Femtosecond-resolved semiconductor dynamics with mid- and far-infrared pulsed lasers

By

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In the name of God, Most Gracious, Most Merciful

Read in the name of your Lord who created

Created man, out of a clot of congealed blood

Read, and your Lord is the most Generous

He Who taught (the use of) the pen

Taught man that which he knew not

Quran, al-Alaq (1-5)
To:

My Precious Parents,

and

My Dear Sisters & Brother
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Abstract

In this thesis mid-infrared time-resolved pump-probe measurements are presented to evaluate spin dynamics in narrow gap semiconductors. The polarization of the pump beam is modulated and the transmittance change in the probe beam is measured. The optical polarization is measured versus time delay and its decay yields the spin lifetime.

Results show that the spin lifetime decreases with increasing temperature. Between InAs and InSb films, although InAs shows longer spin lifetime, it has an accumulation layer in the surface with high current and very short spin lifetime, while InSb has a surface depletion layer. The spin dephasing dominates for the range of temperature 77 – 293 K for thick films, however thin films show stronger spin flip relaxation mechanism. For AlInSb/InSb quantum wells, the spin lifetime depends on the well width, mobility, and carrier concentration, and the dephasing mechanism dominates for high mobility, but for low mobility at low temperature the spin flip scattering is dominant.

We have also investigated the Lande g-factor for the first time at room temperature, using the same technique by adding an external magnetic field causing spin precession. Our experimental results show that with increasing temperature and energy, the absolute g-factor, decreases. In comparison, simple predictions of k.p theory for temperature dependency of g-factor, which changes primarily because of the change in the band gap, suggest the opposite. We find leaving only the dilational part in the temperature dependence of the energy gap, improves the consistency between experimental and theoretical g-factors.

Finally, a far-infrared time-resolved pump-probe study of population dynamics of shallow donor impurity states (P, As) in silicon shows that at low temperatures, the dominant linewidth broadening is due to direct phonon decay rather than dephasing collisions or inhomogeneous broadening. The achieved long lifetimes are convenient for applications such as quantum information processing.
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Recent publications


Conference presentation

Introduction

1.1 Spin injection and spin dynamics

The electron has three properties: charge, mass, and spin. Due to the electron's intrinsic angular momentum (spin), an electron behaves like a small magnet. Besides of controlling electronic charge in semiconductors, recently, there has been extreme interest in using spin. Spin dynamics have drawn much attention which leads to new area called spintronics- "spin transport electronics". There are two possible ways to generate spin-polarized carriers in semiconductors: optically and by electrical injection from a ferromagnet or from a magnetic semiconductor. Spin transport has the advantage compared to charge transport: that quantum coherence can be maintained on much larger time scales. The importance of spin-transport phenomena in condensed-matter physics has increased over the past decade.

When the polarized carriers are created, since the spin interacts with orbital angular momentum, the polarization vanishes quickly. Hence, it is important to find ways to suppress spin relaxation in order to be able to manipulate spin and transport polarized carriers over some distance.

Spin dynamics in semiconductors have been studied extensively, with the majority of work focusing on optically injected spins. It is possible to create spin-polarized carriers by the absorption of circularly polarized light based on the selection rules. Since the same selection rules are valid for recombination as for excitation, the spin-polarization can be deduced from the circular polarization of luminescence. Early research relied on the continuous-wave, polarization-resolved measurement of photoluminescence from these structures. Time-resolved measurements have been used to determine the degree of initial spin polarization and the time dependence of the polarized component of emission from an optically generated and spin-polarized carrier distribution for a wide range of near infrared, mostly for GaAs-based structures. In contrast, little attention has been focussed on spin dynamics in InSb and InAs, although they have narrow band gap suitable for mid-infrared ultrafast spectroscopy of the spin dynamics, which may play a key role in the future of spintronics. In this thesis we study spin dynamics and spin manipulation in such semiconductors.
Exposed to magnetic fields, many materials cause linearly polarized light to rotate its axis of polarization (the Faraday effect). Spin polarized carriers also induce magnetization; thus, the Faraday rotation can be used to detect spin-polarization. Similar to the Faraday effect, band filling effects (different absorption of right- and left-handed circularly polarized light) due to different occupation of the final states can be utilized to measure spin polarization. In this thesis we concentrate on the absorption technique.

Zeeman experiments in solids have been used for the study of electron energy bands. These studies in semiconductors provide information on the location and symmetry of the band edge extrema or critical points in the Brillouin Zone (BZ), the effective mass, and g-factor at and in the vicinity of the critical points, and the energy gaps between the band extrema.

The problem of measuring electron spins is of great interest for a variety of purposes from imaging the structure of molecules to quantum information processing.

### 1.1.1 Applications

Recent developments in spintronics offers opportunities for developing a new generation of devices combining standard microelectronics with spin-dependent effects arising from the interaction between carrier spin and the magnetic properties of the materials. For spintronics applications it is essential not only to have a long spin lifetime, but also to be able to control spin dynamics with an external electric or magnetic field. Spintronics or sometimes magnetoelectronics, which combine both the spin and charge of electrons along with manipulation of electron spin, provide devices with new functionality and increased performance. The advantages of these new devices would be nonvolatility, increased data processing speed, decreased electric power consumption, and increased transistor density compared with conventional semiconductor devices [Prinz (1998), Prinz (1999), Wolf (2001), Das Sarma (2001), De Boeck (2002)]. However, only the control of spin in semiconductors together with modern semiconductor technology can lead to extensive applications. Spin-based devices are closely connected with the development of nanotechnology. They can provide spin transistors, sensors, memories, tunable detectors, and bits for quantum computing. Some of these devices are mentioned here:
Sensors, GMR (Giant Magnetoresistance) read head used in hard disks [Prinz (1998)]; the largest application of nanotechnology to date is the use of GMR materials in read heads for high-density magnetic data recording.

Nonvolatile memories, MRAM (Magnetic random access memory), which has much faster writing times, consume less energy for writing and is insensible to cosmic radiation with much shorter data access time. In nonvolatile memories the information stays in the memory even if the electric power is switched off;

Spin transistors, Spin-FET (spin field-effect transistor) first demonstrated by Datta and Das [Datta (1990)]. It has not yet been realized in experiments, mainly because of large conductivity mismatch between a metal and a semiconductor [Schmidt (2000)].

Quantum computation is perhaps one of the most exciting potential applications of spintronics. The possible use of electron spins in the architecture of a solid-state based quantum computer has raised special attention in the study of the behavior of the electron spin coupled with an external magnetic field. In the single qubit operation it is of fundamental importance to have pure spin states in order to guarantee that no losses occur when the spins transport information [Zutic (2004)]. This goal may be achieved by manipulating the electron g-factor in semiconductor heterostructures and designing appropriate external gate control devices.

The evolution of MRAMs and various spin-based technologies could be critically important in facilitating the development of the first quantum computer. Spin injection into semiconductor [Aronov (1976)] is necessary in order to carry out quantum bit operations required for quantum computing. Electron-spin in an artificial atom can be preserved for a sufficiently long time that it can act as a memory to describe "0" or "1" in a quantum computer. Information storage can be done by direction of an electron spin (quantum bit).

The energy relaxation time is the time required to change from a high-energy excited state to the low-energy ground state, and can be regarded as the retention time of the information memory. In an electron-spin quantum computer, electron-spin in a semiconductor is used as a memory unit (quantum bit); it is one of the best candidates,
because it is expected to have a long relaxation time (more than 1 ms), longer than a typical period required for quantum logic gate operation (a few ps).

Semiconductor quantum dots or electrons bound to impurity atoms provide a potentially ideal electronic structure appropriate for quantum computing. In principle the electronic and spin state in these structures can be coherently controlled on a time scale that is short compared to the quantum decoherence time and that entangled states of quantum bits can be created. At the end of this thesis we investigate electron population lifetimes and decoherence in impurities in silicon.

1.2 \textit{k.p theoretical method}

In III-V zinc-blend semiconductors $k.p$ perturbation due to the periodic lattice is known to cause a mixing of the $\Gamma_6$ states in the conduction band with the $\Gamma_8$ and $\Gamma_7$ states in the valence band, and this mixing affects the optical, electric, and magnetic properties of the electrons. The energy band structure of InAs is shown as an example in Figure 0.1. The $\Gamma$'s are group theoretical symmetry labels for different types of electronic bands.

Multilevel $k.p$ models have been recognized to provide an efficient description of the band structure in the vicinity of the $\Gamma$ point for zinc-blend semiconductors. The only parameters needed for such calculations are the values of various band-gap energies and interband momentum matrix elements.

This model provides a theoretical method to calculate the energy bands of semiconductors, and therefore effective mass and $g$-factor of the carriers. Therefore, we have a powerful tool to compare the experimental data with theoretical results, which is used extensively in this thesis.
Introduction of quantum structures such as QWs and QDs modifies the Bloch functions and eigen-energies of conduction and valence states and changes fundamental properties of the semiconductors. Among these properties spin dynamics and Lande g-factor of electrons are very sensitive to the quantum confinement.

1.3 Narrow Gap III-V semiconductors such as InSb & InAs
Narrow gap semiconductors (NGSs) such as indium antimonide and indium arsenide are interesting semiconductors from the point of view of tests of semiconductor band structure calculations, because of the small electron effective mass, high electron mobility as well as strong spin-orbit coupling, and hence a strong Rashba effect. The heavy constituent atoms of InSb produce large relativistic effects such as spin-orbit coupling (responsible for the large, negative gyromagnetic ratio). InSb is a candidate material for room-temperature spintronic devices such as the Das-Datta spin-transistor [Datta (1990)], which relies on a coherent spin population manipulated by the Rashba effect, and thus detailed investigation of the spin-electronic structure in this material at room temperature is of high topical interest. In this work we have studied the spin dynamics and g-factor in InSb and InAs films, InSb quantum wells (QW) and some quantum dot (QD) samples up to room temperature. At the same time we resolve a
long-standing problem in the literature regarding the g-factor in GaAs at high temperature.

1.4 Far-infrared dynamics of donor transitions in silicon
By forcing crystals of germanium or silicon to grow with impurities such as boron or phosphorus or arsenic, the crystals gain entirely different electrical conductive properties. Because the indirect band gap in silicon prevents fast electron-hole recombination, having silicon based laser source has been complicated. Silicon based device would represent a significant progress in optoelectronic technology since it would open the approach for inexpensive massive integrated optical and electronic components. Quantum cascade lasers (QCLs), however, rely on intersubband transitions, effectively negating the disadvantageous indirect band gap, and they can provide mid- and far-infrared emission, because of small energy gaps of these transitions [Faist (1994)]. Si/SiGe heterostructures have advantages compared to III-V materials. Since Si:Ge bond is non-polar, there is no polar optical phonon scattering. Studies on p-Si/SiGe heterostructures have demonstrated that there is only a very weak temperature dependence on intersubband lifetimes up to 300 K [Kelsall (2005)]. Also, Si is a much more efficient conductor, important for higher temperature operation in QCLs. However, studies of the intersubband lifetime in Si-based QCLs have shown that the lifetimes are rather fast and it is difficult to engineer 4-level lasing schemes as for III-V QCLs, see e.g. [Califano (2007)]. An alternative strategy for light out of silicon is the silicon impurity laser [Pavlov (2000), Borak (2005), Pavlov (2006), Hubers (2005)]. In both of these types of potential laser material it is important to investigate the transition lifetimes.

Similar to spintronics, dynamics of donor transitions in silicon is also important for quantum computation schemes. Architecture for a quantum computer is presented in which spins associated with donors in silicon function as qubits; see for example [Keyes (2004), Kane (2003), Rodriguez (2004)].

Until now there was little or no time-resolved information regarding the lifetime of electrons bound to donor or acceptor impurity atoms, even in silicon, in spite of the technological importance. This is primarily because the wavelength required for transient optical techniques is in the far-infrared, where only free-electron lasers
operate. In this thesis we investigated the relaxation dynamic of both boron-doped p-type Si:Ge heterostructures and phosphor and arsenic doped n-type Si. The results for the p-type samples are complicated and show fast lifetimes (not presented here), whereas the n-type samples show single exponential decays with rather long lifetimes.

1.5 Thesis overview (description of chapter contents)
In this thesis we investigate several subjects in the following chapters:

In chapter 2 we review the three- and five-band $k.p$ model, for calculation of some important parameters in semiconductors such as Lande g-factor. Using perturbation theory, we use the $8 \times 8$ Hamiltonian including spin-orbit interaction and external magnetic field. Solution of this matrix gives the eigen-energies, which helps us to have more exact view of dispersion relation of our samples. It provides a way to evaluate energy dependency of g-factor and non-parabolicity study in the conduction bands. Definition and theoretical view of the spin relaxation mechanisms and the regimes of their domination are also considered in this chapter.

The experimental setup and sample characteristics are presented in chapter 3. We have taken advantage of using time-resolved pump-probe experiment by using a Ti:sapphire ultrafast laser to study spin dynamics. Some previous results related to our samples, which are useful for our discussion, have also been presented in this chapter.

Chapter 4 illustrates the results of spin life time measurements for our samples. We have measured the time evolution of spin polarization in thick-film InAs, thick- and thin-films InSb, AlInSb QW samples, and PbSe and HgTe QD samples. Temperature dependency of spin life time is studied for the samples, and dominant relaxation mechanisms are examined.

The g-factor measurement is demonstrated in chapter 5. In the same experimental setup, by adding an external magnetic field in the plane of the samples, time-resolved spin precession is studied. From this oscillatory behaviour g-factor can be measured at
different temperatures. The comparison between experimental and theoretical \((k.p)\) results for temperature dependency of \(g\)-factor of bulk samples shows some discrepancies. We try to consider several possibilities, which might help to reduce this difference.

Chapter 6 represents a study of transitions between hydrogen like shallow donor impurity states (P, As) in silicon. Time-resolved pump-probe using free-electron laser is employed to evaluate lifetime and also the dominant decoherence mechanism, from population dynamics of the donor impurity states.

Finally, Chapter 7 represents the conclusions, and some suggestions for future work.

In addition, more information has been presented in Appendices:

Appendix A considers a brief development of \(k.p\) theory. In Appendix B, time independent perturbation theory is reviewed. Also, solutions of Hamiltonian equations in the case of presence of spin-orbit splitting and external magnetic field are mentioned. Appendix C shows the 8-band \(k.p\) matrices for non-zero magnetic field by using raising and lowering operators, with and without coupling terms. At last, Appendix D provides the Fortran program for the presented matrices, used for calculation of Landau levels, which provides a tool to investigate energy dependency of Lande \(g\)-factor.
2 Theoretical background

2.1 Theoretical models for semiconductor bandstructure and spin splittings

2.1.1 Introduction and motivation
The electrons in semiconductors can have energies only within certain bands, which each correspond to a large number of discrete quantum states of the electrons. The microscopic behaviour of electrons in a solid is specified by electronic bandstructure. The band structure describes ranges of energy that an electron is forbidden or allowed to have. The periodic nature and symmetrical properties of crystals have interesting effects on the band structures of these materials theoretically. An interesting feature of the III-V compounds is a bulk inversion asymmetry, which results in spin-orbit coupling effects such as spin splitting at non-zero wave vector $k$ even without an applied magnetic field. It is interesting to investigate how this spin splitting behaves in the presence of an external magnetic field $B$. One can ask how the Zeeman spin splitting caused by $B$ combines with the spin splitting caused by inversion asymmetry. These studies provide information on the location and symmetry of the band edge extrema or critical points in the Brillouin Zone (BZ), the effective mass, and g-factor at and in the vicinity of the critical points, and the energy gaps between the band extrema. Of primary interest in this thesis is the description of spin splittings, both with and without magnetic field.

Solution of energy-eigenvalue problem of a semiconductor crystal gives the band structure of the semiconductor. Details of subband energy alignments are important factors in different applications. Conduction and valence subband energy levels are calculated based on three general approaches: $k.p$ [Luttinger (1955a)], Pseudopotential [Wang (1996)], Tight-binding [Vogl (1983)]. The techniques called ‘ab-initio’ have been used to calculate many properties of solids, including semiconductors; calculates the total energy including interactions between electrons and with the nuclei [Shukla (1999)]. These are good at predicting chemical trends and electronic states over the whole BZ, but require significant computational effort.

The small portion of band structure of a material, in the centre of the first BZ known as the $\Gamma$ point, with $k_x = k_y = k_z = 0$, where the lowest points in the conduction band
and the highest points in the valence band are found can determine several important physical characteristics, in particular its magnetic, electronic and optical properties. The \( k.p \) technique focuses on this high symmetry point (or indeed others), and with a small number of experimentally determined parameters can produce an accurate description of the electronic states near this symmetry point with minimal computing effort. Broadly speaking, the energy states at non-zero \( k \) are expressed using perturbation theory in terms of the basis set of states at zero \( k \). The simplest approximation is the effective mass approximation:

\[
E_{n}^{(k)} = E_{n}^{(0)} + \frac{\hbar^2 k^2}{2m^*_n} \quad \text{Eqn 2.1}
\]

Many compound semiconductors made from Group III and Group V elements such as GaP, AlAs, InSb etc all have a common crystal symmetry (the ZnS or "zinc blende" structure) and have their Fermi energy in the gap between a band arising from hybridised bonding p-states (labelled by \( |X\rangle, |Y\rangle, \) and \( |Z\rangle \)), which forms the valence band and anti-bonding s-state \( |S\rangle \), which forms the conduction band. The bandstructure of a zinc-blende semiconductor near the fundamental gap is illustrated in Figure 2.1. It can be seen that each band has a different curvature, and to a first approximation they appear to follow a simple parabolic dependence like free particles each with a different effective mass. It can also be seen that the valence band is split, and this is due to the spin-orbit interaction. It is well known that p-orbitals, which have non-zero orbital angular momentum, experience spin-orbit coupling effects. It is the purpose of \( k.p \) theory to improve upon this simple parabolic approximation of Eqn 2.1.

