rho_{de} = A e^{-i\nu_1 t} + B e^{-(2\nu_1 - \nu_2) t} + O(\Omega_1^3) \]
\[ \rho_{db} = C e^{-i\nu_1 t} + O(\Omega_1^2) \]
\[ \rho_{ac} = C e^{-i\nu_1 t} + O(\Omega_1^2) \]

where the complex amplitudes are slowly varying functions of time.

The coherent wave mixing process can also lead to population pulsation. To the second order of the probe field, populations, as represented by the diagonal matrix elements, can oscillate with a beat frequency of \( \pm 2(\nu - \nu_1) \). In contrast to the conventional population pulsation process, population pulsation in the double-V system arises from electron spin coherence and does not occur in the first order of the probe field. Effects of population pulsation thus do not contribute up to the first order of the probe field. Consider

\[ \rho_{aa} + \rho_{bb} + \rho_{cc} + \rho_{dd} = 2, \rho_{aa}(0) = \rho_{bb}(0) = \rho_{cc}(0) = \rho_{dd}(0) = 0, \rho_{ab}(0) = \rho_{ac}(0) = \rho_{bd}(0) = \rho_{cd}(0) = 1 \]

Since up to the first order of the probe field, \( \rho_{ab} = \rho_{cd} = O(\Omega_1) \),

\[ i\dot{\rho}_{aa}^{(0)} = \left[ -\frac{\Omega}{2} e^{-i\nu_1 t} \rho_{ca}^{(0)} - \text{c.c.} \right] - i\Gamma \rho_{aa}^{(0)} \]
\[ i\dot{\rho}_{bb}^{(0)} = \left[ \frac{\Omega}{2} e^{i\nu_1 t} \rho_{bd}^{(0)} - \text{c.c.} \right] + i\Gamma \left( 1 - \rho_{bb}^{(0)} \right) \]
\[ i\dot{\rho}_{cc}^{(0)} = \left[ -\frac{\Omega}{2} e^{-i\nu_1 t} \rho_{ca}^{(0)} - \text{c.c.} \right] + i\Gamma \left( 1 - \rho_{cc}^{(0)} \right) \]
\[ i\dot{\rho}_{dd}^{(0)} = \left[ -\frac{\Omega}{2} e^{-i\nu_1 t} \rho_{bd}^{(0)} - \text{c.c.} \right] - i\Gamma \rho_{dd}^{(0)} \]

Let \( \delta n_{ac} = \rho_{aa}^{(0)} + \rho_{aa}^{(0)} - 1 \); then \( \frac{d}{dt}(\delta n_{ac}) = -\Gamma \delta n_{ac} \); which in turn give \( \delta n_{ac} = \delta n_{ac}(0) e^{-\Gamma t} \).

Since \( \delta n_{ac}(0) = 0 \), we have \( \rho_{aa}^{(0)} + \rho_{cc}^{(0)} = 1 \) and \( \rho_{bb}^{(0)} + \rho_{dd}^{(0)} = 1 \). The equations of motion for the
relevant populations and complex amplitudes, up to the first order of the probe field but to all orders of the pump field, are given by:

\[
i\dot{A}^{(i)} = \left[ \Delta_i - (\Delta_e + \Delta_{th}) - i\gamma \right] A^{(i)} - \frac{\Omega_i}{2} \left( \rho_{bb}^{(0)} - \rho_{aa}^{(0)} \right) + \frac{\Omega}{2} X^{(i)}
\]

\[
i\dot{A}^{(i)} = \left[ \Delta_i + (\Delta_e + \Delta_{th}) - i\gamma \right] A^{(i)} - \frac{\Omega_i}{2} \left( \rho_{cc}^{(0)} - \rho_{dd}^{(0)} \right) + \frac{\Omega}{2} Y^{(i)\ast}
\]

\[
i\dot{B}^{(i)} = - \left[ \Delta_i - 2\Delta + (\Delta_e + \Delta_{th}) + i\gamma \right] B^{(i)} + \frac{\Omega}{2} Y^{(i)}
\]

\[
i\dot{B}^{(i)} = - \left[ \Delta_i - 2\Delta - (\Delta_e + \Delta_{th}) + i\gamma \right] B^{(i)} + \frac{\Omega}{2} X^{(i)\ast}
\]

\[
i\dot{X}^{(i)} = - \left[ \Delta_i - \Delta - 2\Delta_e - i\gamma_{ad} \right] X^{(i)} - \frac{\Omega_i}{2} C^{(0)\ast} - \frac{\Omega}{2} B^{(i)\ast} + \frac{\Omega}{2} A^{(i)}
\]

\[
i\dot{Y}^{(i)} = - \left[ \Delta_i - \Delta + 2\Delta_e + i\gamma_{ad} \right] Y^{(i)} + \frac{\Omega}{2} C^{(0)} - \frac{\Omega_i}{2} A^{(i)\ast} + \frac{\Omega}{2} B^{(i)}
\]

\[
i\dot{C}^{(0)} = \left[ (\Delta + \Delta_e - \Delta_{th}) - i\gamma \right] C^{(0)} - \frac{\Omega_i}{2} \left( \rho_{bb}^{(0)} - \rho_{aa}^{(0)} \right) + \frac{\Omega}{2} X^{(i)\ast}
\]

\[
i\dot{C}^{(0)*} = \left[ -(\Delta + \Delta_e - \Delta_{th}) - i\gamma \right] C^{(0)*} - \frac{\Omega_i}{2} \left( \rho_{cc}^{(0)} - \rho_{dd}^{(0)} \right) + \frac{\Omega}{2} Y^{(i)}
\]

\[
i\dot{\rho}_{ab}^{(0)} = \left[ - \frac{\Omega}{2} C^{(0)\ast} + \frac{\Omega^*}{2} C^{(0)} \right] - i\Gamma \rho_{aa}^{(0)}
\]

\[
i\dot{\rho}_{bb}^{(0)} = \left[ \frac{\Omega}{2} C^{(0)\ast} - \frac{\Omega^*}{2} C^{(0)} \right] + i\Gamma \rho_{dd}^{(0)}
\]

\[
i\dot{\rho}_{cc}^{(0)} = \left[ \frac{\Omega}{2} C^{(0)\ast} - \frac{\Omega^*}{2} C^{(0)} \right] + i\Gamma \rho_{aa}^{(0)}
\]

\[
i\dot{\rho}_{dd}^{(0)} = \left[ - \frac{\Omega}{2} C^{(0)\ast} + \frac{\Omega^*}{2} C^{(0)} \right] - i\Gamma \rho_{dd}^{(0)}
\]

With continuous-wave pump and probe fields, we need to find the steady-state solution for the above equations of motion:
\[ O(\Omega_1^{(i)}) : \]

\[
\rho_{dd}^{(0)} = \frac{|\Omega_+|^2}{2 \left[ |\Omega_+|^2 + \Gamma \gamma \left\{ 1 + \left( \frac{\Delta + \Delta_e - \Delta_{lh}}{\gamma} \right)^2 \right\} \right]} \\
\rho_{aa}^{(0)} = \frac{|\Omega_-|^2}{2 \left[ |\Omega_-|^2 + \Gamma \gamma \left\{ 1 + \left( \frac{\Delta - \Delta_e + \Delta_{lh}}{\gamma} \right)^2 \right\} \right]} \\
C^{(0)} = \frac{\Omega_+}{2(\Delta + \Delta_e - \Delta_{lh} - i\gamma)} \left( 1 - 2 \rho_{dd}^{(0)} \right) \\
= \frac{\Omega_+}{2(\Delta + \Delta_e - \Delta_{lh} - i\gamma)} \times \frac{1}{1 + \frac{|\Omega_+|^2}{\Gamma \gamma \left\{ 1 + \left( \frac{\Delta + \Delta_e - \Delta_{lh}}{\gamma} \right)^2 \right\}}} \\
C^{(0)} = \frac{\Omega_-}{2(\Delta - \Delta_e + \Delta_{lh} - i\gamma)} \left( 1 - 2 \rho_{aa}^{(0)} \right) \\
= \frac{\Omega_-}{2(\Delta - \Delta_e + \Delta_{lh} - i\gamma)} \times \frac{1}{1 + \frac{|\Omega_-|^2}{\Gamma \gamma \left\{ 1 + \left( \frac{\Delta - \Delta_e + \Delta_{lh}}{\gamma} \right)^2 \right\}}} \tag{1}
\]

\[
O(\Omega_1^{(i)}) : \]

\[
\left[ \Delta - (\Delta_e + \Delta_{lh}) - i\gamma \right] A^{(i)} = \frac{\Omega_1}{2} \left( \rho_{bb}^{(0)} - \rho_{aa}^{(0)} \right) - \frac{\Omega_+}{2} X^{(i)} \tag{2}
\]

\[
\left[ \Delta - 2\Delta - (\Delta_e + \Delta_{lh}) + i\gamma \right] B^{(i)} = \frac{\Omega_1}{2} X^{(i)*} \tag{3}
\]

\[
\left[ \Delta - \Delta_e - i\gamma_{ad} \right] X^{(i)} = \frac{\Omega_1}{2} B^{(i)*} - \frac{\Omega_+}{2} A^{(i)} + \frac{\Omega_1}{2} C^{(0)*} \tag{4}
\]

Equations 2 and 3 give:
\[
\left[ \Delta_1 - \Delta - 2\Delta_e - iy_{ad} - \frac{\Omega_e}{4} \left( \frac{1}{\Delta_1 - 2\Delta - (\Delta_e + \Delta_m) - iy} \right) \right] X^{(1)} = -\frac{\Omega_e}{2} A^{(1)} + \frac{\Omega_e}{2} C^{(0)*}
\] (5)

Equations 1 and 4 give:

\[
\begin{align*}
D &= \left[ \Delta_1 - \Delta - 2\Delta_e - iy_{ad} - \frac{\Omega_e}{4} \left( \frac{1}{\Delta_1 - 2\Delta - (\Delta_e + \Delta_m) - iy} \right) \right] \\
A^{(1)} &= -\frac{\Omega_e}{4D} \frac{\Omega_i}{4D} C^{(0)*} + \frac{\Omega_i}{2} \left( \rho_{bb}^{(0)} - \rho_{aa}^{(0)} \right)
\end{align*}
\] (6)

If pump beam is on resonance, i.e., \( \Delta = 0, \left| \Omega_e \right| = \left| \Omega_i \right| \). This implies \( \rho_{aa}^{(0)} = \rho_{dd}^{(0)}, \rho_{bb}^{(0)} = \rho_{cc}^{(0)} \). Then we have:

\[
I = \frac{\left| \Omega_e \right|^2}{\Gamma \gamma \left( 1 + \left( \frac{\Delta_e - \Delta_m}{\gamma} \right)^2 \right)}
\] (7)

Equations 1, 6 and 7 give:

\[
A^{(1)} = \frac{\Omega_i}{2(1+I) \times \Delta_1 - (\Delta_e + \Delta_m) - iy} \times \frac{1}{\Delta_1 - (\Delta_e + \Delta_m) - iy} \times \left[ \frac{\Omega_i}{4 \Delta_1 - (\Delta_e + \Delta_m) - iy} \right]
\] (8)

Similarly,

\[
A^{(2)} = \frac{\Omega_i}{2(1+I) \times \Delta_1 - (\Delta_e + \Delta_m) - iy} \times \frac{1}{\Delta_1 - (\Delta_e + \Delta_m) - iy} \times \left[ \frac{\Omega_i}{4 \Delta_1 - (\Delta_e + \Delta_m) - iy} \right]
\] (9)
When both pump and probe are TM polarized, we have two independent two level systems. The Hamiltonian in this case, $H = H_0 + H_{\text{ext}}$, where $H_0$ is the unperturbed Hamiltonian and $H_{\text{ext}}$ is the perturbation term for the relevant dipole interaction within the rotating wave approximation, can be written in the form:

$$H_0 = \hbar \omega_a |a\rangle\langle a| + \hbar \omega_b |b\rangle\langle b| + \hbar \omega_c |c\rangle\langle c| + \hbar \omega_d |d\rangle\langle d|$$

$$H_{\text{ext}} = -\hbar \left[ \frac{\Omega_1}{2} e^{-i\nu t} (|a\rangle\langle b| + |d\rangle\langle c|) + \frac{\Omega_p}{2} e^{-i\nu t} (|a\rangle\langle b| + |d\rangle\langle c|) + h.c. \right]$$

The equations of motion for relevant density matrix elements are:

$$i \dot{\rho}_{ab} = \omega_{ab} \rho_{ab} - \left[ \frac{\Omega_1}{2} e^{-i\nu t} + \frac{\Omega_p}{2} e^{-i\nu t} \right] (\rho_{bb} - \rho_{aa}) - i\eta \rho_{ab}$$

$$i \dot{\rho}_{dc} = \omega_{dc} \rho_{dc} - \left[ \frac{\Omega_1}{2} e^{-i\nu t} + \frac{\Omega_p}{2} e^{-i\nu t} \right] (\rho_{cc} - \rho_{dd}) - i\eta \rho_{dc}$$

$$i \dot{\rho}_{aa} = \left[ \frac{\Omega_1}{2} e^{-i\nu t} + \frac{\Omega_p}{2} e^{-i\nu t} \right] \rho_{ba} - c.c. \right] - i\Gamma (\rho_{aa} - \rho_{aa}^{eq})$$

$$i \dot{\rho}_{bb} = \left[ \frac{\Omega_1}{2} e^{-i\nu t} + \frac{\Omega_p}{2} e^{-i\nu t} \right] \rho_{ba} - c.c. \right] - i\Gamma (\rho_{bb} - \rho_{bb}^{eq})$$

$$i \dot{\rho}_{cc} = \left[ \frac{\Omega_1}{2} e^{-i\nu t} + \frac{\Omega_p}{2} e^{-i\nu t} \right] \rho_{ba} - c.c. \right] - i\Gamma (\rho_{cc} - \rho_{cc}^{eq})$$
To include the coherent wave mixing process, we write, as in the (TE, TM) case, the relevant density matrix elements, up to the first order of the probe field, as follows:

\[
\rho_{ab} = Ae^{-i\nu t} + Be^{-i\nu t} + Xe^{-i(2\nu - \nu_1)t} + Ye^{-i(\nu_1 - 2\nu)t} + O(\Omega_i^2) \\
\rho_{dc} = A'e^{-i\nu t} + B'e^{-i\nu t} + X'e^{-i(2\nu - \nu_1)t} + Y'e^{-i(\nu_1 - 2\nu)t} + O(\Omega_i^2) \\
\rho_{aa} = C(De^{i(\nu - \nu_1)t} + c.c.); \quad \rho_{bb} = 1 - \rho_{aa} \\
\rho_{cc} = 1 - \rho_{dd} \\
\rho_{dd} = C'\left(D'e^{i(\nu - \nu_1)t} + c.c.\right)
\]

Within RWA $Y = Y' = 0$. Consider $\rho_{aa}(0) = \rho_{aa}^{eq} = 0; \rho_{bb}(0) = \rho_{bb}^{eq} = 1$. The equations of motion for the relevant complex amplitudes, up to the first order of the probe field but to all orders of the pump field, are given by:

\[
i\dot{A}^{(1)} = \left[\Delta_t - (\Delta_e + \Delta_{th}) - i\nu\right]A^{(1)} - \frac{\Omega_1}{2}\left(1 - 2C^{(0)}\right) + \frac{\Omega_p}{2}2D^{(1)}
\]

\[
i\dot{C}(0) = -i\GammaC(0) - \frac{\Omega_1}{2}A(0)^* + \frac{\Omega_1^*}{2}A(1) - \frac{\Omega_p}{2}B(0)^* + \frac{\Omega_p^*}{2}B(0)
\]

\[
i\dot{D}^{(1)} = (\Delta_t - \Delta - i\Gamma)D^{(1)} - \frac{\Omega_1}{2}B(0)^* + \frac{\Omega_p}{2}A(1) - \frac{\Omega_p^*}{2}X(1)^*
\]

\[
i\dot{B}^{(0)} = \left[\Delta - (\Delta_e + \Delta_{th}) - i\nu\right]B^{(0)} - \frac{\Omega_p}{2}\left(1 - 2C^{(0)}\right) + \frac{\Omega_1}{2}2D^{(1)^*}
\]

\[
i\dot{X}^{(1)} = \left[2\Delta - \Delta_t - (\Delta_e + \Delta_{th}) - i\nu\right]X^{(1)} + \frac{\Omega_p}{2}2D^{(1)^*}
\]

With continuous-wave pump and probe fields, we obtained the steady-state solution for $A^{(1)}$: 
To describe the propagation of the probe in the double-V system, we define two susceptibilities:

\[
A^{(i)} = \frac{\Omega_i}{2(1 + I_+)} \times \frac{1}{\Delta_i - (\Delta_e + \Delta_{lh}) - i\gamma} \times 1 - \frac{\left|\Omega_p\right|^2}{\Delta_i - i\Gamma} \left\{ \frac{1}{\Delta - \Delta_e - \Delta_{lh} - i\gamma} - \frac{1}{2} \left( \frac{1}{\Delta - 2\Delta_e + \Delta_{lh} - i\gamma} + \frac{1}{\Delta - (\Delta_e + \Delta_{lh}) - i\gamma} \right) \right\}
\]

\[
I_+ = \frac{\left|\Omega_p\right|^2}{\Gamma\gamma} \left\{ \frac{\left|\Delta - \Delta_e - \Delta_{lh}\right|^2}{\gamma} \right\}
\]

(17)

To describe the propagation of the probe in the double-V system, we define two susceptibilities:

\[
\chi_N(v_1) = \frac{A^{(i)}(v_1) + A^{(i)}(v_1)}{h\Omega_i(v_1)} N\mu^2
\]

\[
\overline{\chi}(v_1) = \frac{B^{(i)}(v_1) + B^{(i)}(v_1)}{h\Omega_i^+(v_1)} N\mu^2
\]

where \(N\) is an effective density averaged over the cross-section of the probe beam and \(\mu\) is the dipole matrix element for the \(z\)-component of the lh transition. \(\chi_N(v_1)\) is the susceptibility that contains contributions from absorption saturation and the usual EIT process. \(\overline{\chi}(v_1)\) is the susceptibility associated with the coherent wave mixing process.