The \( k.p \) method will also be used here for description of the effects of applied magnetic fields on Zinc-Blend crystal semiconductors, and the rest of this chapter will describe in detail this method. Having obtained accurate descriptions of the energy levels we shall summarize these results comparison with the parabolic approximation. This will lead us to effective masses and g-values that are no longer constant and isotropic. Having Landau energies for a given field orientation, we can identify a field-dependent effective mass, \( m^* \), from \( E(n+1,\pm) - E(n\pm) = \frac{\epsilon h B}{m^*} \), where \( \pm \) shows spin directions. Also, one can calculate the effective Lande factor \( g^* \) as a
function of energy for various field directions. The g value is defined by the relation of \( E(n,+) - E(n,-) = g \* \mu_B B \) (\( n \) is the Landau index).

![Figure 2.1 Schematic bandstructure of direct-gap zinc-blend semiconductor around \( \Gamma \) point (three-band model)](image)

Recently the description of spin splitting, including zero-magnetic field splitting due to the Rashba [Rashba (1960)] and Dresselhaus [Dresselhaus (1955a,b)] effects not normally included in \( k.p \) models, has become highly topical due to interest in semiconductor spintronic devices. Knowledge of the spin-splitting is also necessary for calculation of spin relaxation times [D'yakonov (1974), Boggess (2000), Lau (2001)]. It is therefore important to find the experimental values of effective Lande factor \( g^* \) to test the models and provide a method for evaluation of the band structure parameters. Measurement of these effects in NGSs is of particular interest due to the much stronger interaction between bands and much stronger spin-orbit coupling.

### 2.1.2 \( k.p \) theory – the effective mass tensor and Kane's momentum matrix element

The energy Hamiltonian for an electron in a crystal is:

\[
\left[ \frac{p^2}{2m_0} + V(r) \right] \psi_{nk}(\mathbf{r}) = E_n(\mathbf{k}) \psi_{nk}(\mathbf{r})
\]

Eqn 2.2
where $m_0$ is free electron mass, $p$ is the operator $(-i\hbar \nabla )$, $V(r)$ is periodic potential of the crystal, $n$ and $k$ show the index and wave vector numbers, respectively. Bloch theorem states that the solution of Schrodinger equation for the periodic lattice is of the form:

$$\psi_{nk}(r) = e^{ik \cdot r} u_{nk}(r)$$  \hspace{1cm} \text{Eqn 2.3}

where $\psi_{nk}(r)$ is Bloch wave-function, and $u_{nk}(r)$ is cell periodic function, which has the same translational symmetry as the lattice. Substituting $\psi_{nk}(r)$ in Eqn 2.2 we achieve:

$$\left[ \frac{p^2}{2m_0} + \frac{\hbar^2 k^2}{2m_0} + V(r) \right] u_{nk}(r) = E_n(k) u_{nk}(r)$$

or

$$\left[ H_0 + \frac{\hbar^2 k^2}{2m_0} \right] u_{nk}(r) = E_n(k) u_{nk}(r)$$  \hspace{1cm} \text{Eqn 2.4}

where the $k$-independent terms have been collected together in $H_0 = \frac{p^2}{2m_0} + V(r)$. Assume that we have solved the Schrodinger equation, $H_0 u_{n0} = E_n^{(0)} u_{n0}$, for $H_0$ and obtained the eigen-functions $u_{n0}$, and eigen-values $E_n^{(0)}$. We have:

$$u_{nk}(r) = \sum_m f_{n,m}(k) u_{m,0}(r)$$  \hspace{1cm} \text{Eqn 2.5}

where $u_{m,0}(r)$ form an orthonormal set:

$$\int_{\text{cell}} u_{n0}^* (r) u_{m0}(r) \, d^3 r = \delta_{nm},$$

with $n$, $m$ as band indices, and $\delta_{nm}$ is the Kronecker delta function.

Substituting Eqn 2.5 into Eqn 2.3 gives:

$$\psi_{nk}(r) = e^{ik \cdot r} \sum_m f_{n,m}(k) u_{m,0}(r)$$  \hspace{1cm} \text{Eqn 2.6}

Substituting Eqn 2.5 in Eqn 2.4 multiplying by $u_{n,0}^*$ and integrating over a unit cell, we obtain:
where $p_{n,m}$ is momentum matrix element:

$$p_{n,m} = \int \hat{u}_{n,0}^*(r) \hat{p} \hat{u}_{m,0}(r) \, d^3 r$$  \hspace{1cm} \text{Eqn 2.8}

Therefore, we include the effect of the non-diagonal term in Eqn 2.7, $\frac{\hbar}{m_0} k \cdot p_{n,m}$, using perturbation theory. The perturbed eigen-state is:

$$u_{nk}(r) = u_{n0}(r) + \left[ \sum_{n=m} \frac{\hbar}{m_0} \frac{k \cdot p_{n,m}}{E_n(0) - E_m(0)} \right] u_{m0}(r)  \hspace{1cm} \text{Eqn 2.9}$$

To second order the energy eigen-value is:

$$E_n^{(k)} = E_n^{(0)} + \frac{\hbar^2 k^2}{2m_0} + \frac{\hbar}{m_0} k \cdot p_{n,m} + \frac{\hbar^2}{m_0^2} \sum_{n \neq m} \frac{|k \cdot p_{n,m}|^2}{E_n(0) - E_m(0)}  \hspace{1cm} \text{Eqn 2.10}$$

If the symmetry point of interest has only band extrema (usually assumed to be the case at the $\Gamma$-point in ZnS structure crystals of interest), then the third term which is linear in $k$ must vanish, meaning that $p_{n,m} = 0$, and this is why we need the second order term.

This result can be written in terms of effective mass approximation:

$$E_n^{(k)} = E_n^{(0)} + \frac{\hbar^2}{2} \sum_{\alpha, \beta} D^{\alpha \beta} k_\alpha k_\beta  \hspace{1cm} \text{Eqn 2.11}$$

Where

$$D_n^{\alpha \beta} = \left( \frac{1}{m_n^*} \right)_{\alpha \beta} = \frac{1}{m_0} \left[ \delta_{\alpha \beta} + \frac{1}{m_0} \sum_{n \neq m} \frac{P^\beta_{n\alpha} P^\alpha_{m\beta}}{E_n(0) - E_m(0)} \right]  \hspace{1cm} \text{Eqn 2.12}$$
is inverse effective mass tensor, and $\alpha$ and $\beta$ are the coordinate indices 1,2,3. If the effective mass tensor is diagonal, then the relation between energy and momentum is in the form of $E_n^{(k)} = E_n^{(0)} + \hbar^2 k^2 / 2m_n^*$ and we have recovered the simple approximation of a parabolic energy-dispersion Eqn 2.1.

In the simplest approximation, therefore, electrons and holes in a crystal respond to electric fields as if they were free particles in a vacuum, but with a different mass, called the effective mass, which is usually stated in units of the ordinary mass of an electron $m_0$ ($9.11 \times 10^{-31}$ kg). The deviation from the free-electron mass originates from the interaction with the other bands.

Note at this stage we have a matrix model, Eqn 2.7, that requires knowledge of a number of independent parameters, $E_n^{(0)}$ and $p_{p_{n,m}}^{x,y,z}$, which must be found from experiment. The number of these parameters may be quite large, even for a small number of bands included in the summation. Kane showed that group theory can be used to show that many elements are zero (e.g. we just showed $p_{n,n} = 0$), and that many elements are equal. The most important matrix element of $p$ is that between the conduction bands-like state and the valence band $p$-states [Kane (1982)]:

$$p_{cv} = \langle X | p_x | S \rangle = \langle Y | p_y | S \rangle = \langle Z | p_z | S \rangle$$

Eqn 2.13

and ignoring all other interactions the effective mass equation for the conduction band reduces to:

$$\frac{1}{m_e^*} = \frac{1}{m_0} \left[ 1 + \frac{2E_p}{3E_g} \right]$$

Eqn 2.14

where $E_p = 2P^2/m_0 = 2m_0P^2/\hbar^2$ is the interaction energy [Rössler (1984)] and $P$ is Kane's parameter. It suggests that the effective mass should scale linearly with energy gap if $E_p \gg E_g$, and this turns out to be the case for most ZnS-structure materials, for which $E_p \sim 20$eV. Figure 2.2 compares examples of conduction band effective masses from experiment and $k.p$ theory (straight line).
Figure 2.2 Conduction band effective masses from experiment (points) and k·p theory (straight line). In practice, \( E_p \) is different for each material. Taken from 1

2.1.3 Spin-orbit interaction

So far we have ignored the spin-orbit interaction, which is a relativistic effect that contributes to the energy. When electrons move around the positively charged nucleus, the electric field of the nucleus transformed into the rest-frame of the electron, appears as a magnetic field seen by the electrons, Figure 2.3. The electron spin magnetic moment senses this field, and the electron energy is therefore dependent on the alignment of the spin and orbital angular momentum vectors. Spin-orbit coupling may therefore be said to occur because of the interaction between electron spin with angular momentum of the movable nucleus, and the interaction energy is \( H_{so} = \lambda L \cdot S \), where the orbital angular momentum, \( L = r \times p \), and \( \lambda \) is constant. The eigen-functions of \( H_{so} \) are also eigen-states of the total angular momentum, \( J = L + S \), and its z component \( J_z \). Therefore states with different \( J \) have different Spin-orbit interaction energy. Total angular momentum \( J \) varies from \( L + S \) to \( |L - S| \). For an s-state, \( L = 0 \) and \( S = 1/2 \) there is only one possibility is \( J = 1/2 \). The z-component of \( J \), \( m_J = -1/2 \) and \( 1/2 \), i.e. spin up and spin down. For a p-orbital,

1 http://www.nd.edu/~djene/kdotp.pdf
L = 1 and S = 1/2 then there are two possibilities for J are 3/2 and 1/2. The two possibilities have different angles between L and S and so different spin-orbit interaction energies. We note that for J=3/2, z-component of J is $m_J = -3/2, -1/2, 1/2, 3/2$ i.e. four states, and for J=1/2 there are two possible values of $m_J = -1/2, 1/2$.

**Figure 2.3** The relative motion of a nucleus in a circular orbital about the electron generates a magnetic field resulting in Spin-orbit splitting.

The bonding p orbital that makes up the valence band has L=1, so that just as for the atomic case, we have a J=3/2 and J=1/2 state that are split by the spin-orbit coupling separated by the spin-orbit splitting energy, $\Lambda_0$. The J=3/2 multiplet is doubly degenerate at k=0 (4 states including spin), known as the HH and LH bands, and the J=1/2 band is known as SO state.

The magnitude of $\Lambda$ is more for crystals whose constituent atoms have higher atomic number. In fact, the spin-orbit splitting energy of semiconductors increases as the fourth power of the atomic number of the constituent elements, $Z^4$, Figure 2.4.
2.1.3.1 Spin-orbit Hamiltonian

To calculate the Hamiltonian of the spin-orbit interaction we note that when electrons move around the positively charged nucleus at relativistic speeds, the electric field of the nucleus Lorentz-transforms to a magnetic field seen by the electrons. The transformation is given by: \[ \vec{B}' = -(\vec{v} \times \vec{E}) / (2c^2 \sqrt{1 - v^2 / c^2}) \approx -(\vec{v} \times \vec{E}) / 2c^2 \] (for \( v \ll c \)). The potential energy of interaction of spin with total magnetic field is \(-\mu_s \cdot B\), where the magnetic dipole moment is \( \mu_s = (-e/m_0)S \), and \( S = (\hbar / 2)\sigma \). The magnetic induction is \( B = B_{\text{ext}} - B' = (\vec{v} \times \vec{E}) / 2c^2 \), \( B_{\text{ext}} = 0 \), and \( \vec{v} = \vec{p} / m_0 \). For an electron moving in the periodic potential \( E = -V V(r) / e \), so the spin-orbit Hamiltonian can be shown by:

\[
H_{so} = \frac{\hbar}{4m_0 c^2} (VV \times \vec{p}) \cdot \sigma
\]

Eqn 2.15

where \( V \) is the periodic lattice potential and \( \sigma \)s are the Pauli spin matrices:

\[\text{Figure 2.4 The spin-orbit splitting energy for different semiconductors plotted against the average atomic number } Z_{av}; \text{ the dashed line shows a rough fit to a } Z^4 \text{ polynomial. Taken from }^2.\]

\[\text{http://www.nd.edu/~djena/kdotp.pdf}\]
\[ \sigma_x = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}, \quad \sigma_y = \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix}, \quad \sigma_z = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}; \]

And the eigen-states of \( \sigma_x \) are

\[ \text{spin } \uparrow = \begin{bmatrix} 1 \\ 0 \end{bmatrix}, \quad \text{spin } \downarrow = \begin{bmatrix} 0 \\ 1 \end{bmatrix}. \]

Operating Eqn 2.15 on the Bloch wave-function gives:

\[
H_{so} + H_{kso} = \frac{\hbar}{4m_0^2c^2} (\sigma \times \nabla \psi) \cdot p + \frac{\hbar^2}{4m_0^2c^2} (\sigma \times \nabla \psi) \cdot k \tag{Eqn 2.16}
\]

where \( H_{kso} \) is the \( k \)-dependent spin-orbit term, which near \( k=0 \) is very small compared with \( H_{so} \) and is neglected. The reason is that the main contribution to the spin-orbit interaction comes from the core region of the atom where \( \nabla \psi \) and \( p \) are very large; \( k \) is small compared to \( p \). For more information about solution of the Hamiltonian including spin-orbit interaction, refer to Appendix B.

### 2.1.4 k.p calculation in zero magnetic field

As already mentioned, the eigen-energies problem (Eqn 2.7) requires first that we find the matrix elements \( p_{n,m} \), which in principle is an infinite set. We can restrict ourselves to states close in energy to \( E_n^{(0)} \) so that the denominator in the third term is small, and secondly we can use group theoretical arguments to reduce the number of independent elements of \( H' \). In the discussion that follows we shall show that there are a small number of states and a small number of matrix elements that are required for accurate description of most of the important electronic properties of the technologically useful semiconductors. The states of interest are all near to the band edge, so the momentum \( k \) is small and consequently terms that scale with momentum can be treated as the small perturbation.
Table 2.1 Kane basis set at Γ point [001], using phase convention of Weiler [Weiler (1981)]
(with correction of spin directions in the last one).

| $u_n$ | $|J, m_J\rangle$ | $|\psi_{J, m_J}\rangle$ | $E_n(k=0)$ |
|-------|------------------|-----------------|-------------|
| $a$-set: |
| $u_1$ | C↑ | $|\frac{1}{2}, \frac{1}{2}\rangle$ | $|S\uparrow\rangle$ | $E_g$ |
| $u_3$ | HH↑ | $|\frac{3}{2}, \frac{3}{2}\rangle$ | $-\left(\frac{i}{\sqrt{2}}\right)(X + iY)\uparrow$ | 0 |
| $u_5$ | LH↓ | $|\frac{3}{2}, -\frac{1}{2}\rangle$ | $\left(\frac{i}{\sqrt{6}}\right)(X - iY)\uparrow + Z\downarrow$ | 0 |
| $u_7$ | SO↓ | $|\frac{1}{2}, -\frac{1}{2}\rangle$ | $-\left(\frac{i}{\sqrt{3}}\right)(X - iY)\uparrow - Z\downarrow$ | $-\Delta$ |
| $b$-set: |
| $u_2$ | C↓ | $|\frac{1}{2}, -\frac{1}{2}\rangle$ | $|S\downarrow\rangle$ | $E_g$ |
| $u_6$ | LH↑ | $|\frac{1}{2}, \frac{1}{2}\rangle$ | $-\left(\frac{i}{\sqrt{6}}\right)(X + iY)\downarrow - 2Z\uparrow$ | 0 |
| $u_4$ | HH↓ | $|\frac{3}{2}, \frac{3}{2}\rangle$ | $-\left(\frac{i}{\sqrt{2}}\right)(X - iY)\downarrow$ | 0 |
| $u_8$ | SO↑ | $|\frac{1}{2}, \frac{1}{2}\rangle$ | $-\left(\frac{i}{\sqrt{3}}\right)(X + iY)\downarrow + Z\uparrow$ | $-\Delta$ |

For semiconductors where the energy separation between the conduction and valence bands is small, the four bands CB, HH, LH, SO are usually included and treated exactly (first by Kane [Kane (1957)]). This is often referred to as the 4-band model, or the 8-band model (because there are 8 states if spin is included), and sometimes also to the “three level model” because at $k=0$ two of the bands coalesce into one (here, we have used three-band model and 8×8 matrix). Kane showed that if the states of interest include only $S$, $X$, $Y$ and $Z$, then the basis shown in Table 2.1 will diagonalise the spin-orbit coupling, without seriously complicating the $k.p$ terms.
Kane treated the interactions of higher bands with this set by second order perturbation theory, and the refinement of this model to include the higher bands before diagonalisation and also magnetic fields is due to Luttinger and Kohn [Luttinger (1955b)] and Pidgeon and Brown (PB) [Pidgeon (1966)].