Note that in waveguide scheme the pump beam becomes strongly attenuated as it propagates along the QW waveguide. To avoid strong attenuation of the pump, one can use a pump beam that is still \(x\)-polarized but propagates along the \(z\)-axis (orthogonal to the probe) instead of the \(y\)-axis. This orthogonal configuration, however, can also lead to a peculiar behavior: the optical field induced by the coherent wave mixing process can propagate along \(-k_i\) (counter-
propagating with respect to the probe). This is due to the fact that only the in-plane component of the wave vector needs to be conserved for phase matching since the motion of the carriers are confined to the plane of the QW. Figure 30 shows numerical results with parameters given by the experiment: $\gamma/2\pi = 0.16$ THz, $\gamma_s/2\pi = \Gamma/2\pi = 1$ GHz, $\Delta_e/2\pi = 0.75$ GHz, $\delta_p/2\pi = 48$ GHz. For convenience, we also choose $\Delta_{\eta} = 0$.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig30.png}
\caption{Calculated differential transmission (DT) response. (a) Pump is TE polarized and probe is TM polarized. (b) Both pump and probe are TE polarized. See text for parameters used.}
\end{figure}

**Summary**

In this chapter, we have given general motivation for studying electron spin coherence and decoherence in semiconductors. Theoretical analysis of electron spin coherence without magnetic field has been in detail. In the next chapter, we will present experimental verification [60] of our theoretical analysis.
CHAPTER VI

ELECTRON SPIN COHERENCE WITHOUT MAGNETIC FIELD: EXPERIMENT

In this chapter, we demonstrate the generation and detection of electron spin coherence in a quantum well waveguide without either an external or internal DC magnetic field. In the absence of spin precession, the induced spin coherence is detected through effects of quantum interference in the spectral domain coherent nonlinear optical response. We interpret the experimental results qualitatively using a few-level model with optical selection rules as its basic ingredient.

Introduction

Coherent spin manipulation in semiconductors plays a central role in spin-based electronics and photonics and in spin-based quantum information technologies [59]. Extensive experimental studies in this area have led to the demonstration of remarkable phenomena such as robust electron spin coherence at room temperature and the transfer of electron spin coherence between molecularly bridged quantum dots [60-61]. Coherent spin manipulation can be carried out in the presence of an external DC magnetic field in Voigt geometry. The external magnetic field induces a Zeeman splitting between the two conduction band electron spin states. Coherent superposition of these spin states leads to a Larmor precession of the electron spin, which can be probed with transient optical techniques. Coherent spin manipulation without external magnetic fields in non-magnetic semiconductors has also been demonstrated in a recent study [62], in which an effective internal magnetic field induced by strain results in a spin splitting in the conduction band. An interesting question, which is of both conceptual and practical importance,
is whether coherent spin manipulation in semiconductors can be carried out without either external or internal magnetic fields, i.e., without spin precession.

**FIGURE 31.** Transmission spectra of a GaAs QW waveguide. (a) Optical selection rule for dipole transitions between the conduction and the HH and LH valence bands in a GaAs QW. (b) and (c) Linear transmission spectra of the QW waveguide for TE and TM polarized fields.

**Electron spin coherence without magnetic field**

We demonstrate the generation and detection of electron spin coherence in a GaAs quantum well (QW) without the use of either an external or internal magnetic field, enabling a new approach of accessing spin coherence in semiconductors. We have taken advantage of the spin-orbit coupling in the valence band and have used light-hole (LH) transitions in a QW waveguide to induce coherent superposition of the electron spin states. In the absence of spin precession, the induced
spin coherence is detected through quantum interference in the spectral domain, instead of time domain, coherent nonlinear optical response.

![Graph](image)

**FIGURE 32.** Experimental DT response. DT response obtained with $I_{\text{pump}} = 2 \text{ mW}$ and $I_{\text{probe}} = 0.25 \text{ mW}$. The pump and probe are TE and TM polarized, respectively. The solid line is a fit to Lorentzian.

The band edge in semiconductors such as GaAs is characterized by $s$-like conduction bands and $p$-like valence bands. Spin-orbit coupling in the valence band leads to the formation of heavy-hole (HH) and LH valence bands with $j_z = \pm 3/2$ and $\pm 1/2$, respectively. Figure 31(a) shows the selection rule for dipole transitions between the conduction and the HH and LH valence bands in a GaAs QW. For the LH valence bands, the spin-orbit coupling mixes spin-up and spin-down states, which makes it possible to couple the $s_z = \pm 1/2$ conduction bands to a common LH valence band via two dipole optical transitions. Exciting both of these two transitions can in principle induce a coherent superposition of the two electron spin states. As indicated in Figure 31(a), one of the transitions is for optical fields polarized along the $z$-direction [63]. For a (001) QW, the $z$-
polarized field has to propagate in the plane of the QW. The optical excitation thus has to be carried out in a waveguide configuration.

![Graph](image)

**FIGURE 33.** Percentage change of the induce transparency. DT response obtained with $I_{\text{pump}} = 4 \text{ mW}$ and $I_{\text{probe}} = 0.25 \text{ mW}$. The pump and probe are TE and TM polarized, respectively.

In all previous studies, optical excitations of electron spin coherence through either HH or LH transitions were carried out in a configuration of normal or near normal incidence. In this configuration, the spin coherence can be excited only in the presence of an external or an effective internal DC magnetic field in the plane of the QW (Voigt geometry). As discussed earlier, electron spin coherence in this case leads to spin precession, which can be detected with techniques such as time-resolved Faraday rotation, time-resolved photoluminescence, or transient differential absorption [64-66].

In the absence of external or internal DC magnetic fields, no spin precession is present. Transient techniques are no longer effective for probing the electron spin coherence. The spin coherence,
however, can still lead to quantum interference, inducing a transmission resonance in the spectral
domain coherent optical response, as we will discuss in detail later. The linewidth of the induced
resonance corresponds to the decay rate of the electron spin coherence. A particular challenge is
to identify and single out, from the complex nonlinear optical response, the contribution induced
by the electron spin coherence.

**FIGURE 34.** Power dependence of the induced transparency at $T = 20$ K. DT response obtained with pump
wavelength at 805.6 nm. The pump and probe are TE and TM polarized, respectively.

Experimental studies were carried out in a slab waveguide consisting of a 17.5 nm GaAs/AlGaAs
(001) QW and GaAs/AlGaAs superlattice cladding layers grown by molecular beam epitaxy. The
waveguide was cleaved to a length of nearly 100 $\mu$m and was mounted on a bridge-like sample
holder attached to the cold finger of an optical cryostat. A 20X optical objective was used to
couple the incident laser beams into the waveguide. Figures 31(b) and 31(c) show the linear
transmission spectra of the waveguide for TE (electric field parallel to the QW plane) and TM
(electric field normal to the QW plane) polarized fields, respectively. For the TE polarization
both HH and LH exciton resonances are observed, whereas for the TM polarization HH exciton
resonance is nearly negligible, indicating a nearly ideal polarization selection rule [63]. Fabry-
Perot fringes due to reflection between the two facets of the waveguide are also visible in the
transmission spectra. Note that strong exciton absorption ($\alpha L > 10$) leads to transmission resonances with a nearly flat bottom. To perform differential transmission (DT) measurements, we used a tunable diode laser and a tunable Ti: Sapphire laser as the probe and pump, respectively. The relative frequency jitter between the pump and probe is less than 20 MHz. Both beams propagate along the same direction in the waveguide. To avoid strong optical absorption in the waveguide, the pump was fixed at a spectral position 0.5 to 1 nm below the LH exciton absorption line center. To single out the DT response, we used lock-in detection with dual modulation. The intensity of the pump and probe were modulated at frequencies $\omega_1$ and $\omega_2$, respectively. The DT response, with an intensity modulation frequency of $|\omega_2 - \omega_1|$, was measured with a lock-in amplifier.

**FIGURE 35.** Effect of free carrier injection on electron spin coherence. Differential transmission signal is almost quenched by 5 mW of HeNe excitation. The pump and probe are TE and TM polarized, respectively.

The basic idea of the experiment can be understood with the help of Figure 31(a). For the excitation of the electron spin coherence, the TE-polarized pump and TM-polarized probe couple
the two electron spin states to a common LH valence band state (TE-polarized fields consist of both $\sigma^+$ and $\sigma^-$ polarized fields in Fig. 31(a). The spin coherence is formed to the second order of the applied field (linear to both the pump and probe field).

The pump further interacts with the induced spin coherence, generating a third order polarization. This spin-coherence-induced nonlinearity can be detected in the DT response, i.e. the change in the probe transmission induced by the pump. Specifically, we will show below that the DT response as a function of the pump-probe detuning exhibits an induced resonance associated with the electron spin coherence.

![Image of Figure 36](image)

**FIGURE 36.** Wavelength dependence of the induced transparency at $T = 20\,\text{K}$. DT response obtained with $I_{\text{pump}} = 10\,\text{mW}$ and $I_{\text{probe}} = 0.25\,\text{mW}$. The pump and probe are TE and TM polarized, respectively. Coherent resonance is seen for wavelength window of width around 1.6 nm.

Figure 32 shows the DT response obtained at $T=50\,\text{K}$. A sharp induced resonance occurs at zero pump-probe detuning. The spectral linewidth of the resonance is 1 GHz, corresponding to a
decay time of 300 ps, which is in agreement with an earlier transient measurement of the spin decoherence time in a similar 17.5 nm GaAs QW [67]. Change of transmission is few percent as shown in Figure 33. Power variation of induce transparency is shown in Figure 34. For the pump and probe powers used in our experiment transmission amplitude show nearly linear behavior as a function of intensity. Free carrier injection above the bandgap by 5 mW of HeNe laser excitation lead to carrier induced dipole dephasing or broadening of the excitonic resonance, as a result differential transmission signal is almost quenched as shown in Figure 35(a). Nevertheless, free carrier injection should not affect electron spin coherence as much as dipole coherence. As a result, induced transparency signal due to electron spin coherence is not quenched and still shows a linear behavior as function of probe power, as shown in Figure 35(b). Coherent resonance is seen only for around 1.6 nm wide wavelength window, as shown in the wavelength dependence in Figure 36.

To show that the induced resonance in Figure 32 indeed arises from the electron spin coherence, we carried out additional experimental studies, in which we applied to the QW waveguide an external magnetic field \((B=0.25 \text{ T})\) along the \(z\)-axis. In this Faraday geometry, the external magnetic field induces energy splitting in both the conduction and the valence bands, but does not affect the optical selection rule. The electron spin coherence is thus excited through the same mechanism regardless whether there is an external magnetic field (this would not be the case if the external magnetic field were applied in the Voigt geometry). We stress that while the use of the external magnetic field enables us to demonstrate and clarify the physical origin of the induced resonance in the DT response, the excitation and detection of the electron spin coherence do not rely on the presence of the external magnetic field.

For comparison, we show in Figure 37(c) the DT response obtained with the pump and probe having the same TE-polarization and with otherwise identical conditions to Figure 37(b) (similar
results were also obtained when both the pump and probe are TM-polarized). As expected, the spin-coherence induced resonances vanish since in this case the pump and probe cannot couple the two electron spin states to a common LH valence band. Instead, a sharp resonance is observed at zero pump-probe detuning. This induced resonance is due to exciton population oscillation and the width of the resonance is determined by the exciton lifetime [69].

**FIGURE 37.** Magnetic field dependence of the induced transparency. DT response obtained with $I_{\text{pump}} = 2$ mW and $I_{\text{probe}} = 0.25$ mW and with an external magnetic field ($B=0.25$ T) applied in the Faraday geometry. For (a) and (b), the pump and probe are TE and TM polarized, respectively. For (c), both the pump and probe are TE polarized.

Note that the optical selection rule in Figure 31(a) dictates that population oscillation cannot occur when the pump and probe are TE and TM polarized, respectively, which is shown in the
following theoretical analysis and is also confirmed experimentally by the absence of an induced resonance at zero pump-probe detuning in Figures 37(a) and 37(b).

Figures 38 and 39 show temperature dependence of the induced transparency with magnetic field and without magnetic field. Coherent signal can be seen at temperatures as high as 80 K, which is above liquid nitrogen temperature 77 K. Hence our experiments can be done even with liquid nitrogen as cryogen, and thus will be both economical and experimentally convenient. As can be seen from Figure 38, both the splitting and linewidth of the two resonances is higher for TM pump and TE probe than those for TE pump and TM probe. Difference in the two cases is the powers of TE (4 mW for the first case and 0.25 mW for the second case) and TM (0.25 mW for the first case and 4 mW for the second case) polarizations. Given that TE and TM modes are not supported in our slab waveguides, differences in powers of TE and TM polarizations might be one of the possible reasons of the differences of splitting and linewidth. Also note that at T = 50 K coherent resonances become sharper and then again wider at higher temperatures. This agrees with previous experimental observations in other laboratories.

To gain a qualitative understanding of electron spin coherence and related coherent nonlinear optical processes in the spectral domain, we used a simple few-level model for the QW system. This model consists of the two conduction band states and the two LH valence band states in Figure 31(a). The model Hamiltonian assigns energy to each of the four states and optical transitions between the states as shown in Figure 31(a). A weak magnetic field in the Faraday geometry, when switched on, leads to a Zeeman-splitting of the conduction band denoted by $2 \hbar \Delta_e$. The splitting in the LH band is denoted by $2 \hbar \Delta_{lh}$. We solve the density-matrix equations of motion under this Hamiltonian at steady state, with the two valence band states initially filled and the two conduction band states initially empty.
FIGURE 38. Temperature dependence of the induce transparency. DT response obtained with $I_{\text{pump}} = 4 \text{ mW}$ and $I_{\text{probe}} = 0.25 \text{ mW}$ and with an external magnetic field ($B=0.25 \text{ T}$) applied in the Faraday geometry.

The DT response to the second order of the pump field and first order of the probe field (see e.g. [70]) and with the pump TM-polarized and the probe TE-polarized is given by

$$\Delta T^{(3)} \propto -\frac{|\Omega_p|^2 |\Omega_r|^2}{8} \text{Im}(A_+ + A_-)$$

$$A_\pm = \frac{1}{\delta_\pm + (\Delta_e + \Delta_a) + i\gamma} \left[ \frac{1}{\delta_p - \delta_r \mp 2\Delta_e + i\gamma} \left( \frac{1}{\delta_r \pm (\Delta_e - \Delta_a) - i\gamma} - \frac{1}{\delta_r \mp (\Delta_e + \Delta_a) + i\gamma} \right) \right] + \frac{4\gamma}{\Gamma} I_\pm$$

Here $\delta_p$ ($\delta_r$) is the detuning of the pump (probe) frequency from the LH transition frequency at
zero magnetic field, $\gamma$ is the optical transition linewidth, $\gamma_s$ is the decay rate of the electron spin coherence, $1/\Gamma$ is the lifetime of the conduction band levels, and $I_\pm = 1/(\gamma^2 + [\delta_p (\Delta_e - \Delta_{th})])^2$. The Rabi frequencies for both optical fields are defined with the dipole moment for circular polarizations: $|\hbar \Omega_{p/l} | = |d_\perp| |\tilde{E}_{p/l}|$.

![Figure 39](image)

**FIGURE 39.** Polarization dependence of the induced transparency for different configurations of pump and probe polarizations without magnetic field. DT response obtained with $I_{\text{pump}} = 4\, \text{mW}$ and $I_{\text{probe}} = 0.25\, \text{mW}$.

There are two types of contributions to the DT response (the contribution of the short-lived coherence between the two LH states is ignored in Eq. 1). The first type is represented by the last term in $A_\pm$ and is due to incoherent bleaching. This contribution leads to a spectrally broad response. The second type, represented by the first term in $A_\pm$, is due to the electron spin coherence. With the pump TM-polarized and the probe TE-polarized, our model is a double-V system with the two electron spin states as the two excited states in each V-type 3-level system.
The optical transition path that excites the spin coherence interferes destructively with the direct (first order) probe transition in each V-system. Similar to electromagnetically induced transparency, this destructive interference results in an induced transmission resonance in the DT response [71]. As shown in Eq. 1, the spectral position of the induced resonance is determined by the two-photon resonance condition $\delta_p - \delta_r - 2\Delta_z = 0$. The linewidth of the resonance is determined by the decay rate of the spin coherence. In the absence of an external magnetic field, the induced resonance occurs at zero pump-probe detuning.

When both the pump and probe are TE-polarized, our model becomes that of two decoupled two-level systems. The DT response is then given by

$$\Delta T^{(3)} \propto -\frac{[\Omega_r]^2 [\Omega_p]^2}{16} \text{Im}(B_+ + B_-),$$

$$B_\pm = \frac{1}{\delta_i + (\Delta_z - \Delta_i) \pm i\gamma} \left[ \frac{1}{\delta_i - \delta_p \pm i\Gamma} \left( \frac{1}{\delta_p \mp (\Delta_z - \Delta_{ih}) \pm i\gamma} \right) - \frac{1}{\delta_i \mp (\Delta_z - \Delta_{ih}) \pm i\gamma} \right] \frac{2\gamma I_z}{\Gamma}$$

In this configuration, $\Delta T^{(3)}$ consists of a spectrally broad response due to incoherent bleaching and also a sharp resonance at zero pump-probe detuning. No spin coherence can be created in this case; instead, the sharp resonance is set up by the two optical fields coupling to the same transition: it is the result of a coherent population oscillation [70]. As such, the width of the resonance is determined by the excited state lifetime $1/\Gamma$. Note that the sharp resonance occurs at zero pump-probe detuning even in the presence of an external magnetic field.