The complete 8x8 Pidgeon-Brown matrix Hamiltonian as a part of the block diagonal (coming later, see Figure 2.6) including the band-edge energy $E_g$ and $\Delta$ relative to the $\Gamma_8$ valence band is given in Table C1 (Appendix C), which also contains the momentum matrix element parameters for higher bands such as $\gamma_1$ and $\kappa$ achieved from parameters primarily identified by Luttinger [Luttinger (1956)] shown by superscript L:

$$
\gamma_1^L = \frac{1}{3} \left( \frac{E_p}{E_g} \right) + \gamma_1, \quad \gamma_2^L = \gamma_2 + \frac{1}{6} \frac{E_p}{E_g}; \\
\gamma' = \frac{1}{6} \left( \frac{E_p}{E_g} \right) + \gamma', \quad \gamma'' = \frac{1}{6} \left( \frac{E_p}{E_g} \right) + \gamma''; \\
\gamma'''' = \gamma + \frac{1}{6} \frac{E_p}{E_g}, \quad \kappa^L = \frac{1}{6} \left( \frac{E_p}{E_g} \right) + \kappa.
$$

Eqn 2.17

where $\gamma_1^L, \gamma_2^L, \gamma_3^L$, and $\kappa^L$ are the valence band parameters defined by Luttinger [Luttinger (1956)]. The $\Gamma_6 \times \Gamma_8$ parameters $\gamma_1, \gamma_2, \gamma_3, \kappa$, and $q$ were shown by Roth et al. [Roth (1959)] to involve the split-off band $\Gamma_7$. The $\Gamma_7 \times \Gamma_7$ and $\Gamma_7 \times \Gamma_8$ parameters are denoted by $\gamma'_1, \gamma'_2, \gamma'_3, \kappa', \kappa''$. The conduction band $\Gamma_6$ effective mass parameter $\mu$, the linear-k parameter $c$ for $\Gamma_8$, and the $\Gamma_6 \times \Gamma_8$ parameters $P$ and $G$ were defined by Dresselhaus [Dresselhaus (1955a)], Kane [Kane (1957)], and Dresselhaus et al. [Dresselhaus (1955b)]; the $\Gamma_6 \times \Gamma_7$ and $\Gamma_7 \times \Gamma_8$ parameters are denoted by $C', P'$, and $G'$. Weiler [Weiler (1981)] obtained three other parameters: $N_1, N_2, N_3$. $N_1$ contributes to the conduction band g-factor, similar to $\kappa$ in the valence bands; $N_2$ and $N_3$ represent additional couplings between the $\Gamma_6$ conduction band and the $\Gamma_8$ valence bands. These new parameters like $q$, arise from the spin-orbit splitting of higher bands. The parameters $q$ and $\mu = (\gamma_3 - \gamma_2)/2$ are called Ge warping parameters because, if they were zero in a material with inversion symmetry, the energy bands would be isotropic (independent of the direction of $k$). The parameters
$C, G, N_2$ and $N_3$ are called the inversion asymmetry parameters because they are non-zero in zinc-blend materials without inversion symmetry. The parameters $q, C, G, N_1, N_2$, and $N_3$ are small in most materials of interest and usually neglected. It is also usually assumed that $\kappa = \kappa' = \kappa'', \mu = \mu', \gamma_i = \gamma_i'$, $C = C'$, $P = P'$ and $G = G'$ in order to further reduce the total number of parameters.

Terms in the Hamiltonian contain the following operators:

\[
\begin{align*}
\mathbf{F}_1^+ &= \mathbf{k}_x^+ + \mathbf{k}_y^+ + \mathbf{k}_z^+, \\
\mathbf{F}_2^+ &= \mathbf{k}_x^+ - \mathbf{k}_y^+ - \mathbf{k}_z^+, \\
\mathbf{F}_3^+ &= \sqrt{3} (\mathbf{k}_x^2 - \mathbf{k}_y^2), \\
\mathbf{F}_4^+ &= \{\mathbf{k}_x, \mathbf{k}_y\} = (\mathbf{k}_x^+ \mathbf{k}_y^+ + \mathbf{k}_y^+ \mathbf{k}_x^+), \\
\mathbf{F}_4^- &= \{\mathbf{k}_x, \mathbf{k}_y\}, \\
\mathbf{H}_z &= i [\mathbf{k}_x, \mathbf{k}_y], \\
\mathbf{H}^+ &= \pm [\mathbf{k}^+, \mathbf{k}_z].
\end{align*}
\]

Eqn 2.18

Some of these operators are zero in the absence of magnetic field, but are included here simply to avoid writing the matrix out again later when we include the field.

We stress that the matrix of Table C1 is an operator matrix, and that $\mathbf{k}^2$ etc are all operators. However, in the zero-magnetic field case it is trivial to convert this into an algebraic matrix because the Bloch functions are eigen-functions of these operators, i.e. operation on a Bloch function does not change $\mathbf{k}$, and states of different $\mathbf{k}$ are not coupled. Any given state $|n, \mathbf{k}\rangle$ is only coupled within a block of $8 \times 8$ whose corresponding matrix can be diagonalised using a straightforward numerical process for any choice of vector $\mathbf{k}$, so long as the momentum parameters and band-edge energies are known. We shall see that this process is not so simple in non-zero field (and similarly it would also pose difficulties in non-zero electric field, though this case does not concern us here).
2.1.5 The five-band $k.p$ (14×14 matrix)

Another development can be achieved by going from the three-level model to the five-level model (14×14 Hamiltonian). It includes the $\Gamma_7^\sigma$, $\Gamma_8^\sigma$, $\Gamma_6^\sigma$, $\Gamma_7^\sigma$, $\Gamma_8^\sigma$ levels in the $k.p$ description, plus including the contributions from far levels, to account for the experimentally observed anisotropy of the conduction band for example in GaAs as a medium-gap material [Weisbuch (1977), Pfeffer (1990), Pfeffer (1996)]. We note here in passing that for semiconductors where the band gap is large and comparable with the separation of the higher bands it becomes necessary to include the higher bands in the set that will be diagonalised exactly. This next higher band is due to the anti-bonding p-orbitals, and looks very similar to the valence band multiplet. This model may be called the 7 band model, the 14 band model or sometimes the five level model (here, we have used five-band model, 14×14 matrix).

The non-sphericity is related to the $Q$ terms (momentum matrix elements between valence band and higher conduction band) and the difference between $\gamma_2$ and $\gamma_3$ values (Luttinger parameters) [Luttinger (1956)]. For $Q = 0$ the energies do not depend on the direction of magnetic field i.e. it results in a spherical conduction band. The spin splitting, which is in general due to the lack of inversion symmetry in zinc-blend crystals, is related to the nonzero values of $E_p$, $E_p'$, $Q$ (momentum matrix elements) and $\Delta^-$ (interband spin-orbit coupling between $(\Gamma_8^\sigma, \Gamma_7^\sigma)$ and $(\Gamma_8^\sigma, \Gamma_7^\sigma)$ multiples). For comparison with the three-band model (8×8), the bandstructure of a Zinc-Blend semiconductor near the fundamental gap for the five-band model (14×14) is illustrated in Figure 2.5.
Figure 2.5 Five-level model (14×14 model) for bandstructure of GaAs-type semiconductors near the Γ point of BZ. Energy gaps, interband matrix elements of momentums, and spin-orbit interactions along with far-band contributions are indicated.

2.1.6 Magnetic field effects

We will consider the effect of an externally applied magnetic field on the electron motion. Landau showed that in a uniform magnetic field, \( B \), the motion of an electron in the plane perpendicular to \( B \) becomes quantized. The three-dimensional dispersion is split into a series of one-dimensional subbands. These magnetic subbands are known as Landau levels, shown by index \( n \).

In addition to the effect on the orbital motion, an external magnetic field produces an effect on the magnetic dipole due to the electron spin. It produces the magnetic potential energy as \( U = -\mathbf{\mu} \cdot \mathbf{B} \), where the magnetic dipole moment (related to the orbital angular momentum) is given by \( \mathbf{\mu}_L = -eL/2m_0 \). Assuming a magnetic field in the z-direction and quantization of angular momentum, we find the equally spaced energy levels by \( U = eL_z B / 2m_0 = m_0 e\hbar B / 2m_0 = m_1 \mu_B B \), where the Bohr magneton is \( \mu_B = e\hbar / 2m_0 = 9.2740154 \times 10^{-24} \text{ J} / \text{T} = 5.788382 \times 10^{-5} \text{ eV} / \text{T} \). This displacement of the energy levels from the zero field level gives the uniformly spaced multiplet splitting of the spectral lines which is called the Zeeman effect. After discovering the
spin of electron by Goudsmit and Uhlenbeck, it was found that the magnetic field also interacts with the electron spin magnetic moment, \( \mathbf{\mu}_s = -\gamma e S / 2m_0 \), where \( \gamma \) is approximately 2 (for free electron). This factor is called the spin \( \gamma \)-factor or gyromagnetic ratio. The \( \mathbf{S} \) and \( \mathbf{L} \) vectors are both precessing around the magnetic field and are not in general in the same direction. The resulting geometric factor \( g_L \) or simply \( g^* \) is called the Lande \( g \)-factor, which allows the resultant splitting of the spectral lines in terms of quantum number \( m_j \) is expressed, where \( m_j \) is the \( z \)-component of the total angular momentum.

Combining the Landau and Zeeman effects, the energy states of the electron in a magnetic field, in the parabolic effective mass approximation is given by:

\[
E^\pm_{n,k}(B) = E_n + \frac{\hbar^2 k_z^2}{2m^*} + (n + \frac{1}{2}) \frac{\hbar e B}{m^*} \pm \frac{1}{2} g^* \mu_B B
\]

Eqn 2.19

where \( k_z \) is the component of momentum parallel to the magnetic field \( B \), and the quantum numbers \( \pm \) refer to spin. In the parabolic approximation, the effective mass and \( g \)-value, \( m^* \) and \( g^* \), are constants and independent of \( n, k_z, B \).

The Hamiltonian for a charged particle moving in a transverse magnetic field can be written as \( H_B = (p - eA)^2 / 2m \), (\( A \) is the vector potential of \( B \)); or in terms of the ladder operators can be rewritten as \( H_B = \hbar \omega_c \sigma^+ \sigma^+ + 1/2 \), which has eigen-values of \( E_n = \hbar \omega_c (n + 1/2) \) or Landau energies with \( \omega_c = eB / m \), the cyclotron frequency. It is similar to the harmonic oscillator, each associated with a highly degenerate set of eigen-vectors, known as Landau levels.

The initial one-electron eigen-value problem with spin-orbit interaction in the presence of an external magnetic field gives:

\[
\left[ \frac{\mathbf{p}^2}{2m_0} + V(\mathbf{r}) + \frac{\hbar}{4m_0^2 c^2} (\mathbf{\sigma} \times \nabla V) \cdot \mathbf{p} + \mu_B \mathbf{B} \cdot \mathbf{\sigma} \right] \Psi = E \Psi \]

Eqn 2.20
where \( \mathcal{V} \) is the crystal potential, \( \mathbf{p} \rightarrow \mathbf{p} - eA(r) \) is the kinetic momentum. In a similar fashion to the zero-B case (see Appendix B), we look for solutions in the form of:

\[
\psi_{l,k_z}(r) = \exp(ik_z z) \sum_{n,m} \phi_n(x, y, z) f_{l,m}^n(k_z) u_{n,m}(r),
\]

Eqn 2.21

where \( \phi_n \) are the harmonic oscillator functions and \( f_{l,m}^n \) are numerical coefficients.

Note that whereas in previous sections (Eqn 2.1 - Eqn 2.12) \( n \) was used as the band index, we now use \( n \) as the Landau quantum number and \( l \) as the band index. Also note that the sum is now running over the infinite set of Landau quantum numbers \( n \) as well as the 8 bands \( m \).

For a magnetic field \( B \) in an arbitrary direction identified by the spherical polar angles \( \theta \) and \( \phi \), it is possible to make a coordinate transformation. In this thesis we are only interested in experiments with \( B \) along [001], therefore \( \theta = \phi = 0 \), \( k_x = k_1 \), \( k_y = k_2 \), and \( k_z = k_3 \).

\[
k_x = (\frac{1}{\lambda} \sqrt{2}) (a + a^+) \quad \text{and} \quad k_y = (\frac{1}{\lambda} \sqrt{2}) ((a - a^+) \quad \text{Eqn 2.22}
\]

where \( \lambda = (c/eB)^{1/2} = s^{-1/2} \), \( \lambda \) is the Landau radius, and \( s \) is the free-electron cyclotron energy, \( \hbar k_z \) is the component of the momentum along the direction of the applied magnetic field, and \( a^+ \), \( a \) are raising and lowering operators for harmonic oscillator functions \( \phi_n \):

\[
a^+ \phi_n = \sqrt{n+1} \phi_{n+1}, \quad a \phi_n = \sqrt{n} \phi_{n-1}
\]

Eqn 2.23

Note that it follows that

\[
N \phi_n = a^+ a \phi_n = n \phi_n
\]

and

\[
[a, a^+] = a a^+ - a^+ a = 1.
\]

Eqn 2.24

where \( N \) is the number operator.

The \( 8 \times 8 \) operator Hamiltonian after substitution of Eqn 2.22 is shown in Table C2. Unlike the zero-field case where states of different \( k \) do not interact so that the only
couplings are within sets of $8 \times 8$, it now becomes clear that the operator matrix of Table C2 has a mix of raising and lowering operator terms and therefore states of different $\phi_n$ do interact. In other words, because the trial solutions are not eigenfunctions of the operators in the matrix the basis states of the system are all coupled. Writing out the determinental form of the Hamiltonian for the infinite basis-set of $\phi_n u_m$ does not lead to a block diagonal, and therefore it is not possible to simply rewrite Table C2 in algebraic $8 \times 8$ determinental form as before.

However, by judicious choice of the order of the basis functions, and by neglection of small terms proportional to $C, G, q, N_1, N_2, N_3$, and $\mu$, it is possible to produce a block diagonal form (see below), that can be diagonalised; (although, note that $\mu$ is not in fact small for materials of interest here). Instead of grouping together states all having the same $\phi_n$, we group together the following sets of 8:

$$
|\psi_{n,k}\rangle = \begin{bmatrix}
  f_1^n \phi_n u_1 \\
  f_2^n \phi_{n-1} u_2 \\
  f_3^n \phi_{n+1} u_3 \\
  f_4^n \phi_{n+1} u_7 \\
  f_5^n \phi_{n+1} u_2 \\
  f_6^n \phi_{n+2} u_6 \\
  f_7^n \phi_{n+2} u_4 \\
  f_8^n \phi_{n+2} u_8 \\
\end{bmatrix} \exp(i k_z z) \quad \text{Eqn. 2.25}
$$

Because $\phi_n = 0$ for $n < 0$, with $n \geq -2$, we have

$$
f_1^{-2} = f_2^{-2} = f_3^{-2} = f_4^{-2} = f_5^{-2} = f_6^{-2} = f_7^{-2} = f_8^{-2} = f_1^{-1} = f_5^{-1} = f_6^{-1} = f_8^{-1} = \quad \text{Eqn. 2.26}
$$

For $n \geq 1$ there are eight independent solutions for each $n$. They are denoted, in order of decreasing energy, conduction band (if $f_1^n$ or $f_2^n$ is large), heavy hole (if $f_3^n$ or $f_4^n$ is large), light hole (if $f_5^n$ or $f_6^n$ is large), and split-off band (if $f_7^n$ or $f_8^n$ is large).
Now we get a Hamiltonian which can be divided into blocks [Cohen (1988), Trebin (1979)] although the first few blocks are smaller than $8 \times 8$ because of Eqn 2.26: $H_{11}=1 \times 1$, $H_{22}=4 \times 4$, $H_{33}=7 \times 7$, and $H_{44}=H_{55}=H_{66}=\cdots=H_{nn}=8 \times 8$, see Figure 2.6. The off-diagonal matrices, which are coupling terms among the diagonal blocks, now contain only terms proportional to $C$, $G$, $q$, $N_1$, $N_2$, $N_3$, and $\mu$, and may be neglected because most of the coupling terms are small. For preventing enormous complexity in the calculations, they are usually ignored.

![Figure 2.6 Schematic of a block diagonal](image)

Reintroducing the coupling terms introduces other elements that mean the infinite matrix is no longer block-diagonal, and renders the problem much more difficult (see Appendix C, Tables C1, C2).