Figure 40 shows numerical results from Eqs. 1 and 2 with parameters given by the experiment: $\gamma/2\pi = 0.16$ THz, $\gamma_s/2\pi = \Gamma/2\pi = 1$ GHz, $\Delta_z/2\pi = 0.75$ GHz, $\delta_p/2\pi = 48$ GHz. For convenience, we also choose $\Delta_{ih} = 0$. This simple model describes qualitatively the principal features of the experiment, confirming that the sharp resonance in the DT response for TE pump and TM probe
is induced by an electron spin resonance. While the above few-level model yields good qualitative agreement with the experiment, nonlinear optical processes in semiconductors are also strongly affected by inherent many-body Coulomb interactions. A quantitative analysis should be possible with fully microscopic theories, for example along the lines of Ref. [72-79].

**FIGURE 40.** Calculated differential transmission response. (a) Pump is TE polarized and probe is TM polarized. (b) Both pump and probe are TE polarized. See text for parameters used.

**Summary**

In summary, by taking advantage of spin-orbit coupling in the valence band and by using LH transitions in a waveguide, we have demonstrated the generation and detection of electron spin coherence in semiconductors without either an external or internal DC magnetic field. In the absence of spin precession, the electron spin coherence is detected through effects of quantum interference in spectral domain coherent nonlinear optical response. This new approach of coherent spin manipulation removes the complexity and/or added decoherence of external or
effective internal magnetic fields and potentially opens up new avenues for spintronics and spin-based photonics.
CHAPTER VII

STARK EFFECTS IN GaAs QUANTUM WELL WAVEGUIDES

In the previous chapter we have presented induced transparency arising electron spin coherence. In this chapter, we will present experimental results of Rabi oscillations arising from dipole coherence in GaAs QW waveguide.

Introduction

Observation of Rabi oscillations in a system is an indication that the system can be used for further coherent control experiments. Rabi oscillation is a quantum mechanical effect with no classical analog [100-101]. In a two-level system, Rabi oscillations are the sinusoidal time evolution of the population difference occurring at Rabi frequency for time scales shorter than the dephasing time. Effects of Rabi oscillations can be observed in time domain [102-104] or frequency domain [105-106]. In contrast to atomic systems, Rabi oscillations in semiconductors become complicated and interesting due to many-body effects [107-108]. Rabi oscillations have been experimentally observed in semiconductor quantum wells (QWs) [109-111] as well as in quantum dots [112-113].

Rabi oscillations in frequency-domain and time-domain

We present direct experimental evidence of Rabi oscillations in GaAs quantum well waveguides. In waveguide geometry light propagates in the quantum well plane, and so the optical interaction length is given by the length of the waveguide in contrast to all the references above where light propagates along growth direction (thus optical interaction length is limited by QW width). Given
this clear and useful advantage of long optical interaction length in the waveguide, it is quite natural to ask if one can perform coherent control experiments with waveguide. Direct evidence of Rabi oscillations in coherent pump-probe experiment in waveguide opens up an interesting and new way of coherent manipulations in QW waveguides.

**FIGURE 41.** Experimental scheme for Rabi oscillation experiments. (a) Schematic of the experimental configuration. (b) Optical selection rules for dipole transitions in GaAs QW.
Experimental studies were carried out in a slab waveguide consisting of a 17.5 nm GaAs/AlGaAs (001) single QW and GaAs/AlGaAs superlattice cladding layers grown by molecular beam epitaxy. The waveguide was cleaved to a length of nearly 100 μm and was mounted on a bridge-like sample holder attached to the cold finger of an optical cryostat. Figure 41(a) shows the experimental scheme. In our transient nonlinear technique both pump and probe are derived from a 82 MHz tunable mode-locked Ti:Sapphire pulsed laser. Probe pulse with duration around 200 fs propagates in the quantum well plane. A 20X optical objective was used to couple the incident laser beams into the waveguide. Pump pulse with duration around 8 ps obtained by spectrally filtering 200 fs pulse propagates along the growth direction. A 20 cm plano-convex lens was used to focus pump on the sample. Long pump pulse drives the Rabi oscillations, whereas, much shorter probe pulse time gates the transient event on a scale shorter than dipole dephasing time. A

FIGURE 42. Transmission spectra of a GaAs QW waveguide made from 17.5 nm GaAs QW at 10 K.

...
FIGURE 43. Rabi oscillation in the frequency-domain and time-domain. (a) Rabi splitting due to the Rabi oscillations of HH excitonic transitions. (Inset) shows dependence of the Rabi splitting on pump intensity. (b) Rabi oscillation can be seen directly in the time-domain also.

grating spectrometer with photomultiplier tube detector is used to spectrally resolve and measure the probe transmission. Figure 41(b) shows the polarization selection rules. Figure 42 shows the linear transmission spectra of the waveguide for TE (electric field parallel to the QW plane) and TM (electric field normal to the QW plane) polarized fields, respectively. For the TE polarization both HH and LH exciton resonances are observed, whereas for the TM polarization HH exciton resonance is nearly negligible, indicating a nearly ideal polarization selection rule [114].

Figure 43(a) shows the Rabi splitting in the absorption spectrum measured by the probe when the pump is at heavy-hole exciton transition. The measured Rabi splitting is consistent with an order-
FIGURE 44. Effect of pump-probe delay on Rabi oscillations. For (a)-(e) pump-probe delays are +8, +2, 0, -2 and -5 ps respectively. Positive delay means pump comes before probe.

of-magnitude calculation using the average pump intensity over pump pulse duration. As shown in Figure 43(b), we also observed Rabi oscillations by measuring probe absorption around applied pump wavelength as a function of pump-probe delay. Observed oscillations around zero pump-probe delay are not coherent spectral oscillations because coherent oscillations occur when both pump and probe are short compared to dipole dephasing time [115]. In our experiments, pump duration ~ 8 ps is not short compared to dipole dephasing time of ~ 5 ps in our sample. The measured Rabi periods of order 1 ps agrees with spectral domain absorption measurements as in Figure 43(a). Inset of Figure 43(a) shows Rabi splitting measured from absorption spectra as a function of pump intensity. For the range of intensities studied we find an approximate square-root dependence of Rabi splitting on pump intensity in good agreement with theoretical
expectations. Even though the effective Rabi frequency and hence Rabi splitting is renormalized by Coulomb effects, renormalization does not modify basic square-root dependence very much based on basic Hartree-Fock form of the semiconductor Bloch equations [107].

![Graph showing polarization dependence of the Rabi splitting at T = 10 K.](image)

**FIGURE 45.** Polarization dependence of the Rabi splitting at $T = 10$ K. (open circle) for TE probe propagating in the QW plane and TE pump propagating along growth direction. (close circle) for TE probe propagating in the QW plane and TM pump propagating along growth direction. (solid curve) transmission spectrum for TE polarized probe in absence of the pump.

To prove that the observed splitting is not incoherent spectral hole burning, we have shown dependence of Rabi splitting on the pump-probe delay in Figure 44. The exciton linewidth in our sample includes some inhomogeneous broadening. A pump with spectral width narrower than the inhomogeneous linewidth may cause spectral hole burning (SHB). However, because exciton population relaxation time in our sample is ~ 500 ps, we would expect spectral hole burning to persist for this duration. Appearance of Rabi splitting only within a short range around zero
pump-probe delay proves that the splitting observed is not spectral hole burning. Moreover, we observe Rabi splitting larger than the inhomogeneous line width which is not possible if the effect is due to SHB. Figures 45 and 46 show polarization dependence of the Rabi oscillations in frequency domain and time-domain respectively. As expected for orthogonal polarizations of pump and probe, Rabi oscillation signatures are relatively weak. Reason is that for orthogonal polarizations pump and probe can not create population pulsation efficiently [49].

![Figure 46](image)

**FIGURE 46.** Polarization dependence of the Rabi oscillations in time-domain at $T = 10$ K. (a) for TE probe propagating in the QW plane and TE pump propagating along growth direction. (b) for TE probe propagating in the QW plane and TM pump propagating along growth direction.

**Summary**

In conclusion, we have presented direct experimental evidence of Rabi splitting in GaAs quantum well waveguide. Our results open up the possibilities of optical coherent control experiments in
waveguides where we have definite advantage of long optical interaction length. Long optical interaction length in waveguides might be useful for slow light, quantum memory for photons etc.
CHAPTER VIII

TUNABLE OPTICAL PULSE DELAY IN GaAs QW

Electron spin coherence without magnetic field in previous chapters was studied in waveguide geometry with light propagating in the QW plane. In the next two chapters, we return to usual normal geometry experiments where light propagates perpendicular to the QW plane and along growth direction. We report the experimental realization of a tunable optical delay by exploiting unique incoherent nonlinear optical processes in semiconductors. The tunable optical delay takes advantage of the strong Coulomb interactions between excitons and free carriers and uses optical injection of free carriers to broaden and bleach an exciton absorption resonance. Fractional delay exceeding 200% has been obtained for an 8 ps optical pulse propagating near the heavy-hole excitonic transition in a GaAs quantum well structure. Tunable optical delay based on optical injection of free carriers avoids strong absorption of the pump beam and is also robust against variations in the frequency of the pump beam.

Tunable optical delay via carrier induced dipole decoherence

Recent dramatic experimental demonstration of slow and fast light has stimulated considerable interest in dynamic control of the group velocity of light and in the development of tunable all-optical delays for applications such as optical buffers [12-15]. Earlier slow light studies have used electromagnetically induced transparency (EIT) and coherent population oscillation (CPO). To realize tunable optical delays, more recent experimental studies have also pursued a variety of other physical mechanisms, including stimulated Brillouin and stimulated Raman scatterings [24-
27], and optical wavelength conversion [28]. The materials used in these studies range from atomic vapors [12-14], doped ions in crystals [15], optical fibers [24-28], and semiconductors [31-35]. For compact all-optical buffers, chip-scale semiconductor-based tunable optical delays are highly desirable.

In EIT and CPO, optical interactions between a signal and a pump induce a narrow transparency window within an absorption resonance. Tunable optical delay can be achieved via the pump intensity dependence of the spectral width as well as the depth of the transparency window. In stimulated light scattering, the parametric gain depends on the intensity of the pump beam, which can be used to generate tunable optical delay for the signal beam. In optical wavelength conversion, the spectral dependence of the group velocity in a fiber provides a convenient mechanism for tunable optical delay. All these schemes are based on the use of coherent nonlinear optical processes.

We propose and demonstrate experimentally a scheme that uses incoherent nonlinear optical processes to realize tunable optical delays in semiconductors. The proposed scheme exploits the strong Coulomb interactions between excitons and free carriers and uses optical injection of free carriers to broaden and bleach an exciton absorption resonance. A fractional delay exceeding 200% has been obtained for an 8 ps optical pulse propagating near the heavy-hole (HH) excitonic transition in a GaAs quantum well (QW) structure. Tunable optical delay employing free carrier injection avoids strong absorption for the pump beam and is also robust against variations or fluctuations in the frequency of the pump beam.

When an optical pulse with frequency, \( \nu \), propagates in a dielectric medium, the phase velocity is \( c/n \), where \( n \) is the refractive index, and the group velocity is given by

\[
\nu_g = \frac{c}{n + \nu \left(\frac{dn}{d\nu}\right)}.
\]
Near an absorption resonance, the group velocity depends strongly on both the spectral lineshape as well as the transition strength associated with the resonance. Varying the spectral lineshape as well as the transition strength modifies the group velocity, resulting effectively in a tunable optical delay.

In a direct gap semiconductor, optical transitions near the band edge are characterized by excitonic resonances. Nonlinear optical properties of an excitonic system are strongly modified by many-body Coulomb interactions [48]. In particular, exciton-exciton scattering or exciton-carrier scattering can induce significant dephasing or spectral broadening of the exciton resonance. These excitation-induced dephasing processes play an important role in both coherent and incoherent nonlinear optical responses in semiconductors [40-41].

To realize tunable optical delay via spectral broadening and bleaching of an excitonic resonance, we propose to optically inject free carriers with a pump beam slightly above the band gap. At relatively low excitation levels, exciton-carrier scattering is considerably more efficient than exciton-exciton scattering in broadening the excitonic resonance. Perhaps more importantly, the use of an off-resonant pump avoids the strong pump absorption occurring when the pump is resonant or nearly resonant with the relevant optical transition.

**FIGURE 47.** Schematic of setup for the time-of-flight measurement of a signal pulse.
The experimental studies are carried out in a high quality undoped (001) GaAs/Al$_{0.3}$Ga$_{0.7}$As QW sample grown by molecular beam epitaxy. The sample contains 50 periods of 17.5 nm GaAs wells and 15 nm Al$_{0.3}$Ga$_{0.7}$As barriers. For transmission measurements, the substrate of the QW sample is removed with selective chemical etching. The QW sample glued onto a sapphire disc is mounted on a cold finger of a helium flow cryostat. The signal pulse used in our study features duration of 8 ps and a spectral linewidth of 0.2 nm (see Figure 48). A spectral pulse shaper is used to derive the nearly transform-limited signal pulse from a femtosecond mode-locked Ti: Sapphire laser. The output coming directly from the femtosecond mode-locked Ti:Sapphire laser has a pulse duration of 150 fs and is used to measure the delay and temporal lineshape of the signal pulse via sum frequency generation in a BBO crystal, as shown schematically in Figure 47. For optical injection of free carriers, a separate picosecond mode-locked Ti: Sapphire laser with pulse duration of 60 ps is used. The two mode-locked lasers are synchronized with a repetition rate of 80 MHz.

**FIGURE 48.** Typical signal pulse in (a) time-domain and (b) frequency domain. Solid lines are Gaussian fit to the data.
Figure 49 shows absorption spectra of the QW sample near the band edge with and without optical injection of free carriers. In the absence of free carrier injection, the absorption spectra are characterized by well resolved HH and light-hole (LH) exciton absorption resonances. The HH exciton resonance features a linewidth of 0.78 nm at T=80 K and 0.55 nm at T=20 K. For optical injection of free carriers, we set the wavelength of the pump beam to $\lambda=795$ nm, slightly above the band gap of the GaAs QW. From the absorption of the sample at the pump wavelength, we estimate that a pump beam with an average power of 1 mW (using a pump spot size of order 200 $\mu$m) generates approximately a carrier density of $3 \times 10^9$/cm$^2$. The free carrier injection leads to highly efficient broadening of the exciton resonance. At T=80 K, the exciton resonance nearly vanishes at a pump power near 10 mW, as shown in Figure 49(a).

![Figure 49](image)

**FIGURE 49.** Carrier induced dephasing of excitonic transitions. Absorption spectra near the band edge obtained in the presence of free carrier injection by a pump beam at $\lambda=795$ nm and with the pump power indicated in the figure. (a) T= 80 K. (b) T=20 K. The dashed lines show, as a reference, the absorption spectra obtained in the absence of free carrier injection.

The spectral broadening of the exciton resonance depends on details of the exciton and carrier dynamics as well as details of the underlying many-body Coulomb interactions. A thorough
discussion of these processes is beyond the scope of this paper. Within the context of tunable optical delays, we wish to point out that as shown in Figure 49, free carrier injection induces a much greater broadening of the exciton resonance at $T=80$ K than that at $T=20$ K. Earlier studies of transient four-wave mixing have indicated that at relatively low excitation levels, the collision coefficient for exciton-free carrier scattering is nearly eight times greater than that for exciton-exciton scattering [42]. At low temperature, a significant fraction of free carriers injected can subsequently form excitons. As a result, the effective carrier density at lower temperature can be considerably smaller than that at higher temperature, leading to a strong temperature difference in the spectral broadening of the exciton resonance.

![Figure 50](image)

**Figure 50.** Line broadening as a function of the intensity of the control beam. As the power increases, increased carrier density increases the dipole dephasing and broadens the exciton resonance.
It should be pointed out that at relatively high carrier densities, screening of Coulomb interactions becomes important. The exciton spectral broadening is thus not expected to scale linearly with the carrier density at these high carrier densities. Figure 50 shows linewidth of excitonic resonance as a function of the power of the control beam. We also note that in addition to optical injection of free carriers, other approaches, such as Franz-Keldysh effects, for which an external electric field is applied in the plane of the QW, can also be used to broaden and bleach the exciton resonance [43].

To illustrate how the exciton absorption affects the group velocity of a nearly resonant optical pulse, we plot in Figure 51 the result of a time-of-flight measurement of a weak signal pulse after

![Time-of-flight measurement of pulse with different center wavelengths after its transmission through the QW sample. The central wavelength of the signal pulse is indicated in each figure. The results were obtained at 20 K and with the sum frequency generation shown schematically in Figure 47. Note that t=0 was set to the peak of the signal pulse when the pulse was tuned far below the HH exciton resonance. The average intensity of the signal pulse is 0.2 W/cm².](image)

**FIGURE 51.** Time-of-flight measurement of pulse with different center wavelengths after its transmission through the QW sample. The central wavelength of the signal pulse is indicated in each figure. The results were obtained at 20 K and with the sum frequency generation shown schematically in Figure 47. Note that t=0 was set to the peak of the signal pulse when the pulse was tuned far below the HH exciton resonance. The average intensity of the signal pulse is 0.2 W/cm².
its transmission through the QW sample (no free carrier injection was used in these measurements). As shown in Figure 51, the signal pulse becomes more delayed, i.e. arrives at a later time, as the central wavelength of the pulse approaches the heavy-hole (HH) resonance. The overall experimental results are in general agreement with the theoretical expectation. Note that at the exciton absorption line center and in the region of the anomalous dispersion, the group velocity can become negative. In this case, the peak of the pulse can emerge from the sample before the peak of the pulse enters the sample. As shown in earlier experimental and theoretical studies, this negative group velocity arises from pulse reshaping; namely, the leading edge of the pulse is less attenuated than the trailing edge of the pulse [39]. Since the primary emphasis here is to demonstrate tunable optical delay by modifying the group index, we will discuss in more detail pulse propagation in the anomalous dispersion region in semiconductors in a separate publication.