Table C3 is in energy units since $E_e$, $\Lambda$, $E_p$, and $s$ have the dimensions of energy and $F$, $\kappa$, $N_1$, and $\gamma$'s are dimensionless. The Hamiltonian of Table C1 is equal to those of Pidgeon and Brown [Pidgeon (1966)] and of Aggarwal [Aggarwal (1972)], except for corrected sign errors and when account is taken of different basis sets [Weiler (1981)]. The matrix is Hermitian and invariant under time reversal.
2.1.7 Approximate solutions to the k.p matrix eigen-problem

Aggarwal [Aggarwal (1972)], has given approximate solutions for the eigenvalues of the Hamiltonian equation. The solutions are exact for $B = k_z = 0$, i.e. band edge. The solutions of the 4×4 Pidgeon-Brown matrices (Table C3) for the a- and b-sets have the following form:

$$
\begin{vmatrix}
    sA - E & B\sqrt{s} & C\sqrt{s} & D\sqrt{s} \\
    B\sqrt{s} & E_0 - E & 0 & 0 \\
    C\sqrt{s} & 0 & E_0 - E & 0 \\
    D\sqrt{s} & 0 & 0 & G_0 - E
\end{vmatrix} = 0
$$

Eqn 2.27

where we have neglected the higher band interactions with the valence band (i.e. $\gamma_1^L$, $\gamma_2^L$ etc. all zero), $s$ is the free electron cyclotron energy, and $A$, $B$, $C$, $D$ are constants involving the band parameters and are different for each set. We are interested in the conduction band, so in this equation energies are measured relative to the conduction band edge $E_0 = -E_g$ and $G_0 = -E_g - \Delta$ are the energies of the valence band edge and the spin-orbit split-off band edge. Finding the determinant and rearranging leads immediately to:

$$
E = sA - \frac{(B^2 + C^2)s}{E_0 - E} - \frac{D^2s}{G_0 - E}
$$

Eqn 2.28

Substituting the values of $A$, $B$, $C$, $D$ for each set and replacing $E_a$ and $E_b$ with $\bar{E} \pm \delta E$ we find:

$$
\bar{E} + \delta E = s\left[(2F + 1)(n + \frac{1}{2}) + (N_1 + \frac{1}{2})\right] - \frac{\left(\frac{3}{2} E_p n + \frac{1}{6} E_p s\right)s}{E_0 - \bar{E} - \delta E} - \frac{\frac{1}{3} E_p (n + 1)s}{G_0 - \bar{E} - \delta E}
$$

Eqn 2.29

and

$$
\bar{E} - \delta E = s\left[(2F + 1)(n + \frac{1}{2}) - (N_1 + \frac{1}{2})\right] - \frac{\left(\frac{3}{2} E_p n + \frac{1}{6} E_p s\right)s}{E_0 - \bar{E} + \delta E} - \frac{\frac{1}{3} E_p n s}{G_0 - \bar{E} + \delta E}
$$

Eqn 2.30
The values of \( m^* \) and \( g^* \) are defined from Eqn 2.19 (with \( k_z = 0 \)), \( E_a - E_b = g^* \mu_B B \) and \( E_a + E_b = (2n+1) \hbar e B / m^* \). Neglecting \( 8E \) on the right hand sides of Eqn 2.29 and Eqn 2.30, the effective mass and Lande g-factor are:

\[
\frac{m^*}{m_0} = \frac{2E}{(2n+1)s} = 1 + \frac{E_p}{3} \left( \frac{2}{E_g + E} + \frac{1}{E_g + \Delta + E} \right) + 2F \quad \text{Eqn 2.31}
\]

\[
\frac{g^*}{g_0} = \frac{2sE}{s} = 1 - \frac{E_p}{3} \left( \frac{1}{E_g + E} - \frac{1}{E_g + \Delta + E} \right) + 2N_1 \quad \text{Eqn 2.32}
\]

where \( g_0 = 2 \). The band edge values of \( g^* \) and \( m^* \) are given by setting \( E = 0 \). It can be shown that the band edge formulas are exact, and may also be derived without the approximation of small energy and apart from \( F \) and \( N_1 \) are independent of other higher band parameters such as \( \gamma_1^L, \gamma_2^L \) etc.

Because Kane considers only the mutual coupling of the conduction and valence bands, therefore in the above expression the band parameters \( F \) and \( N_1 \), which represent the interaction of conduction band with other bands, would be zero in Kane's model.

The higher-band contributions \( F \) and \( N_1 \) were also further expanded from a 14-band model by Johnson and Dickey [Johnson (1970)]. The five-band model was also used by Hermann and Weisbuch [Hermann (1977)] for calculation of electron effective mass and g-factor at the band edge:

\[
\frac{m^*}{m_0} = 1 + \frac{E_p}{3} \left( \frac{2}{E_g + E} + \frac{1}{E_g + \Delta} \right) - \frac{E_p'}{3} \left( \frac{2}{E_\Gamma^+ - E_g} + \frac{1}{E_\Gamma^+ - E_g} \right) + C \quad \text{Eqn 2.33}
\]

\[
\frac{g^*}{g_0} = 1 - \frac{E_p}{3} \left( \frac{1}{E_g + E} - \frac{1}{E_g + \Delta} \right) - \frac{E_p'}{3} \left( \frac{-1}{E_\Gamma^+ - E_g} + \frac{1}{E_\Gamma^+ - E_g} \right) + C' \quad \text{Eqn 2.34}
\]

With a negative sign in front of \( E_p \) with other positive energies, in the g-factor equation; \( E(\Gamma^+_\Gamma) \) and \( B(\Gamma^+_\Gamma) \) denote the energies of the next higher conduction bands at the \( \Gamma \) point – see Figure 2.5. The energies \( E_p \) and \( E_p' \) describe the interaction of the first conduction band with the nearest valence and conduction band, respectively. \( C \)
and $C'$ are included as corrections to take into account the interaction with the upper conduction bands. With $E_p' = C = C' = 0$, the known three-band model formulas are achieved.

## 2.2 Spin dynamics theory

### 2.2.1 Introduction

Spin relaxation and spin dephasing are processes that lead to spin equilibration and are thus important for spintronics. For spintronics applications, relaxation time of the electron spins, $\tau$, must be sufficiently long to process information stored in the form of the polarization of spin ensembles. For the spin field effect transistor (spin-FET) [Datta (1990)] it is also essential for spintronics to be able to control spin dynamics with an external electric field. It has been shown that only in the case of the DP process is it possible to alter the spin lifetime with an applied electric field, by modulating the strength of the spin-orbit coupling through the Rashba effect [Zawadzki (2004)]. The EY process is not strongly affected by electric field. The control of the DP lifetime has also been demonstrated in some systems [Karimov (2003), Nitta (1997), Koga (2002), Hall (2005)]. It is therefore essential to be able to find regimes in which DP dominates, especially in NGSs, which have large predicted Rashba effect. In this thesis, time-resolved pump-probe experiments have been performed to investigate the spin lifetimes, to identify the regimes in which the different spin relaxation processes dominate, and to understand how to control them. For example, the influence of temperature on the spin relaxation and the importance of the different relaxation mechanisms are studied.

Typically, the magnitudes for spin lifetimes measured at low temperature are in the range 1-10 ns, obtained from spin resonance in degenerate n-type InSb at helium temperatures. At elevated temperatures the lifetimes can be of order a few picoseconds.

There are four major physical mechanisms responsible for spin equilibration: Elliot-Yafet (EY) [Elliot (1954), Yafet (1963)], D'yakonov-Perel' (DP) [D'yakonov (1971), D'yakonov (1972)], and Bir-Aronov-Pikus (BAP) [Bir (1976)] and hyperfine-interaction processes; see e.g. [Paget (1977)]. In n-type semiconductors, two main spin relaxation processes have been found to be important in optical orientation experiments: the DP and the EY mechanisms [Zutic (2004), Song (2002), Kikkawa...
While, the Bir-Aronov-Pikus (BAP) is only significant in p-type semiconductors, is not related to our cases in this thesis.

### 2.2.2 Elliott-Yafet spin flip scattering

#### 2.2.2.1 Film

In EY mechanism, due to the spin-orbit interaction, spin up and spin down states mix. This means that scattering events can change the carrier's spin orientation. This mechanism is responsible for the fast spin relaxation of holes but only in narrow bandgap semiconductors are the electrons affected markedly.

EY spin relaxation rate is [Song (2002)]:

$$\frac{1}{\tau_{EY}} \approx A\alpha^2 \left(\frac{E_k}{E_g}\right)^2 \frac{1}{\tau_p}$$

Eqn 2.35

where \(\alpha = \eta \frac{(1-\eta/2)/(1-\eta/3)}{\eta} \frac{\Delta}{(E_g+\Delta)}\), and \(\Delta\) is the spin-orbit splitting of the valence band. \(E_g\) and \(E_k\) are the energy gap and electron kinetic energy, respectively.

The momentum relaxation time, \(\tau_p\), is determined by temperature, concentration and mobility. For low concentrations where inelastic scattering (by phonons or ionised impurities) dominates, \(\tau_p = \frac{\mu m^*}{e}\). For high concentrations electron-electron scattering (which does not cause dissipation and is therefore not measured by the mobility) may become more important than mobility scattering. \(A\) is a dimensionless constant that varies depending on the orbital scattering interaction process between 2 (for lattice scattering) and 6 (for ionized impurity scattering). In the degenerate limit \(E_k\) may be replaced by \(E_F\), the Fermi energy, and in the high temperature limit, it may be replaced by \(k_B T\).

#### 2.2.2.2 QW

The EY process leads to spin relaxation due to mixing of the valence band states into the conduction band, leading to a nonzero transition rate even for spin-conserving scattering processes. An expression for the spin relaxation rate for the EY process in a QW has been given by [Tackeuchi (1999a,b)]:
where $m^*/m_0$ is the electron effective mass ratio, $E_{1e}$ is the confinement energy for the lowest electron subband, and the other symbols have the same meaning as before. $C_{BE}$ is a dimensionless constant and is predicted to be of the order of unity.

### 2.2.3 D'Yakonov-Perel spin-dephasing

#### 2.2.3.1 Film

DP mechanism happens because of the absence of inversion symmetry, which leads to a $k$-dependent splitting of the conduction band. Since this splitting is analogous to the splitting induced by an external magnetic field, it can be viewed as an internal $k$-dependent magnetic field. Between two scattering events, the spin precesses around this internal field. This means that scattering suppresses this mechanism.

The DP spin relaxation rate in bulk material is given by [Song (2002)]:

$$\frac{1}{\tau_{sp}} = Q\beta^3 \frac{E_k^3}{\hbar^2 E_g} \tau_p$$  \hspace{1cm} \text{Eqn 2.37}

where

$$\beta = \frac{4\eta m^*}{\sqrt{3-\eta} m_0}$$  \hspace{1cm} \text{Eqn 2.38}

where $\eta$ was defined before, $Q$ is a dimensionless factor, predicted to be in the range of 0.8-3, depending on the dominant momentum relaxation process [Song (2002)]. For scattering on ionized impurities and defects $Q \approx 32/21$, whereas for lattice scattering $Q \approx 96/35$ [D'yakonov (1972)].
2.2.3.2 QW

The DP spin dephasing arises because spin-orbit coupling removes spin degeneracy and provides an effective magnetic field that causes the electron spin to precess at a rate determined by the splitting. For small $k$ in bulk crystals this splitting can be written as $\Delta E = \gamma k^3$, where $\gamma$ is the Dresselhaus spin splitting coefficient [Cardona (1988)]. For QWs, the spin splitting is linear in $k$ and the Rashba coefficient $\beta$ related to the spin splitting, is given by [Tackeuchi (1997)]:

$$\gamma = \beta \hbar^3 (2m^* E_s)^{-1/2}$$

Eqn 2.39

The DP spin relaxation rate is:

$$\frac{1}{\tau_s^{DP}} = C_{DP} \frac{m^* (2m^* E_s)^2}{\hbar^8} E_s \tau_p = C_{DP} \frac{\beta^2}{2} \frac{E_{1s}^2}{\hbar^2 E_g} E_s \tau_p$$

Eqn 2.40

where again $C_{DP}$ is a dimensionless constant, predicted to be 16 [Tackeuchi (1997)]. Using Eqn 2.38 and Eqn 2.40 for InSb, $\beta=0.045$ which agrees well with other calculations [Cardona (1988)].

DP lifetime can be tuned by application of a vertical electric field (in a way that depends on growth direction [Cartoixa (2005)], which changes the magnitude of the structural (Rashba) inversion asymmetry and hence also $C_{DP}$. The attractiveness of narrow gap semiconductors for spintronics comes from the large predicted Rashba effect, i.e. the strong tuning of $\tau_s$ with field, and the consequent faster switching and lower power consumption [Bandyopadhyay (2004)].

The scaling of spin relaxation time with electron confinement energy can serve as a criterion for distinguishing between the spin-relaxation mechanisms, assuming the momentum relaxation time can be kept constant. It was shown that $\tau_s \propto E_{1s}^{-2.2}$, in close agreement with the form of Eqn 2.40, for two series of GaAs/AlGaAs QWs, which is consistent with DP being the dominant spin-relaxation mechanism.
[Tackeuchi (1999a)]. On the other hand, in the same work for a set of InGaAs/InP QWs, it was found that $\tau_s \propto E_{1e}^{-1}$, similar to Eqn 2.36 indicating dominance of EY.

It was suggested [Tackeuchi (1997)] that the efficiency of EY process could become much more significant in narrow band gap semiconductors, as can be seen from the $1/E_g^2$ dependence of Eqn 2.35 to Eqn 2.36. However, DP also becomes faster, with $1/E_g$ from Eqn 2.37 to Eqn 2.40, and the cross-over from EY to DP dominated regimes occurs for quantum wells when

$$\tau_x = \left( \frac{C_{el}}{C_{DP}} \right) \frac{3 - \eta}{8} \frac{\hbar}{E_{1e} E_g} \left( \frac{m_o}{m^*} - 1 \right)^2$$

Eqn 2.41

which is not explicitly temperature-dependent. At this point, the spin relaxation lifetime is maximum:

$$\tau_s^{max} = \frac{1}{\sqrt{2 C_{DP} C_{el}}} \frac{\sqrt{3 - \eta}}{2 \eta^2} \frac{m_o}{m^*} \left( 1 - \frac{m^*}{m_o} \right)^{-1} \left( \frac{E_g}{E_{1e}} \right)^{3/2} \frac{\hbar}{k_B T}$$

Eqn 2.42

### 2.2.4 Regimes of domination by EY and DP

The most important difference between EY and DP mechanisms is their opposite dependence on $\tau_s$. It can be seen that the spin lifetime in the case of DP relaxation mechanism is inversely proportional to the electron mobility (Eqn 2.37 and Eqn 2.40), whereas in the case of EY mechanism it is linearly proportional (Eqn 2.35 and Eqn 2.36). This means that for materials with low electron mobility the role of EY mechanism is increased, and the cross-over temperature is higher.

In the limit of degenerate statistics the typical electron kinetic energy is given by $E_F$, the Fermi energy, and in the opposite limit of non-degenerate statistics, it is $k_BT$. For example, in the limit of non-degenerate statistics the EY expression for bulk material Eqn 2.35 becomes:
\[ \frac{1}{\tau_s^{\text{EY}}} \approx A \alpha^2 (k_B T / E_g)^2 1/\tau_p \]  
Eqn 2.43

whereas in the case of degenerate statistics the DP expression Eqn 2.37 becomes:

\[ \frac{1}{\tau_s^{\text{DP}}} \approx Q_d \beta^3 \frac{E_p^3}{\hbar^2 E_g} \tau_p \]  
Eqn 2.44

Therefore the DP dominates at high temperature because of the higher power of \( T \).

The crossover between DP and EY mechanisms may be found by equating Eqn 2.35 and Eqn 2.37:

\[ \mu^{(\omega)} = \frac{\alpha e \hbar}{\beta m} \sqrt{\frac{A}{Q E_g E_k}} \]  
Eqn 2.45

It was shown [Song (2002)] that in high mobility semiconductors the EY mechanism dominates only at very low temperatures even in NGS, where the efficiency of the EY process is larger. The situation is different in low mobility materials. It was already demonstrated that the crossover between the EY and DP process in doped InSb quantum wells could occur even at room temperature [Litvinenko (2006a)].

Figure 2.7 provides the total spin relaxation time \( \tau_s = (1/\tau_s^{\text{EY}} + 1/\tau_s^{\text{DP}})^{-1} \) for n-type samples. \( \tau_s \) ranges from 1 ps to 100 ns for n-type materials. For low concentration \( n < 10^{16} \) cm\(^{-3} \) for InSb at 300K, and assuming dominance of lattice scattering, we can use non-degenerate EY and DP spin relaxation rates, with \( A = 2 \) (2-6), and \( Q_{\text{ad}} = 0.8 \) (0.1-3) [Song (2002)].
Figure 2.7 Total spin relaxation time for n-InAs and n-InSb. The colour of each cell represents $\tau_s$ for the point at the lower left corner of the cell according to the colour map at the right-hand side. $N_D$ is in cm$^{-3}$ and $\tau_s$ is in second. $N_D$ is fixed to $5 \times 10^{13}$ cm$^{-3}$ [Song (2002)].

For n-type materials, $\tau_s$ increases as $T$ decreases with the longest $\tau_s$ found at $N_D = 10^{17}-10^{18}$ cm$^{-3}$ instead of in purer materials. This is because the regime shown in Figure 2.8 is dominated solely by the DP process and $1/\tau_{DP}$, which is proportional to $\tau_p$, increases as the impurity concentration decreases. The same qualitative feature has also been found in another experiment [Kikkawa (1998)].

The dependence of spin relaxation time at this range of temperature for doping densities between $5.2 \times 10^{16}$ and $8.8 \times 10^{17}$ cm$^{-3}$ for n-InAs, showed that the results are in accord with the DP model, particularly by including electron-electron scattering [Murdin (2005), Song (2002)].

Theoretical prediction indicates that for bulk III-V n-type semiconductors the transition temperature from the DP-dominated regime to the EY-dominant regime is at $T \sim 5$ K [Song (2002)]. The EY process is more effective for low mobility InSb at low temperature and at small doping concentrations ($N_D < 10^{18}$ cm$^{-3}$) [Chazalviel (1975), Bichard (1979)], whereas the DP process dominates at room temperature.
Typically, the magnitudes for spin lifetimes measured at low temperature are in the range 1-10 ns, obtained from spin resonance in degenerate n-type InSb at helium temperatures. At elevated temperatures the lifetimes can be of order a few picoseconds.

For QWs, of the two main spin relaxation processes, only in the case of DP-dominated scattering will an electric field applied in the growth direction cause a modulation of the strength of the spin-orbit coupling through the Rashba effect and hence also the rate of DP spin relaxation. This is an essential component for spintronic devices requiring modulation of the spin lifetime with an electric field. It is therefore important to establish the conditions in which spin polarization lifetimes are both long and dominated by the DP process.

Similarly, changing from InSb wells to InAs wells is predicted to increase the maximum spin lifetime by about an order of magnitude. From Eqn 2.40, DP can be expected to dominate at room temperature even in narrow gap QWs unless the mobility is low.
3 Experimental technique

We describe in detail the experimental techniques developed for spin dynamics studies at Surrey, which pertains to both chapters 4 and 5. The other main experiment described in this thesis - the far-infrared dynamics of silicon donors - is very similar, and will be described along with the results in Chapter 6. In addition to the experimental techniques, this chapter also contains details about the samples used, including mobility and density results. Some previous published works by collaborators on these samples about spin lifetime measurements are included to be used for our discussion. These are included here rather than in the spin lifetime chapter (Chapter 4) in order to clearly separate them from the work of the author.

3.1 Experimental Method for spin dynamics in narrow gap semiconductors

A circularly polarized pump-probe experiment was performed to study the spin dynamics. The light source was a difference frequency generator, (DFG, Coherent Inc.), which mixes the signal and the idler beams of an optical parametric amplifier, itself pumped by an amplified Ti:sapphire oscillator (refer to section 3.2). The DFG provides pulses shorter than 100 fs duration at a repetition rate of 250 kHz. The wavelength of the laser radiation could be continuously tuned from 3 to 13 μm. The time resolution of the experiments was about 200 fs. In all cases, the pump and probe photon energy was maintained just above the bandgap. The laser power before the beam splitter was 4 mW. The mid-infrared beam from the DFG was split into two beams, a stronger pump and a weak probe (with intensity of a few percent of that of the pump), which passed along a time delay stage. After polarization both beams were focused on the sample aperture in the N₂ cryostat, temporally and spatially, via focusing mirrors. After passing through the material, the pump beam was dumped and the probe beam was detected with a cooled InSb detector, which is sensitive for 2-5 μm and fast enough for the repetition rate of 250 kHz. The sample was mounted in the vacuum on the cold finger of a liquid nitrogen bath cryostat. To have circularly polarized probe beam, a variable quarter wave plate has been used. A ZnSe photoelastic modulator (PEM) was used to modulate the polarization of the pump beam (Figure 3.1).

The induced transmission change of the probe beam was measured as a function of the time delay between pump and probe pulses having the same circular polarizations
as the probe (SCP), and opposite circular polarizations (OCP). The optical polarization $(SCP - OCP)/(SCP + OCP)$ is proportional to the spin polarization in the sample. The change in transmission of the probe beam due to the modulation (i.e. the circular dichroism, $\Delta T_{CD}$) was detected as a function of the time delay between pump and probe pulses. It is a measure of the spin-polarization, which can be detected using phase sensitive techniques [Boggess (2000), Murzyn(2003a,b), Murdin (2005)].

The sum of the OCP and SCP signals is a measure of the total population (recombination time of photo-generated electrons) and is reproduced by a linearly polarized pump with the PEM modulator replaced by an optical chopper. The optical polarization is defined by

$$P_{opt} = \frac{\Delta T_{SCP} - \Delta T_{OCP}}{\Delta T_{SCP} + \Delta T_{OCP}} = \frac{\Delta T_{CD}}{\Delta T_{LP}} = P_0 \exp (-t/\tau_c)$$

Eqn 3.1

where $\Delta T$ (the probe transmission change) decays exponentially with a decay constant equivalent to the spin life-time. $\Delta T_{LP}$ is the linearly polarised transient transmission change, which is measured separately by switching off the PEM and

![Figure 3.1 Schematic of apparatus for pump-probe experiment, (note that magnetic field is added for g-factor measurement, chapter 5)](image-url)
inserting an optical chopper (with everything else unchanged). In zero magnetic field $P_0$ is a constant that depends on the absorption coefficient, i.e. on the matrix elements. For III-V narrow gap semiconductor films and heterostructures (i.e. almost all of the samples studied here) the carrier recombination proceeds via the Auger process [Lau (2001)] and does not follow a single exponential decay, but the fastest component is much longer than the observed spin relaxation times and therefore $\Delta T_{LP}$ is observed to be as a constant. In most such cases we have extracted spin relaxation times from $\Delta T_{CD}$ decay only.

### 3.2 Pulse source - Ti:sapphire mode locked laser system

The Ti:sapphire laser is a solid-state laser based on solid-state gain media - titanium-doped sapphire crystals ($\text{Ti}^{3+}: \text{Al}_2\text{O}_3$), which exhibit a broad absorption band, located in the blue-green region of the visible spectrum with a peak around 490 nm. The great interest in this material arises from the widest known gain bandwidth, which allows tuneable laser output between 650-1100 nm, with the peak of the gain curve around 800 nm. Besides having favourable spectroscopic and lasing properties, the sapphire host has high thermal conductivity, exceptional chemical inertness and mechanical rigidity. A very important application of Ti:sapphire lasers is the generation and amplification of femtosecond mode locked pulses, for example see [Svelto (1998), Koechner (1995)]. Mode-locking describes the set of techniques that are used to generate a known correlation between the phases and therefore make it possible to predict when the intensity maxima will occur. There are several different ways a laser can be mode-locked, such as the Kerr lens mode-locking (KLM).

Since in some spectral regions, it is impossible to find materials with similar properties to Ti:sapphire, we can convert the wavelength range by nonlinear optical mechanisms.

Our Ti:sapphire laser system (Coherent) [The Coherent, Inc. laser group CA Operator’s Manuals; including Mira, RegA, OPA, DFG], which emits ultrashort ($<100$ femtosecond or $10^{-13}$ second) pulses, comprises two diode pump lasers (Verdi), a titanium-doped sapphire mode-locked laser oscillator (Mira), regenerative amplifier (RegA), parametric amplifier (OPA), and difference-frequency generator (DFG) (tuneable in the mid-infrared range), see Figure 3.2.
The pump lasers for the Mira and RegA systems, two Verdi available in 5-watt and 10-watt power, respectively, are CW diode-pumped lasers, frequency-doubled Nd:YVO$_4$ laser providing single-frequency green output at 532 nm with vertical polarization.

### 3.2.1 Oscillator
The Mira seed is a mode-locked ultrafast laser that uses Ti:sapphire as the gain medium to produce ultrashort, wide bandwidth, femtosecond pulses. The Mira which is pumped with 5 W Verdi, can produce less than 100 fs pulses at 76 MHz with about 32 nm bandwidth, 50 fs pulse width, from 780 to 840 nm, with an output power of more than 700 mW (CW), or 450 W (mode lock), at linear horizontal polarization. This output is used for seeding titanium:sapphire RegA where maximum seed bandwidth enables short amplified pulse width generation. The seed is designed for optimum bandwidth and amplifier seeding.

### 3.2.2 Regenerative amplifier
The regenerative amplifier (RegA) is a Ti:sapphire regenerative amplifier system, which is pumped by the 10-watt Verdi. It uses a short pulse Mira Seed, combined with an external stretcher/compressor, to provide amplified pulses with 1.5 W power (before the compressor), with about 150 fs pulsewidth, in linear horizontal polarization with repetition rate of more than 250 KHz.

### 3.2.3 Optical parametric amplifier
The optical parametric amplifier (OPA) as a non-linear optical phenomenon converts one fundamental wavelength from the source laser into a tuneable spectrum of long wavelengths. The OPA uses the RegA output to produce amplified femtosecond output pulses tuneable from 1.1 to 2.6 $\mu$m, at 250 kHz repetition rate, with nearly 200 mW output power. The OPA is white-light-continuum (460–1200nm) seeded to generate an idler (1.6 to 2.6 $\mu$m) and a signal (1.1 to 1.6 $\mu$m), which with white light are output from OPA. It uses type II non-linear BBO crystal to extend the wavelength coverage of the RegA.
Femtosecond Laser

I – 800 nm, 500 mW, 50 fs;
II – 800 nm, 1.5 W, 100 fs;
III – from 1.1 to 2.5 μm, 150 fs (not used here)
IV – from 1.1 to 2.5 μm, (~200 mW), 150 fs;
V – from 3 to 12 μm, 6 mW, 150 fs

Figure 3.2 Coherent Ti:sapphire laser set up in the ultrafast laboratory a) schematic – the components are described in the text b) photograph.
3.2.4 Difference frequency generator

Wavelength conversion is very important to generate radiation of much longer wavelength than Ti:sapphire lasers. The Difference Frequency Generator (DFG) uses difference-frequency mixing of the signal and idler outputs from the optical parametric amplifier (OPA) to generate tunable light in the mid-IR spectral region. The signal and idler from the OPA are focused into a non-linear crystal, $\text{AgGaS}_2$, and phase-matched, to produce light in the mid-IR range. The DFG allows continuous tuning from around 3 to 13 $\mu$m, with output power about 6mW (for $\lambda = 4\mu$m), and 250 kHz repetition rate suitable for ultrafast spectroscopy in the mid-IR range. The possibility of time-resolved studies in the mid-IR spectral region allows real-time studies of semiconductor responses. Typical tuning ranges of DFG can be achieved by accessing external control knobs on the OPA and DFG units.

3.3 Narrow Gap Semiconductor Samples

Indium antimonide and indium arsenide are examples of III-V compounds NGSs. Therefore, they are interesting for testing the band structure calculations, because of small electron effective mass, high electron mobility as well as strong spin-orbit coupling, and hence a strong Rashba effect [Rashba (2003)].

3.3.1 Material parameters

In this thesis we have used the well known literature values of several important parameters. The materials InAs and InSb were taken have the parameters shown in Table 3.1 and Table 3.2.
Table 3.1 InAs Material properties, (all parameters are defined in chapter 2 except $\alpha$ and $\beta$ which are from Varshni form (Eqn 5.3), and the last three, which show, respectively, lattice constant, $1/\kappa$ - where $\kappa$ is compressibility, and pressure coefficient of the bandgap).

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<td>[Littler (1985), Ioffe]</td>
</tr>
<tr>
<td>$\Delta_{so}$ (eV)</td>
<td>0.80</td>
<td>[Zwerdling (1961)]</td>
</tr>
<tr>
<td>$\alpha$ (eVK$^{-1}$)</td>
<td>$6 \times 10^4$</td>
<td>[Littler (1985), Ioffe]</td>
</tr>
<tr>
<td>$\beta$ (K)</td>
<td>500</td>
<td>[Littler (1985), Ioffe]</td>
</tr>
<tr>
<td>$m^*/m_0$</td>
<td>0.014</td>
<td>[Lawaetz (1971), Ioffe]</td>
</tr>
<tr>
<td>$E_p$ (eV)</td>
<td>23</td>
<td>[Cavassilas (2001), Luttinger (1956)]</td>
</tr>
<tr>
<td>$E'_p$ (eV)</td>
<td>10.5</td>
<td>[Berolo (1973)]</td>
</tr>
<tr>
<td>$E(\Gamma^*_s)$ (eV)</td>
<td>3.49</td>
<td>[Cardona (1967)]</td>
</tr>
<tr>
<td>$E(\Gamma^*_c)$ (eV)</td>
<td>3.16</td>
<td>[Cardona (1967)]</td>
</tr>
<tr>
<td>$C$</td>
<td>-2</td>
<td>[Hermann (1977)]</td>
</tr>
<tr>
<td>$C'$</td>
<td>-0.02</td>
<td>[Hermann (1977)]</td>
</tr>
<tr>
<td>$\gamma_1, \gamma_1^L$</td>
<td>3.1, 34.8</td>
<td>[Wiley (1975), Vurgaftman (2001)]</td>
</tr>
<tr>
<td>$\gamma_2, \gamma_2^L$</td>
<td>-0.60, 15.5</td>
<td>[Lawaetz (1971), Vurgaftman (2001)]</td>
</tr>
<tr>
<td>$\gamma_3, \gamma_3^L$</td>
<td>0.67, 16.5</td>
<td>[Lawaetz (1971), Vurgaftman (2001)]</td>
</tr>
<tr>
<td>$\kappa, \kappa^L$</td>
<td>-1.3, 14.76</td>
<td>[Wiley (1975), Lawaetz (1971)]</td>
</tr>
<tr>
<td>$F$</td>
<td>-0.5</td>
<td>[Pidgeon (1967)]</td>
</tr>
<tr>
<td>$N_i$</td>
<td>-0.65</td>
<td>[Jiménez-González (1994)]</td>
</tr>
<tr>
<td>$d(A'^*)$ (RT)</td>
<td>6.4858</td>
<td>[Tsay (1972)]</td>
</tr>
<tr>
<td>$\nu(\partial P / \partial V)_T$ (Kgcm$^{-2}$)</td>
<td>$-4.6 \times 10^5$</td>
<td>[Stradling (1970)]</td>
</tr>
<tr>
<td>$(\partial E_g / \partial P)_T$ (eVKg$^{-1}$cm$^2$)</td>
<td>$1.5 \times 10^{-5}$</td>
<td>[Stradling (1970)]</td>
</tr>
</tbody>
</table>
3.3.2 InAs samples

From a practical point of view, both in terms of cost, and also for optical experiments the need for transparency, NGS materials are often grown as epi-layers on GaAs substrates by molecular-beam epitaxy (MBE). Although the lattice mismatch between InAs and GaAs (7%), results in the formation of an interface accumulation layer with a large defect concentration, this can be minimised with sophisticated buffering schemes. The InAs samples used here was Si doped, grown on semi-insulating GaAs by MBE, Table 3.3.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Thickness (µm)</th>
<th>doping</th>
<th>mobility µ (RT) ×10^4 cm^2 V^{-1} s^{-1}</th>
<th>density, n_0 (×10^{17} cm^{-3}) (RT)</th>
</tr>
</thead>
<tbody>
<tr>
<td>IC311</td>
<td>4</td>
<td>Si doped</td>
<td>1.72</td>
<td>1.22</td>
</tr>
<tr>
<td>[Wang (1992)]</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>U24041</td>
<td>0.15</td>
<td>-</td>
<td>1.43</td>
<td>8.1</td>
</tr>
<tr>
<td>U24T4</td>
<td>0.27</td>
<td>-</td>
<td>5.75</td>
<td>2.4</td>
</tr>
<tr>
<td>U24T24</td>
<td>1.00</td>
<td>-</td>
<td>9.22</td>
<td>3.1</td>
</tr>
</tbody>
</table>

3.3.2.1 InAs thick films

Temperature, doping, and pump wavelength dependence of spin relaxation in n-InAs bulk-like samples grown on semi-insulating GaAs substrates have been studied by Murdin et al. [Murdin (2005)]. Their results are consistent with the DP mechanism for temperatures between 77 and 300 K. The temperature dependence of the concentration and hall mobility for sample IC311, which we have used in our study, is shown in Figure 3.3. The temperature dependence of the spin life time in n-InAs determined experimentally and theoretically is illustrated in Figure 3.3c.
Figure 3.3  

a) The carrier concentration $n$ (filled symbols) and mobility $\mu$ (open symbols) as a function of temperature measured experimentally with the Hall effect for InAs sample IC311;  
b) The inferred chemical potential $E_F$ (filled symbols) and orbital scattering time $\tau$ (open symbols);  
c) The temperature dependence of the spin life time in n-InAs determined experimentally; filled circles show sample IC311. From theory, dotted curve shows non-degenerate DP, and dashed curve for degenerate DP. Inset: comparison of the spin lifetime versus temperature for non-degenerate DP and EY mechanisms [Murdin (2005)].