**FIGURE 52.** Time-of-flight measurement of pulse with and without control after its transmission through the QW sample. Data shown with (open circles) and without (squares) free carrier injection by a pump beam at $\lambda=795$ nm. (a) $T=80$ K and $I_{\text{pump}}=2$ mW. (b) $T=20$ K and $I_{\text{pump}}=4$ mW. The solid lines are numerical fit to a Gaussian. The central wavelength of the signal pulse is at $\lambda=815.69$ nm and $\lambda=811.72$ nm for (a) and (b), respectively. Note that here, $t=0$ was set to the peak of the signal pulse in the presence of optical injection of free carriers.
The spectral broadening and optical bleaching of the exciton absorption resonance shown in Fig. 2 provides an effective mechanism for realizing tunable optical delay. Figure 52 compares directly time-of-flight measurements of the signal pulse after its transmission through the QW sample with and without the optical injection of free carriers by a pump beam. For clarity of display, normalized intensity is shown in Figure 52. The free carrier injection leads to a 2-fold increase in the signal transmission for Figure 52(a) and an 8-fold increase in the signal transmission for Figure 52(b). Fractional pulse delay (the ratio of the pulse delay over the pulse duration) exceeding 200% has been observed at T=20 K (see Figure 52(b)).

**FIGURE 53.** Control power dependence of the pulse delay at T = 80 K. Different powers of the control beam inject different amount of free carriers, and hence different dipole dephasing.

Smaller fractional delays have been observed at higher temperatures. The large delay, however, is also accompanied by a broadening or reshaping of the temporal line shape of the signal pulse, as expected theoretically. The fractional delay observed in Figure 52 agrees well with what is
expected for the group delay induced near the HH exciton absorption resonance shown in Figure 51. Figure 53 shows controllable delay at $T = 80$ K for different control powers.

The primary limitation for achieving greater fractional delay is the strong absorption of the signal beam near the exciton resonance. The bandwidth of the tunable optical delay is limited by the spectral linewidth of the exciton resonance. Much greater bandwidth can be achieved at higher temperature or with exciton resonances that are strongly inhomogeneously broadened.

Finally, we note that dynamically shifting the exciton resonance can also be used as a mechanism for tunable optical delay since near an exciton resonance, the group index and thus the optical delay depends strongly on the relative spectral position between the exciton resonance and the signal pulse, as shown in Figure 51. Resonant or near resonant excitation of excitons can shift and broaden the exciton resonance via exciton-exciton interactions and/or optical Stark effects. Quantum confined Stark effects also provide a highly effective mechanism for shifting the exciton resonance [43].

Summary

In summary, we have demonstrated tunable optical delay by broadening and bleaching exciton absorption resonance with optical injection of free carriers. Fractional delay exceeding 200% has been achieved with an 8 ps signal pulse propagating near the exciton absorption resonance. We hope that these studies will stimulate further activities of exploiting unique optical properties of semiconductors for applications in tunable optical delay.
In this chapter, we have studied both linear and nonlinear Gaussian pulse propagation in GaAs quantum wells. Near resonances pulse breaks up into two parts where one output Gaussian pulse precedes the input pulse, whereas, the other output Gaussian pulse follows the input pulse. Velocity changes sign as we scan through a resonance. Pulse break up can also be induced using a pump beam to spectrally broaden and optically bleach the resonance. Pulse velocity has been measured using a time-of-flight technique.

**Pulse propagation through dispersive medium**

As mentioned in the introduction chapter of this dissertation, the propagation of light pulse through a dispersive medium has been a topic of interest for long [2, 38]. Fundamental meaning of negative, positive, zero, superluminal and ultraslow group velocity have been the topic of interests for both conceptual and practical reasons. Gaussian pulse propagation through dispersive media has been studied theoretically in detail [119-120, 122]. Effects of both coherent [123, 128-129] and incoherent [127] nonlinear optical processes on pulse propagation have been studied. Pulse propagation in the context of optical fiber communications [130] and also in the context of recent observations of supercontinuum generation [131] has been reported. Still there remain some issues to be resolved and optical control of pulse propagation is not studied in detail. In this context, we report breakdown of the definition of group velocity and influence of nonlinear effects such as carrier induced dephasing on pulse propagation.
In our studies, we have injected free carriers optically by using a control beam. Phase velocity is given by \( c / n \), whereas, group velocity is given by

\[
\nu_g = \frac{c}{n + \nu \frac{dn}{dv}}
\]

where \( c \) is the velocity of light in vacuum and \( \nu \) is the center frequency of the light pulse. For normal dispersion \( dn/d\nu > 0 \) and the group velocity is less than \( c \). But if the medium has an absorption line near \( \nu \), dispersion becomes anomalous and group velocity can be larger than \( c \), or even negative [124] because of \( dn/d\nu < 0 \) in the anomalous dispersion region. Meaning of group velocity tends to breakdown for anomalous dispersion because relativity doesn’t allow information to be propagated faster than \( c \). Sommerfeld and Brillouin [38] studied the propagation of a sinusoidal signal through anomalous dispersion with the signal suddenly turned on at some time \( t_0 \). They showed that even though the precursors of the pulse can travel faster than \( c \), the main signal travels slower than \( c \). Optical precursors have been experimentally observed [125-126]. To resolve the problem, different kinds of velocity were defined such as energy velocity and signal velocity. Nevertheless, it has been shown [119] that under certain

**FIGURE 54. Schematic of setup for time-of-flight measurements with control. Control beam is used to manipulate both pulse delay and shape.**
realistic conditions a Gaussian pulse can travel through anomalous dispersion with group velocity $v_g$.

**FIGURE 55.** Pulse breakup around HH resonance. Pulse breaks up as the center wavelength of the signal pulse scans through the HH resonance at $T = 20$ K.

Their theoretical predictions were confirmed by Chu et.al. [39] who reported experimental results showing that pulse indeed travels with group velocity through anomalous dispersion. They also showed that i) pulse velocity measured in their experiment is, in fact, group velocity rather than energy velocity and ii) both shape and width of the pulse remains unchanged after propagation. Pulse velocity that is more than c or is negative can be attributed to pulse reshaping and does not necessarily mean violation of relativity or causality. Apparent contradiction between energy
velocity description and group velocity description is shown to be just different parameter regimes of the modified asymptotic description of the Gaussian pulse propagation. Still there is no single analytical solution with general applicability, even modified asymptotic description becomes invalid for certain conditions.

**FIGURE 56.** Pulse breakup around LH resonance. Pulse breaks up as the center wavelength of the signal pulse scans through the LH resonance.

We report experimental results that indicate that neither energy velocity nor group velocity description can be applied straightaway. We see a nearly Gaussian pulse breaking up into two parts when the pulse center frequency is tuned near excitonic resonances in GaAs quantum well
Analytical approaches so far are not sufficient, instead numerical approach have been used to explain our results.

Pulse velocity has been measured by a time-of-flight technique. The experimental studies are carried out on MBE grown high quality GaAs/AlGaAs QW sample consisting of 50 periods of 17.5 nm wide GaAs QWs. The sample is mounted on c-axis normal sapphire disc and GaAs substrate is removed by selective chemical etching. Nearly Gaussian pulses used in our study are of duration around 8 ps with spectral bandwidth of 0.2 nm. A spectral pulse shaper is used to derive nearly transform-limited signal pulses from a mode-locked Ti: Sapphire laser. Direct output of the mode-locked laser has pulse duration of around 150 fs and is used to measure the delay and temporal line shape of the signal pulses after propagation through the sample using sum frequency generation in a BBO crystal. To study nonlinear optical effect on pulse propagation, we injected free carriers optically using a separate picosecond mode-locked Ti: Sapphire laser with pulse duration of around 60 ps. The two mode-locked lasers used are synchronized with the same repetition rate of 80 MHz.

Figures 55 and 56 show a set of cross-correlation data as we scan the pulse center wavelength through heavy-hole (HH) and light-hole (LH). Note that when pulse center wavelength is tuned near resonance pulse breaks up into two parts – one part moves with positive pulse velocity and the other part moves with negative pulse velocity. A monotonic increase in pulse delay is observed (more than 20 ps) as the pulse center wavelength is tuned from higher wavelength towards resonance. Greater pulse delay near resonance is also accompanied by stronger absorption. Pulse broadening and greater delay near resonance can not be attributed to possible multiple reflections in the sample because of high attenuation near resonance. The average intensity of the signal pulse is 0.2 W/cm² that is well below the saturation intensity in this time scale and therefore Figure shows pulse propagation in linear regime where refractive index is not
modified due to the presence of the signal pulse. Self-induced transparency or spectral hole burning effects as reasons for pulse break up can be ruled out for the intensities used in the experiment.

**FIGURE 57.** Pulse delay (left) and output pulse width (right) vs. center wavelength of the signal pulse show different behaviour around LH and HH resonances. Behavior around HH is highly asymmetric. (Inset) shows the absorption spectrum of the GaAs QW sample used.

Pulse delay and output pulse width as a function of pulse center wavelength is shown in Figure 57. Inset shows the absorption spectrum of the sample used. Dispersive nature of the plot is quite different from results reported before. It follows neither group velocity description nor energy velocity description. Pulse velocity changes sign abruptly near resonance with associated pulse break up. The fact that we see pulse break up at both the HH and LH resonances indicates that it is not just the absorption length but other parameters such as pulse bandwidth are important deciding factor for pulse propagation.