3.3.2.2 InAs thin films  
The effect of inversion layers on the spin dynamic properties of InAs films with different mobility ($\mu$) and carrier concentration ($n$) in different layers of the film, grown on semi-insulating GaAs substrates by MBE has been already considered by our group [Litvinenko (2006b)]. We introduce some results of that paper here, to be able to compare them with our results for InSb films. The InAs samples characteristic has been mentioned in Table 3.3.
Theoretical band-bending profiles of three InAs films (calculated by J. J. Harris, Imperial College) at 300 K, is demonstrated in Figure 3.4. The figure shows three samples, each of which shows three distinct regions; a surface accumulation region, an accumulation region at the interface between the film and the substrate, and a central semiconducting region.

Figure 3.4. Theoretical band-bending profiles Calculated by J. J. Harris, Imperial College, for the three samples with different thicknesses at 300K.

Figure 3.5 shows the time evolution of the spin population measured at room temperature for these layers with three different thicknesses. The results show that the longer lifetime in the thicker sample is due to the central semiconducting layer. The thinner film results and their similar short components of the spin lifetime suggest that they have the same origin.
Figure 3.5 Left: The time evolution of spin population measured at RT for the undoped InAs films of different thicknesses: 0.15 µm (green triangles), 0.27 µm (pink circles), and 1 µm (blue squares); right: The temperature dependence of the spin lifetime from the population decay curves in a): 0.15 µm (green triangles), 0.27 µm (pink circles), and 1 µm (blue squares). In the case of the 0.27 µm film the decay has two components; the slow component is shown with open circles and the fast component with solid circles [Litvinenko (2006b)]. Also shown are data for a doped 4 µm thick InAs film lightly Si-doped $5.2 \times 10^{16} \, \text{cm}^{-3}$ sample (IC313) (solid diamond) from [Murdin (2005)].

Figure 3.6 shows the temperature dependence of the derived mobility of the InAs interface layers (diamonds) [Litvinenko (2006b)] and bulk-like layers (circles) [Murdin (2005)]. The crossover mobility between DP and EY processes is shown by the solid line for the case of lattice scattering and nondegenerate statistics.

Figure 3.6 Temperature dependence of the derived mobility of the InAs interface layers (diamonds) and bulk-like layers (circles) deduced by multi-channel differential analysis. The crossover mobility between DP and EY processes is shown by the solid line for the case of lattice scattering and nondegenerate statistics [Litvinenko (2006b)].
3.3.3 **InSb samples**

3.3.3.1 **InSb films**

The InSb samples used for our study were grown by MBE on a semi-insulating GaAs substrate, see Table 3.4.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Structure</th>
<th>mobility ( \mu ) (RT) ( \times 10^4 \text{cm}^2\text{V}^{-1}\text{s}^{-1} )</th>
<th>density, ( n_e ) (cm(^{-3})) (RT)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ME1655</td>
<td>4</td>
<td>6.37</td>
<td>2.8\times10^{15}</td>
</tr>
<tr>
<td>U20017h</td>
<td>0.1</td>
<td>0.69</td>
<td>0.62\times10^{17}</td>
</tr>
<tr>
<td>U20004g</td>
<td>0.3</td>
<td>1.6</td>
<td>0.24\times10^{17}</td>
</tr>
<tr>
<td>U20028a</td>
<td>1</td>
<td>4.0</td>
<td>0.18\times10^{17}</td>
</tr>
</tbody>
</table>

3.3.4 **InSb/AlInSb QWs**

Semiconductor quantum wells (QWs) are important components for future spintronic transistor device applications because the method for electrical manipulation of spin polarizations in semiconductors is likely to be the Rashba effect [Rashba (2003)]. Among the III–V semiconductors, InSb with the smallest electron effective mass and the largest g-factor, is used to explore some aspects of electron spin in InSb quantum wells. The QW samples under study are represented in Table 3.5.

<table>
<thead>
<tr>
<th>samples</th>
<th>L (nm)</th>
<th>Al, %</th>
<th>( \mu ) (( m^2 V^{-1}s^{-1} )) at 77K (RT)</th>
<th>( n ) (( 10^{11} \text{cm}^{-2} )) at 77K (RT)</th>
<th>Te-doping</th>
</tr>
</thead>
<tbody>
<tr>
<td>ADJ1127</td>
<td>10</td>
<td>8</td>
<td>-</td>
<td>-</td>
<td>undoped</td>
</tr>
<tr>
<td>ADJ1129</td>
<td>30</td>
<td>8</td>
<td>-</td>
<td>-</td>
<td>undoped</td>
</tr>
<tr>
<td>ADJ1126</td>
<td>40</td>
<td>8</td>
<td>-</td>
<td>-</td>
<td>undoped</td>
</tr>
<tr>
<td>me1831</td>
<td>20</td>
<td>15</td>
<td>0.9 (1.37)</td>
<td>5.7 (7.3)</td>
<td>uniformly doped</td>
</tr>
<tr>
<td>me1833</td>
<td>20</td>
<td>15</td>
<td>4.87 (2.1)</td>
<td>3.6 (5.3)</td>
<td>( \delta )-doped 20nm from QW</td>
</tr>
<tr>
<td>me2501</td>
<td>30</td>
<td>8</td>
<td>- (4.0)</td>
<td>- (1.8)</td>
<td>undoped</td>
</tr>
<tr>
<td>me2502</td>
<td>30</td>
<td>8</td>
<td>5.27 (2.2)</td>
<td>8.95 (14.1)</td>
<td>( \delta )-doped 20nm from QW</td>
</tr>
<tr>
<td>me2504</td>
<td>30</td>
<td>8</td>
<td>16 (3.7)</td>
<td>4.24 (6.2)</td>
<td>( \delta )-doped 20nm from QW</td>
</tr>
<tr>
<td>me2507</td>
<td>30</td>
<td>8</td>
<td>11.9 (2.9)</td>
<td>5.43 (10.3)</td>
<td>( \delta )-doped 20nm from QW</td>
</tr>
<tr>
<td>me2509</td>
<td>30</td>
<td>8</td>
<td>14.5 (3.5)</td>
<td>2.26 (5.99)</td>
<td>( \delta )-doped 20nm from QW</td>
</tr>
</tbody>
</table>
The first three QW samples have been used in our work. Some other samples were studied to measure the temperature and mobility dependence of spin relaxation time in narrow gap antimonide single QWs (InSb/AlInSb) [Litvinenko (2006a)], Figure 3.7. They showed that for sample ME1833, $\tau_s T$ is inversely proportional to $\mu$ and is well described by the DP process. For sample ME1831F, at high temperature and the high

Figure 3.7 a) Temperature dependence of the electron mobility for samples ME1831F and ME1833 (on logarithmic scales); b) Experimental temperature dependence of the spin relaxation time for samples ME1833 (circles) and ME1831F (triangles). The labels refer to the lines which represent theoretical predictions for $\tau_s$. The thick lines use the interpolated mobility data of Figure 3.7a for sample ME1833, the thin lines use that of sample ME1831F. The solid lines are according to the DP model of equation (Eqn 2.40) with $C_{DP}=32$, and the dashed lines use the EY model of equation (Eqn 2.36) with $C_{EY}=7.5$; c) The comparison of experimental product of spin relaxation time with temperature, $\tau_s T$ taken from b) (open symbols) on mobility, $\mu$, from a). Solid line: DP process using $C_{DP}=32$ as a fitting parameter; dotted line: EY process with $C_{EY}=7.5$ as fitting parameter [Litvinenko (2006a)].
mobility end of the range the dependence of $\tau_s T$ is again inversely proportional to $\mu$ and the DP process dominates. At lower temperature and mobility, the EY process appears to become comparable to the DP process and a clear distinction cannot be made.

For these samples the optimum mobility is about $1 \text{ m}^2\text{V}^{-1}\text{s}^{-1}$ (from Figure 3.7), and if this is achieved at room temperature, then $\tau_{s_{\text{max}}} \approx 0.5 \text{ ps}$ (from $\tau_{s_{\text{max}}}T=150 \text{ psK}$). Because $E_\text{1d} \approx L^{-2}$, $\tau_{s_{\text{max}}}$ scales with $L^3$ (from Eqn 2.42) so strong increases in spin lifetime can be achieved with small increases in well width. The DP spin relaxation mechanism dominates at high mobility, while EY dominates at low mobility. For narrow gap QWs the cross-over mobility (given by Eqn 2.41) is increased, but generally speaking the mobility of NGS is also high. DP can be expected to dominate at room temperature even in narrow gap QWs unless the mobility is low.

### 3.4 QD samples

Quantum dot semiconductor materials have attracted much attention because their physical and chemical properties can be systematically tuned by variation of the size. For example in some II–VI QDs great progress has been achieved in the area of synthesis, structural characterization, electronic and optical properties and the potential for their applications. HgTe as II–VI compound has zero-gap in the bulk material, and may also have useful near- and mid-infrared transition energies when quantum confinement is introduced. However, IV–VI QDs such as PbSe have been given less attention even though they have the effects of strong confinement on electrons and phonons [Wise (2000), Wehrenberg (2002), Wehrenberg (2003)], and they have transitions in a technologically important region of the spectrum (1.3-1.5\text{\mu m}). Since PbSe has narrow bandgap, is therefore used extensively in the mid-infrared optoelectronic device applications. The absorption peak of PbSe QDs depends on their radius and can be shifted into the near-infrared telecommunications wavelength band. Compared to QWs, which require expensive and complicated growth technology, lots of QDs can be provided with simple chemistry based on cheap technology by mixing liquid chemicals in glass. They can be put in special places, and they show interesting physical properties. Moreover, QDs are potential elements for spintronic application ranging due to their long spin lifetime as well as
the possible control over structural and electronic environment of localized carriers. In this work, HgTe and PbSe QDs have been studied.

The spectra of representative PbSe QD samples used for our study are shown in Figure 3.8 and of HgTe in Figure 3.9.

Figure 3.8 Spectrum of PbSe QD samples with slightly different chemical synthesis times (i.e. different nanocrystal size distributions). The nanocrystals are encased in a matrix of PMMA and on a glass substrate.

Figure 3.9 Photoluminescence spectra of two HgTe QD samples with slightly different chemical synthesis times (i.e. different nanocrystal size distributions).
4 Experiment on spin polarisation decay

4.1 Introduction
Spin dynamics in semiconductors have been studied broadly, because of their importance for the development of spintronic devices [Ziese (2001), Zutic (2004)]. Due to its very well established growth and the band gap energy being within the range of widely available ultrafast Ti:sapphire oscillators, most investigations have been focused on bulk or heterostructures of GaAs-based semiconductors [ Awschalom (2002)].

NGS materials could be also very beneficial for designing future spintronic devices because of their strong spin-orbit coupling. The presence of space-charge accumulation or inversion layers at the surfaces of $Hg_{1-x}Cd_xTe$ ($x<0.3$), InAs, InSb and $InAs_{1-x}Sb_x$ could be used to design a contact with metallic materials without a significant resistance mismatch and consequent need for Schottky barrier [Zwierzycki (2003)]. This makes them potential candidates for applications utilizing spin injection from ferromagnetic metals. The effect of surface accumulation due to a native surface defect above the bulk conduction band edge is known to be particularly strong with InAs [Wieder (1974), Washburn (1979), Tsui (1971)] and $Hg_{1-x}Cd_xTe$ [Agnihotry (1998)]. Electron spin relaxation strongly depends on the scattering rate and electron kinetic energy. Thus layers with different carrier concentration and mobility have different spin lifetime and different spin relaxation processes could dominate.

Among narrow-gap semiconductors InAs and InSb have been of particular interest. There was much attention on InSb because of the recent development of extraordinary magneto-resistive sensors [Solin (2000)], high speed electronic transistors [Ashley (1995)], and possible hybrid spintronic devices based on high mobility InSb epi-layers [Murzyn (2003a)]. Moreover, a small electron effective mass as well as strong spin-orbit coupling, and hence a strong Rashba effect, make InSb a promising candidate for spintronic application.

These materials can be constructed with particular characters for investigation as films, quantum wells, or quantum dots. Some studies have been done by our group on InAs films (see section 3.3.2) [Murdin (2005), Litvinenko (2006b)] and on
InSb/AlInSb quantum wells (QWs) (see section 3.3.4) [Litvinenko (2006a)]. In this work, spin dynamics on InAs and InSb films, InSb quantum wells, and some quantum dot samples has been investigated. All sample characteristics have been mentioned in section 3.3, Tables 3.1 to 3.4.

4.2 Results- spin lifetime

4.2.1 InAs thick film

The spin relaxation time of n-InAs sample (IC311) 4 \( \mu \text{m} \) thick grown on semi-insulating GaAs substrates by MBE has been studied for different temperatures between 77 K and 293 K. We have used a circularly polarized pump-probe experiment (refer to section 3.1) to study spin dynamics. The change in transmission of the probe beam due to the modulation (i.e. the circular dichroism, \( \Delta T_{\text{CD}} \)) along with the linearly polarised transmission change (\( \Delta T \)) was detected as a function of the time delay between pump and probe pulses at different temperatures. The decay times, are measured from the curve fitting by exponential decay functions. The spin lifetime is found by dividing the two results. The result at \( \lambda=3.4 \ \mu\text{m} \) for RT is shown in Figure 4.1.

The temperature dependence of spin lifetime is presented in Figure 4.2. During experiment, with changing the temperature wavelength was tuned to the InAs band edge. It can be seen when temperature is increased, spin lifetime is decreased, in accord with both the EY (Eqn 2.35) and DP (Eqn 2.37) processes.

Of course the reason why the DP process has temperature dependency is because the spin precession rate in zero externally applied magnetic field increases with increasing electron kinetic energy (which increases with temperature) due to the Dresselhaus \( k^3 \) terms in the band structure. The EY spin scattering rate increases with electron kinetic energy because it arises from the mixing of states, which increases with increasing \( k \) (as illustrated by Eqn 2.9).
Figure 4.1 Time evolution of the $\Delta T_{CD}$ and $\Delta T_{LP}$ and their division for InAs IC311 at RT for 3.4 $\mu$m; by using curve fitting via a single exponential decay function, decay time can be obtained.

Our result confirms the results of Murdin et al. [Murdin (2005)] and validates the reproducibility of the experiment for InAs sample IC311. They have reported that spin relaxation mechanism for n-InAs at this range of temperature for doping densities between $5.2 \times 10^{16}$ and $8.8 \times 10^{17}$ cm$^{-3}$ agrees with the DP model and the experimental behaviour of $\tau_s (T)$, follows the mixture of theoretical degenerate DP (at lower temperatures), and non-degenerate DP (at higher temperatures); also in agreement with the reference of [Song (2002)], which specifies that the spin relaxation mechanism for n-InAs is governed by the DP mechanism in this range of temperature and doping density, (Figure 2.8).

The DP mechanism dominates at high temperature because of the higher power of $T$ (see sections 2.2.3 and 2.2.4). When the non-degenerate DP relaxation mechanism dominates, Eqn 2.37 ($E_k \rightarrow kT$), the lifetime is inversely proportional to $\mu$ and to $T^3$. At high temperature usually $\mu$ decreases (Figure 3.3a), but the effect of this is negligible because of the very strong explicit temperature dependence, resulting in a decrease of $\tau_s$. The degenerate spin relaxation lifetime is concentration dependent via $E_F$ from Eqn 2.37 ($E_k \rightarrow E_F$). As it can be seen from Figure 4.2 the spin lifetime is
tending to \( \sim 40\text{ps} \) at low temperature, and this agrees with the prediction from Eqn 2.37. At low temperatures the effects of degeneracy and of the EY mechanism must be considered.

![Graph](image)

**Figure 4.2** Spin life-time versus temperature for \( n \)-InAs (IC311). At this range of temperature, when the nondegenerate DP mechanism is dominant, \( \tau_{\text{DP}} \) is inversly proportional to \( T^3 \) and by increasing the temperature, the spin lifetime decreases.

### 4.2.2 InSb films

#### 4.2.2.1 InSb thick film

Similar to InAs thick film (IC311), pump-probe time resolved experiment (refer to section 3.1), has been used at temperatures between 77 K and 293 K to study the spin relaxation time of an undoped InSb sample (ME1655) 4 \( \mu \text{m} \) thick grown on semi-insulating GaAs substrates by MBE. The mobility and carrier concentration of this film were 6.37 \( m^2/\text{Vs} \) (3 \( m^2/\text{Vs} \)) and 1.4\( \times10^{16} \text{ cm}^{-3} \) (2.8\( \times10^{15} \text{ cm}^{-3} \)) at 300 K (77 K). The typical difference in the probe transmission is obtained versus time delay at \( \lambda=5.5 \mu\text{m} \) for RT, can be seen from Figure 4.3. It shows an example of the measured \( \Delta T_{\text{CD}} \) and \( \Delta T_{\text{LP}} \) decay, where it can be seen that the spin decay occurs on a time-scale of a few picoseconds, whereas the population recombination occurs on a scale of
several hundred picoseconds. Therefore, we have extracted spin relaxation times from $\Delta T_{CD}$ decay only.

![Graph showing InSb ME1655 at 293 K](image)

Figure 4.3 Evolution as a function of time delay between the pump and probe pulses of the recombination (right) and spin population (left) for InSb at $\lambda=5.5 \ \mu$m at RT.

The temperature dependence of the spin life time from 77K to 293 K achieved for this sample is shown in Figure 4.4. For our range of temperature in n-InSb bulk, DP mechanism dominates [Zutic (2004), Song (2002), Kikkawa (1998), Murzyn (2003b)], and by increasing the temperature $T_s$ decreases.

![Graph showing temperature dependence of spin life time for InSb sample](image)

Figure 4.4 Temperature dependence of spin life time for InSb sample (ME1655) 4 $\mu$m thick.
The error bars show the least square of the fitting for repeated measurements. In reality it includes the effect of other experimental errors such as intensity modulation of PEM, and temperature dependency of magnetic field, etc.

### 4.2.2.2 InSb thin films - structures and transport measurement

Following the investigation of InAs thin films by others in our group (section 3.3.2.2) we have investigated the spin relaxation in undoped InSb films grown on GaAs in the temperature range from 77K to 293K [Litvinenko (2007)]. We note that the semi-insulating GaAs substrates do not themselves contribute to the transport of the samples, and have been used for InSb growth because they are readily available, are transparent in the spectral region of interest and offer the possibility of integrating with advanced GaAs technology.

The lattice mismatch (14.6%) results in the formation of an interface accumulation layer with a large defect concentration. The electron mobility in this layer could be several orders of magnitude smaller than that in bulk material, and is the main obstacle to the manufacturing of thin InSb films of high mobility. Due to native surface defects, a low-barrier depletion region is formed on the surface of the InSb films. The existence of an inversion layer at the surface would be preferable, to form a contact with metallic materials without a significant resistance mismatch, and, consequently, to increase the efficiency of spin injection from ferromagnetic materials. Of course, as observed in InAs, the spin lifetime of an accumulation layer would be much shorter than for a depletion layer, and this might mean the advantage of better resistance mismatch is destroyed.

In this work we investigated three high quality undoped InSb films of thicknesses 0.1, 0.3, and 1μm were grown on semi-insulating GaAs (100)-oriented substrates with a 20 nm low temperature (LT) grown InSb buffer, in a VG Semicon V80 MBE system. The improved two-step growth recipe with the LT buffer has been developed in Imperial College in order to reduce the influence of the defects originating at the interface of the InSb films [Zhang (2004a)]. The samples were then annealed at a high temperature (HT). An additional investigation was performed to optimise the temperature of the HT second growth step, which was found to be 380°C [Zhang
a remarkable improvement of the transport properties has been reported for these samples. The samples are described in Table 3.4.

In order to obtain the mobility $\mu$ and carrier concentration $n$, Hall measurements were performed on 5mm diameter clover-leaf Van der Pauw patterns in the temperature range from 77 to 300K.

Although the density and mobility profiles are expected to be gradually varying, for simplicity InSb films grown on GaAs have been considered as consisting of three regions shown in Figure 4.5: a surface depletion region, a low mobility interface, and a high mobility bulk-like region. Because of the strong dependence of the spin relaxation processes on the mobility and carrier concentration, the transport properties and spin dynamics are different in these layers.

![Figure 4.5 Schematic of the structure of the InSb thin films](image)

The analysis of the mobilities and carrier concentrations uses the standard multi-layer Hall effect technique, derived from the basic 2-layer method of Petritz [Petritz (1958)]. In this model, the combined effective sheet carrier density, $n_s$, and average effective mobility, $\mu$, are related to the sheet densities and mobilities in the $i$ component layers, $n'_s$ and $\mu_i$, by:

$$n_s \mu = \sum_i n'_s \mu_i,$$

Eqn 4.1
and

\[ n_s \mu^2 = \sum_i n_s^i \mu_i^2 \]  \hspace{1cm} \text{Eqn 4.2}

A detailed investigation of the transport properties of this set of high quality InSb films of different thicknesses is presented elsewhere [Zhang (2004a,b), Harris (2004)]. It was found that the transport properties of the films are well described by a two-channel conductivity model considering only the interface and bulk-like region [Harris (2004)]. In order to deconvolute the depth dependence of the mobility and carrier concentration from any pair of films in the set, it was assumed that the properties of the thinner film and the region of the same thickness of the thicker film are the same. Thus the properties of the remaining top region of the thicker film can be found differentially by means of the standard two-layer Hall formulae Eqn 4.1 and Eqn 4.2. The differential mobility and carrier concentration of these layers of the InSb films at room temperature are presented in Table 4.1. Note that the layers \( i = 0, 1, 2, \) and 3 do not intentionally correspond to the interface, bulk and surface regions discussed above, they simply represent the average carrier density and mobility over a specific region of the film depth. However, Poisson-Schrödinger modelling of the band structure of these layers (see below the inset Figure 4.8c) shows that the interface accumulation layer thickness and surface depletion depth (for low-doped material) are about 50nm and 100nm respectively, so that to a good approximation the interface accumulation region consists of the LT layer \( (i = 0) \) and first layer \( (i = 1) \), and the surface depletion region is a small fraction of the third layer \( (i = 3) \), the rest of which, along with the second layer \( (i = 2) \), are in the bulk-like intrinsic region. The latter is not quite homogeneous and in fact shows a gradual improvement in properties with increasing thickness. The sheet conductivity of each layer is equal to the product \( \mu \cdot n \cdot d \), where \( d \) is the thickness of the channel, and thus the ratio of the current carried by each channel to the total current through the whole film can be estimated. As one can see from Table 4.1, in the thickest film (1\( \mu \)m) the interface (sum of \( i = 0 \) and \( i = 1 \) layers) of the InSb film, which we have found to have a short spin lifetime (see below), carries not more than 7.5\% of the total current. Over 92\% therefore flows through the bulk-like material, which is much greater than for the InAs films [Litvinenko (2006b)], where for the same thickness 10\% of the current flows through the interface region and 60\% flows in the surface accumulation layer. Clearly in thinner films the proportion of current carried in the accumulation layers is greater,
and this accounts for the change in relative strengths of the observed lifetimes in the samples of different thickness.

Table 4.1 The depth-dependent electrical properties of the layers of the InSb films deduced from the differential model. \( i \) is the number of the layer (\( i=0 \) corresponds to the LT buffer layer); \( d \) – the width of the layer; \( x \) – relative amount of the total current flowing through the layer, for a 1\( \mu \)m thick film. The 100nm, 300nm and 1\( \mu \)m films consist of the first two, three and four layers, respectively.

<table>
<thead>
<tr>
<th>( i )</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>( d, \text{ nm} )</td>
<td>20</td>
<td>100</td>
<td>200</td>
<td>700</td>
</tr>
<tr>
<td>( \mu, \text{ m}^2/\text{Vs} )</td>
<td>0.035</td>
<td>0.85</td>
<td>1.5</td>
<td>4.2</td>
</tr>
<tr>
<td>( n, 10^{16}\text{cm}^{-3} )</td>
<td>49</td>
<td>4.9</td>
<td>3.9</td>
<td>1.6</td>
</tr>
<tr>
<td>( x, % )</td>
<td>( \approx 0.5 )</td>
<td>( \approx 7 )</td>
<td>( \approx 18.5 )</td>
<td>( \approx 74 )</td>
</tr>
</tbody>
</table>

4.2.2.2.1 Spin dynamics of InSb thin films

The temperature dependences of the derived mobility of each conducting channel are shown in Figure 4.6. The temperature at which the predicted crossover between EY and DP processes occurs for the non-degenerate case is also shown in this figure. For the degenerate case, which is relevant only for the 0.1 and 0.3\( \mu \)m films, the mobility of all constituent layers is in the EY dominated regime over the whole temperature range. Although the concentration of defects was significantly reduced by the improved growth recipe for distances greater than 100 nm from the LT buffer layer [Zhang (2004b)], the electron mobility is too small to remove EY spin flip scattering and make such thin films suitable for spintronic applications at the room temperature. DP domination is only achieved at distances further than 300nm from the interface.

Spin dynamics of these thin films was studied by a circularly polarized pump-probe experiment for the three films. The energy of the photons was chosen to be close to the band gap in order to increase the penetration, so that we can assume a uniform excitation through the thickness of the films.
Figure 4.6 Temperature dependence of the derived mobility of the layers, deduced by two-channel differential analysis of the set of InSb films: 100nm layer (diamonds), 200nm layer (triangles), 700nm layer (circles) The parameters of each layer are presented in Table 4.1. The crossover mobility between the DP and EY processes (Eqn 2.41) is shown by the dotted line for the case of lattice scattering and nondegenerate statistics, which defines the dominancy of each process for different temperatures. The crossover mobility depends on temperature, here it happens at 175 K, before and after this temperature EY and DP mechanisms dominate, respectively.

As mentioned above, the mobility and carrier concentration gradually change with the depth of the films, nevertheless the evolution of the spin population, shown in Figure 4.7 for all samples can be well described by either a single or dual exponential decay.

From Figure 4.7, it can be seen that the 0.1 \mu m film exhibits single exponential spin decay for the whole temperature range, whereas the thicker films are found to have two exponential decays. The short component of the spin lifetimes has a similar value and temperature dependence for all samples and thus originates from the layers in common, i.e. the interface accumulation region. The intermediate thickness film (0.3\mu m) exhibits two exponential decays, and the long component originates from layer i = 2, i.e. the bulk-like material. At 300 K the long lifetime contribution of layer i = 3 in the thickest (1\mu m) sample is similar to that of i = 2.
Figure 4.7 Time evolution of the spin population measured at 77K (a), and room temperature (b), for undoped InSb films of different thicknesses: 0.1μm (green triangles), 0.3μm (pink circles), 1μm (blue squares)

However, at 77K there is a clear difference in the long lifetime components of the 0.3 μm and 1 μm films, and there is some evidence of the $i = 2$ layer decay at short delay times in the signal from the 1 μm sample.

The temperature dependences of the spin lifetime of the 0.1 μm film as well as the long components of the spin lifetime of the other two samples are shown in Figure 4.8. The spin lifetime temperature dependence of the 4 μm bulk-like InSb, also shown in Figure 4.8c demonstrates that the top layer of the 1μm film exhibits similar properties.
Figure 4.8 The temperature dependence of the spin lifetime from the population decay curves. In the case of 0.3 μm and 1 μm films only the long components are shown. Also shown are the theoretical calculations of the spin lifetime (combination of DP and EY mechanisms) for degenerate (dashed line), non-degenerate (dotted line), and Fermi-Dirac statistics (solid line). The experimental spin lifetime of an undoped 4μm-thick InSb film is shown by solid diamonds in (c). The Poisson-Schrödinger modelling of the band structure of the 1 μm film is shown in the inset (the numbers correspond to the HT layers in Table 4.1). Because of dominance of the surface effects in the thin films, the spin lifetimes illustrate nearly similar results in samples 0.1 and 0.3 μm, while the thicker film, 1μm, shows comparable result with the bulk-like sample (4μm).
The mobility and carrier concentration of layers \( i = 1-3 \) were used to calculate the spin lifetimes using degenerate and non-degenerate approximations; the degenerate and non-degenerate limits are recovered by substituting \( E_k \) by the Fermi energy \( E_F \) or the thermal kinetic energy \( k_B T \), respectively, (refer to section 2.2). As expected from the mobility analysis, the spin lifetimes for 0.1 and 0.3 \( \mu m \) films are well described by the EY relaxation mechanism (see Figure 4.8a,b). The DP process predicts a spin lifetime two orders of magnitude larger for these samples. For the 1\( \mu m \) film the DP process dominates at room temperature, and the EY at low temperatures.

For the top layer of the 0.1 \( \mu m \) (0.3 \( \mu m \)) film the Fermi energy is approximately 26 meV (22 meV) at 77 K, but drops down to a few meV at room temperature. Therefore the transition from degenerate to nondegenerate statistics is within this temperature range. Both degenerate and nondegenerate cases are presented in Figure 4.8a,b. In order to obtain the spin lifetime in the intermediate regime, the average \( \langle E_k^m \rangle \) where \( E_k \) is the electron kinetic energy is used to calculate the spin lifetime governed by both the DP \((m=3)\) and EY \((m=2)\) processes.

\[
\langle (E_k)^m \rangle = \frac{\int(E_k)^m f(E_k)\rho(E_k)dE_k}{\int f(E_k)\rho(E_k)dE_k} \quad \text{Eqn 4.3}
\]

where \( f(E) \) is the Fermi function, and \( \rho \) is the density of states. The result of this calculation is shown in Figure 4.8 by solid lines. Only the non-degenerate case is shown for the 1\( \mu m \) film. The values of \( A \) (Eq 2.35) and \( Q \) (Eq 2.37) were used as the only fitting parameters and found to be \( A=18 \) for the 0.1 \( \mu m \) film (Figure 4.8a); \( A=9.5 \) for the 0.3 \( \mu m \) film (Figure 4.8b); and \( A=9.5 \) and \( Q=1.5 \) for the 1 \( \mu m \) film (Figure 4.8c).

In order to estimate the spin lifetime of the low mobility, high carrier concentration LT buffer layer \( i = 0 \) the degenerate EY relaxation mechanism was used, as shown in Figure 4.8a by dash-dot line. In spite of such a short spin lifetime, the LT buffer layer carries only 0.5% of the total current and therefore is not expected to affect the transport and spin properties of the films.
It should be mentioned that the spin lifetime in intrinsic bulk InSb under high intensity laser excitation using a free electron laser (FELIX) for spin population creation, which has reported to be 12 ps at room temperature [Murdin (2006)], is three times larger than the spin lifetime presented in Figure 4.8c for the sample ME1655 under weak excitation. The peak energy of FELIX was about 1 μJ. Such intense laser pulses generated a significant photohole density, which more than doubled the spin lifetime [Litvinenko (2006b)]. The peak energy of the laser pulses used in the experiment presented here was 10nJ. The density of photoholes generated by such pulses is negligibly small in comparison with the FELIX experiment, and the measured spin lifetime is therefore the true electron spin DP lifetime, which was estimated to be about 6 ps or less [Litvinenko (2006b)]. This value agrees very well with the 4 ps room temperature spin lifetime obtained in the present work and proves that with low energy laser pulses the real electron spin lifetime may be measured without the influence of photoholes.

### 4.2.3 Comparison of InAs and InSb thin films

Our results for InSb thin films are compared with the information about InAs films from [Litvinenko (2006b)], refer to 3.3.2.2.

InAs uniquely has electron accumulation in the near-surface region [Heine (1965)]. In contrast, a low-barrier depletion region is formed on the surface of the InSb films (refer to Figure 4.10). The difference between InAs and InSb is that the defect levels are located within the conduction band and in the band gap, respectively. The InAs surface accumulation layer is very useful for fabricating nanometer-scale conducting semiconductor structures, nanowires [Thelander (2003)], and nanoelectromechanical systems [Yamaguchi (2002)]. However, its sub-picosecond spin lifetime could rule out the usefulness of InAs for spintronic applications. On the other hand, InSb films have all advantages of NGS materials, but without the complication of the surface accumulation layer.
We assume that both InAs and InSb films (grown on GaAs) consist of three regions: (i) the interface layer (with high carrier concentration and low mobility); (ii) the bulk-like layer (with low carrier density and high mobility); (iii) the surface accumulation layer for InAs with high carrier concentration and high mobility and the surface depletion layer for InSb (Figure 4.9).

![Figure 4.9 Schematic layers of InAs and InSb films](image)

In InAs an accumulation surface layer carries most of the current [Litvinenko (2006b)], whereas for InSb the influence of the depletion surface layer on the transport property could be ignored [Litvinenko (2007)].

Theoretical band-bending profiles for the 1 μm films at 300K including the accumulation layer in InAs surface and the depletion layer in InSb surface has been shown in Figure 4.10.
The DP mechanism dominates in both materials at room temperature in the bulk-like region. In the interface layers and surface layer the EY mechanism dominates for the whole temperature range. (The magnitude of the mobility at the cross-over between EY and DP (see the dashed line in Figure 4.6) for the ionized impurity scattering case is more than two times the mobility required for the lattice scattering case).

For InAs films it was found that the surface electron Fermi energy is estimated to be about 300meV above the band edge, which leads to a large Moss-Burstein shift in the absorption edge, and the impossibility of optical creation of a spin population in that part of the InAs samples.

Room temperature results of the films are gathered in Table 4.2. The current carried by each layer is proportional to $\mu nd$, so that the fraction of the current flowing through each layer can be estimated. It was found that more than 60% of the current flows through the surface layer of the InAs films. These films have short spin
lifetimes, which are controlled by the degenerate EY process. Although in InSb bulk-like region the spin lifetime is 4 times less than in InAs bulk-like regions, the fraction of current flowing through such regions is considerably higher (92.5%). Therefore, we infer that InSb could be a more useful for spintronic application than InAs.