To study pulse propagation in nonlinear regime we have used a far detuned pump beam to inject free carriers. The spectral broadening and optical bleaching of the excitonic resonance due to this
pump is shown to be effective in controlling the signal pulse propagation. Figure 58 shows the time-of-flight measurement of the signal pulse as we vary the intensity of the pump beam. Pulse break up is induced and pulse velocity changes sign due to the pump beam.

![Figure 58](image)

**FIGURE 58.** Manipulation of pulse shape using control beam. Different intensities of the control beam results in different output pulse shapes.

Depending on where we tune the pulse center wavelength the effect of pump beam can be different because spectral broadening and optical bleaching are different at different wavelengths. Figure 59 shows the probe power dependence of pulse propagation at T = 80 K.

To model Gaussian pulse propagation through a dispersive medium let us consider an input Gaussian pulse of the form

$$E_0 = e^{i\omega_0 t - \tau(0) t^2}$$

where $\omega_0$ the pulse is center frequency and $\tau(0)$ is the initial complex pulse width.
After propagating a distance $z$ through a dispersive medium characterized by frequency dependent propagation constant $k(\omega)$, the output pulse is given by

$$E(z, t) = \frac{e^{i(\omega(0) t - k(\omega(0)) z)}}{2\pi} \int_{-\infty}^{\infty} e^{i(\omega - \omega(0))(t - k z) - (\omega - \omega(0))^2/4\tau(z)} d(\omega - \omega(0))$$

(1.1)

where $\tau(z) \equiv \frac{1}{\tau(0)} + 2i k^-$; $k^-$, $k_-$ are first and second derivative of dispersion curve $k(\omega)$ with respect to $\omega$. We solve (1.1) numerically to explain our results. Numerical results seem to explain our results along with previous works where two different descriptions of pulse velocity seem to be valid.

**FIGURE 59.** Probe power dependence of the pulse propagation at $T = 80$ K.
Failure of group velocity description in our case can be attributed to the fact that our experimental conditions do not match the conditions for which Garrett and McCumber’s predicted [119] and Chu et.al. [39] experimentally verified the validity of group velocity description. While Sommerfeld and Brillouin considered a step-function envelope of the pulse, Garrett and McCumber considered Gaussian pulses with smoothly time-varying front and back edges. They also assumed i) that the bandwidth of the pulse is much less than that of the absorption line i.e. $\Delta \nu_{\text{laser pulse}} \ll \Delta \nu_{\text{absorption line}}$, and ii) that the absorption length is much less than $\left(\frac{\Delta \nu_{\text{absorption line}}}{\Delta \nu_{\text{laser pulse}}}\right)^2$. These conditions lead to the result that the pulse propagates with group velocity and emerges with almost identical input pulse shape. In our experiments, at 20 K, absorption line width is around 0.6 nm and absorption length around 12. Given the bandwidth of around 0.2 nm of the laser pulses used, it is clear that the above conditions are not met and expectedly our experiments do not fit group velocity description. In the polariton [117] picture where light is strongly coupled to the excitons, wide bandwidth of the laser pulse compared to resonance line width reinforce the break down of the group velocity description.

On the other hand, validity of energy velocity description based on the asymptotic approximation decreases as the pulse width is increased. Pulse width of around 8 ps used in our experiments is around four orders of magnitude higher than that used in Oughstun and Balicstsis’ analysis. Experimental results presented in this paper motivate further experimental and theoretical studies to differentiate different possibility of pulse break up mechanism. Given extensive interests in ultrafast optics and plethora of transient phenomena, it is of utmost important to understand pulse propagation through anomalous dispersion region.

**Summary**

In summary, we have shown that under conditions where both group velocity description and energy velocity description become insufficient signal pulse breaks up into two parts moving with
velocities of opposite signs. We have also shown that using a pump beam propagation of the signal pulse can be controlled. We hope these results will motivate further pulse propagation studies in both linear and nonlinear regimes.
CHAPTER X

SUMMARY AND FUTURE WORK

Summary
Research work presented in this dissertation is based on two general themes that are related in some respect. First, tunable all-optical delay to improve the speed of communications. Second, coherent spin manipulation in semiconductors, particularly electron spin.

The dramatic experimental demonstration of slow light in atomic vapors via electromagnetically induced transparency (EIT) has stimulated considerable interest in the dynamic control of the group velocity of light and in the development of tunable all-optical delays. In EIT, destructive quantum interference arising from quantum coherence leads to a narrow transparency window within an absorption resonance, leading to low group velocity of light. To realize tunable optical delays, recent studies have also used coherent processes like coherent population oscillation, stimulated Brillouin and stimulated Raman scatterings, and optical wavelength conversion.

In this dissertation, we have presented experimental studies of realizing all-optical tunable delays via nonlinear optical processes in semiconductor quantum wells (QWs). Two different approaches have been pursued. The first employs EIT arising from electron spin coherence in semiconductors and the second is based on efficient carrier induced exciton dephasing in QWs.

For the EIT-based approach, we have taken advantage of spin-orbit coupling in the light-hole valence band in GaAs QW waveguides. We have also developed techniques to carry out pump-probe spectroscopy in semiconductor waveguides at temperature as low as 4 K. Induced
transparency due to electron spin coherence has been observed in the differential transmission. However, relatively small transparency limits the fractional delay that can be achieved in this approach. Narrow transparency window also limits the bandwidth. Nevertheless, our studies have provided a new approach for inducing and manipulating electron spin coherence in the absence of external magnetic fields.

We have overcome shortcomings of EIT-based approach by exploiting unique incoherent nonlinear optical processes in semiconductors. Optical injection of free carriers leads to bleaching due to saturation and broadening due to enhanced dipole dephasing induced by strong Coulomb interactions between excitons and free carriers. Spectral broadening and bleaching of excitonic absorption due to free carrier injection by a control beam essentially modifies the dispersion and hence provides an efficient optical control of group velocity and delay. Fractional delay exceeding 200% has been obtained for an 8 ps optical pulse tuned near the heavy-hole in a GaAs QW. Our method provides delay better than other methods, higher bandwidth, lower control absorption and stability against control fluctuations.

On the other hand, coherent spin manipulation in semiconductors plays a central role in spin-based electronics and photonics and in spin-based quantum information technologies. Extensive experimental studies in this area have led to demonstration of remarkable phenomena such as robust electron spin coherence at room temperature, the transfer of electron spin coherence between molecularly bridged quantum dots, and evidences of multispin entanglement. One way of coherent spin manipulation is applying magnetic fields. For example, in GaAs quantum well (QW) the magnetic field can induce a Zeeman splitting between the two conduction band electron spin states. Dynamics of coherent superposition of these spin states can be probed with transient optical techniques by measuring the Larmor precession of the optically oriented electron spin around the applied magnetic field. Application of external magnetic fields is an extra
complication to add. Moreover, inhomogeneity of the applied magnetic field can degrade the much desired electron spin coherence. An interesting question of both conceptual and practical importance is whether coherent spin manipulation in semiconductors can be carried out without either external or internal magnetic fields.

In this dissertation, we have presented our studies on coherent spin manipulation in semiconductors, particularly in semiconductor waveguides. Observed signature of both the electron spin coherence and excitonic coherence along with the added advantage of long interaction length make semiconductor waveguides promising for coherent optical studies. Semiconductor waveguides provides long interaction length because light propagates in the QW plane, whereas, normally light propagates along growth direction perpendicular to the QW plane. In waveguides interaction length is decided by the length of the waveguide, whereas, in the other case interaction length is given by the QW width.

As an interesting outcome of our research, we have shown that in our experimental regimes definition of both the group velocity and energy velocity breaks down for pulse propagation through anomalous dispersion. We have also demonstrated that the shape of a pulse can be manipulated using a control beam motivating further studies of pulse shaping using nonlinear optical effects.

Some of our main results are summarized below:

Physics

1. Manipulations of electron spin coherence without magnetic fields. Electron spin coherence time observed is more than 300 ps (compared to dipole coherence time around 5-10 ps).

2. Breakdown of group velocity and energy velocity description of pulse propagation through anomalous dispersion
3. Signature of Rabi oscillations associated with excitonic transitions

Applications

1. New approach of tunable all-optical delay for better performance. Delay-bandwidth product obtained is more than 2 with much higher bandwidth compared to other methods.

2. Developed techniques of pump-probe spectroscopy in semiconductor waveguides from room temperature to as low as 4 K

3. Demonstrated pulse shaping using nonlinear optical effects

Future Work

For eventual device applications such as all-optical buffer, we need optically tunable delay-bandwidth product in thousands. Even though our approach has better performance than other existing approaches, our delay-bandwidth product of little more than 2 is not enough. But our approach has the potential to be extended to get desired delay-bandwidth product. Further studies need to be done to improve our results, possibly in combination with other approach. One such possibility is to extend our approach with tapered QW structure so that excitonic resonance varies as the well width varies. By dispersing our signal pulse and then going through a tapered QW structure, we can increase the bandwidth and hence the delay-bandwidth product [37].

Transparency arising from quantum interference due to electron spin coherence in semiconductor waveguides is particularly encouraging for further studies. For spintronics applications, electron spin coherence in semiconductors has many potential applications. To improve the observed electron spin coherence time of about 300 ps [60], further studies should be done to optimize the material, dimensionality and waveguide design.


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