**Table 4.2** The depth-dependent properties of the InSb and InAs 1μm films deduced from the differential model. \( d \) is the width of the layer, \( x \) is relative amount of the total current flowing through the layer, and \( \tau_s \) is the spin lifetime. For InSb film the property of the LT buffer layer is also shown.

<table>
<thead>
<tr>
<th>layers</th>
<th>InSb</th>
<th>InAs</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>LT buffer</td>
<td>Interface HT</td>
</tr>
<tr>
<td>( d, \text{nm} )</td>
<td>20</td>
<td>100</td>
</tr>
<tr>
<td>( \mu, \text{m}^2/\text{Vs} )</td>
<td>0.035</td>
<td>0.85</td>
</tr>
<tr>
<td>( n, 10^{16} \text{cm}^{-3} )</td>
<td>49</td>
<td>4.9</td>
</tr>
<tr>
<td>( \tau_s, \text{ps} )</td>
<td>&lt;0.017</td>
<td>0.56</td>
</tr>
<tr>
<td>spin relaxation process</td>
<td>EY</td>
<td>EY</td>
</tr>
<tr>
<td>( x, % )</td>
<td>\approx0.5</td>
<td>\approx7</td>
</tr>
</tbody>
</table>

The spin lifetime in the interface layer is nearly one order of magnitude less than in the bulk-like region, but this layer carries not more than 10% of the total current. Because of the high carrier concentration and consequently large Fermi energy in the surface accumulation layer of InAs films, the spin lifetime in the layers is estimated to be less than 30 fs (laser pulse-width), while 60% of the whole current flows through the surface layer of the 1 μm InAs film. The EY spin relaxation mechanism dominates in the surface and interface regions of the both InAs and InSb films. In the 1 μm InSb films the bulk-like region carriers more than 92% of the whole current, the spin lifetime is in the picosecond range and governed by the DP process, which makes it a more attractive candidate for spintronic application.
4.2.4 QWs

In this work, we have studied spin dynamics of undoped indium antimonide single n-InSb/AlInSb QWs grown by MBE on GaAs substrates with different well widths (first three samples of Table 3.5). Naturally, the different samples also have different carrier concentration and mobility, and this also affects the spin life time measurement. The consideration of only the well-width effect can not be achieved easily. Even when studying the temperature dependence of a single sample, \( \mu \) and \( n \) change as the temperature is changed, making comparison difficult. However, since generally speaking it is desirable to have high concentration and mobility for eventual spintronic devices it is important to try to study the possibility of control of the spin lifetime via other parameters such as the well width (which enters into the formulas for both EY and DP via the confinement energy \( E_{1e} \) in Eqns 2.36 and 2.40).

The mobility of all doped QWs was measured by means of the Hall effect. Unfortunately, because of low carrier concentration in undoped wells, it was not possible to measure the electron mobility by Hall effect.

The typical result of optical polarisation versus time delay for well and barrier at 3 and 5 \( \mu m \) for temperature between 77 and 293 K has been presented in Figure 4.11.

![Figure 4.11](image)

**Figure 4.11** Time evolution of the spin population for different temperatures, **left**: QW 20 nm width (ME1831) at 5 \( \mu m \), \( \tau \) (RT) ~ 0.5 ps, **right**: barrier at 3 \( \mu m \) (ADJ1129)
Because there are two different materials with different bandgaps in a QW (well and barrier), we should be careful to separate the regions in our measurement. Photons of 3μm wavelength have enough energy to create high energy electrons in the small gap material of the well, and also low energy electrons in the wide gap material of the barrier. Reducing $E_k$ and increasing $E_g$ both have the effect of increasing the spin lifetime (both for EY and DP Eqns 2.36, 2.40) so the barrier exhibits much longer lifetime, and is also much thicker and more easily observed. The control of the excitation laser wavelength allowed us to separate the influence of the barrier material on the spin dynamics in the quantum wells. The luminescence spectra have been measured. For similar material the QW emission line was observed at 5μm, whereas the barrier material radiated at around 3μm. The calculated barrier bandgap wavelength was 3.7μm. The peak in the spin lifetime with wavelength corresponds closely to that of the barrier edge, and the spin lifetime of the QW must be measured at much longer wavelength (Figure 4.12-left). Figure 4.12-right shows the temperature dependence of spin lifetime measured for 30nm-well, barrier, and bulk by using wavelengths 4.5, 3.5, and 5 μm, respectively. As it is clear, $\tau_s$ decreases with temperature, and the barrier shows longer spin lifetime, the bulk and QW (with the same material) show comparable results.

![Figure 4.12](image)

**Figure 4.12** Left: wavelength dependence of spin lifetime at RT, the longer spin lifetime corresponds to the barrier edge; right: comparison of temperature dependence of spin lifetime for 30nm-well, barrier, and bulk using different wavelengths.
It was found that the spin lifetime in the barrier material for all samples is around 5 ps at room temperature, whereas the spin lifetime in quantum wells depends on the well width and the level of doping, and alters from 200 fs for high doped narrow QWs to 2.2 ps for un-doped wide QWs at RT.

The carrier concentration dependency of spin lifetime has been also studied. Figure 4.13-left with results of optical polarization versus time delay for doped and undoped samples, illustrates that decay time for undoped sample is much longer than the doped ones as expected for degenerate spin relaxation, where $E_g \rightarrow E_F$ in Eqns 2.36 and 2.40, (for QW we have $E_F \propto n$), confirms that spin lifetime is inversely proportional to the carrier concentration, Figure 4.13-right.

![Figure 4.13 Left: optical polarization versus time delay for doped and undoped samples, shows higher doping shows shorter decay time; right: concentration dependency of spin lifetime, shows that $\tau_s$ versus $1/n$ is linear.](image)

The results for temperature dependence of spin lifetime for doped and undoped QWs (with different widths) and bulk samples can be seen in Figure 4.14. It is obvious that the doped sample shows shorter spin lifetime and for undoped QWs spin lifetime is longer. The results show that spin lifetime in QW samples depends on doping level and mobility much more than the well-width.
The sample descriptions are shown in Table 3.4. As it can be seen the undoped samples (30- and 40- nm QWs and bulk) show similar results with long lifetimes, while the doped sample 20- nm QW with shorter lifetime, shows very different result, confirming that the effect of doping is more important than well-width.

We would still like to establish the well width dependence. Because $E_{1e} \propto L^{-2}$ (where $L$ is well-width), for EY mechanism from Eqn 2.36 we find that $\tau_s^{EY} \propto L^2$ and for DP from Eqn 2.40 we have $\tau_s^{DP} \propto L^4$; in the latter, from 20 nm to 30 nm width spin lifetime five time increases and from 20 to 40 nm, it gets 16 times longer. So strong increase in spin lifetime can be achieved with small increase in well width, and this increase is more sensitive when the case of DP mechanism dominates. The spin lifetime as a function of well-width is shown in Figure 4.15 at 77 K and RT. However, neither degenerate EY nor degenerate DP satisfactorily explains the results, and this is
again due to the fact that the mobility and density are changing from sample to sample as well as the width.

Figure 4.15 Spin lifetime versus well-width provides a tool for comparison between two relaxation mechanisms at 77 K and RT, showing that neither EY nor DP can be used without consideration of the density and mobility.

In degenerate statistics, $E_F = (\pi \hbar^2 / m^*) n$, for DP mechanism we have $\tau_s^{DP} \cdot \mu \propto (1 / E_F) \propto 1/n$, (Figure 4.16). The proportionality constant contains no explicit dependence on $n$, $\tau_s$, $\mu$, and $T$, but it is well width dependent ($\propto L^4$). Therefore it should be the same for all samples of the same material and well width, for any temperature, doping, or mobility. [NB, the proportionality constant also contains factors of bandgap etc, which are slightly temperature sensitive, though we ignore that for the moment]. A graph of $\tau_s \mu$ versus $1/n$ should therefore give a straight line with slope that is proportional to $L^4$, even if the samples of the same width have different transport characteristics. This is illustrated in Figure 4.16.

We conclude that DP relaxation mechanism dominates for whole temperature range of our experiment and demonstrate the temperature dependence of the spin lifetime. Density and temperature dependences of spin lifetime can be analysed in terms of DP only if density and mobility dependence on temperature is taken into account. The differences in density and mobility of the different wells dominated any dependency on the well width.
4.2.5 QDs

Bulk spin relaxation arises from spin-orbit coupling mediated by phonon or impurity scattering. Relaxation in QDs is different because the electronic states are quantized, which make it more difficult to satisfy energy conservation in QDs, and thus the coherence lifetimes are considerably larger. We started a preliminary investigation of QD spin lifetimes, though the number of samples and experiments performed were too small to have allowed us to make much analysis. A small selection of representative data is shown in this section for completeness. The characterization of the samples used in this work (HgTe and PbSe QDs), have been mentioned in section 3.4.

The spin lifetime in chemically synthesised PbSe QDs has been measured for the first time by a resonant circularly polarized pump-probe experiment. The profile of the differential transmission of PbSe QDs can be well fitted by a two exponential decay. The shorter lifetime \( \tau_1 = (52 \pm 2) \) ps is attributed to the nonradiative decay of free-carriers and their relaxation to luminescent states. The longer decay time \( \tau_2 = (820 \pm 15) \) ps is due to the relaxation of the luminescent or localized states, see Figure 4.17. The relaxation of the spin population is shown in Figure 4.17b and it can also be described by a two exponential decay. The short component is found to be...
(8±3)ps, whereas the long component is of the order of nanoseconds. Two spin lifetimes have already been reported for CdSe QDs, where it was speculated that the biexponential decay could be attributed to the spin relaxation of electrons and holes.

![Graph](image)

**Figure 4.17** (a) Differential transmission of PbSe QDs fitted by a two exponential decay, (b) two exponential decay of spin population relaxation

There are two HgTe QD samples labelled by S45, S46, their photoluminescence spectra have been presented in Figure 3.9. Figure 4.18 shows the optical polarization for S45 at 1.45 µm wavelength, 40 mW power, which gives the spin lifetime of $\tau_s = 1.46$ ps. We also measured the spin lifetime of 2.7 ps for this sample, at 1.1 µm and 35 mW (not shown here).
Figure 4.18 Typical optical polarization for S45 at 1.45 μm, 40 mW, shows short spin lifetime of 1.5 ps.

Figure 4.19 represents the spin and recombination time decays for sample S46. At room temperature with 40 mW power for wavelengths 1.2 and 1.4 μm, spin lifetimes are 4.3, 3 ps, respectively. From the photoluminescence spectrum (Figure 3.9) it can be seen that the wavelengths chosen are at the short wavelength side of the main peak, in order to match with the absorption.

At this range of the wavelength, both HgTe samples show that by increasing the energy slightly (shorter wavelength) the spin lifetimes are slightly increased. However, we measured longer spin lifetime (19 ps) for sample S46 at 1.45 μm and 40 mW, Figure 4.20. It might be because of the better match with the absorption peak of the sample, or alignment, or illuminating different parts of the sample.

Clearly, further investigation is needed to be able to describe the spin lifetime behaviour of these samples at different situations (such as different powers, wavelengths, temperatures, and QD sizes).
HgTe #46
Laser: 1.2 μm (40mW) RT

HgTe #46
Laser: 1.4 μm (40mW) RT

**Figure 4.19** Spin and recombination time decays for sample S46 at 40 mW power, **left**: 1.2 μm wavelength, **right**: 1.4 μm wavelength; fitted by bi-exponential decay functions including two decay times: fast cooling (e.g. by LO phonons) followed by the spin relaxation.

<table>
<thead>
<tr>
<th>Sample</th>
<th>CD/ΔT</th>
<th>Time delay, ps</th>
</tr>
</thead>
<tbody>
<tr>
<td>S46 HgTe</td>
<td>1450nm (40mW)</td>
<td></td>
</tr>
</tbody>
</table>

**Figure 4.20** Typical optical polarization for S46 at 1.45 μm wavelength, 40 mW power; this shows longer lifetime than the previous figure of 19 ps, which might be because of slightly different wavelength, or resulted from a different intensity.
4.3 Conclusion
We have found that among two tested bulk samples n-InAs and n-InSb, the former has longer spin lifetime, which is more suitable for spintronics applications. On the other hand, the surface effects of InAs thin films on InAs reduce the applicability. Investigation of the spin relaxation in undoped InSb films of different thicknesses between 0.1 and 1μm grown on GaAs in the temperature range from 77K to 293K, has shown that the spin relaxation has several components. The longer time (~2.5ps at 293K) is associated with the central intrinsic region of the film. The spin lifetime in the interface accumulation layer was shown to be in the sub-picosecond range. This amount was not measured, however it has been calculated based on knowledge of the density of electrons in the accumulation layer achieved from the other work, \( \tau_s \propto n \). Fortunately, its influence on the properties of the whole InSb film should not be significant, because the LT layer carries less than 1% of the total current in thick films. The rest of the accumulation layer, which was found to have a spin lifetime only a few times shorter than that in the bulk-like region (1 ps), carries less than 7% of the total current and therefore will not considerably affect the properties of thick InSb films either. Whereas investigation on InAs films grown on GaAs, showed that the native surface defect resulted in an additional charge accumulation layer with high conductivity but very short spin lifetime [Litvinenko (2006b)], in InSb layers the surface states introduce a depletion region. We infer that InSb could be a more attractive candidate for spintronic applications than InAs. For InSb films it has been also shown that while DP spin dephasing dominates in the low concentration, high mobility semiconducting region in the centre of the films, in the low mobility high concentration accumulation layers, EY spin-flip relaxation dominates, even at room temperature. The influence of the surface depletion layer was not detected in either the transport or spin properties of the films under investigation.

Spin lifetime of InSb QWs have been also studied. We found out that the dependency of relaxation times on different parameters such as carrier concentration and mobility is much stronger than well width. However, a noticeable effect of well width was observed when we plotted \( \tau \mu \) against \( 1/n \). The change in the slope of this graph with well width agrees with expectation for degenerate DP spin relaxation, which shows that \( \tau_s \propto 1/L^4 \).
For QD samples, different spin lifetimes have been achieved at slightly different wavelengths. In order to get more quantitative information about the timescale and mechanisms of spin relaxation in QDs and to verify the preliminary results presented here, further experiments should be done. The future plan would be more investigation about spin lifetime of QD samples with good qualities at larger range of wavelength and power, at different temperatures, and QDs with different sizes. Then by applying an external magnetic field the g-factor could be also studied.
5 Experiment on time-resolved spin precession

5.1 Introduction

As already mentioned every electron has spin and magnetic moment. According to the Zeeman effect, in the presence of an external magnetic field with strength $B$, the electron's magnetic moment aligns itself either parallel ($-1/2$) or anti-parallel ($+1/2$) to the field, each having a particular energy. The parallel alignment corresponds to the lower energy state, and the separation between two states is $\Delta E = g\mu_B B$, where $g$ is the Lande electron $g$-factor and $\mu_B$ is the Bohr magneton. This equation implies that the splitting of the energy levels is directly proportional to the magnetic field. When a small external magnetic field is applied in the sample plane, the spins injected by the normal incidence pump beam are initially polarized normal to the sample and precess about the magnetic field. The result can be observed as oscillations in $P_{opt}$ which decay with the spin life-time.

The electron Lande gyromagnetic ratio, $g^*$, is a fundamental parameter in semiconductors, and is different from the free electron $g$ factor in vacuum ($g_0 = 2.0023$) because of spin-orbit interaction. This interaction is the key for understanding spin-related phenomena and spintronics, because it determines the strength of effects such as the control of spin polarisation in an applied external magnetic field (and also electric field via the Rashba effect). Precise measurement of spin $g$-factor is expected to offer invaluable information on electronic structure and electron confinement in the quantum structures.

Traditionally, electron $g$-factors have been measured using a variety of spin-resonance techniques but measurements above liquid helium temperature are very difficult due to the very small energy and population differences between the spin split states. For example, temperature dependence of $g$-factor has been measured for lower temperatures, between 4.2 K and 66 K by electron paramagnetic resonance (EPR) for CdTe [Meyer (1998)]. To our knowledge, experimentally, high temperature electron $g$-factor measurement up to RT has been described for GaAs and InP only by spin-quantum-beats of electrons by polarization- and time-resolved near-infrared photoluminescence spectroscopy under applied magnetic field in the sample plane [Oestreich (1995), Oestreich (1996)]. In that experiment a circularly polarised optical
pulse creates a spin polarisation, which precesses about the in-plane magnetic field. The time-correlated photoluminescence has a polarisation that oscillates between the same as the pump and opposite with frequency equal to the Larmor frequency. At the same time the polarisation decays away with the spin-relaxation time. The authors reported that the temperature dependence of the g-factor for GaAs could not be reproduced using standard \textit{k.p} theory with the literature values for the temperature dependence of the bandgaps and interactions.

Because the luminescence efficiency of such narrow gap materials is low and because inexpensive photon counting systems are not available in this wavelength range (mid-infrared), we measured instead transient absorption of circularly polarized light by the pump-probe technique. With these time-resolved methods we can measure spin lifetime ($\tau_s$), and Lande g-factor in the same set-up, even at room temperature (RT).

Modeling of InSb and InAs using \textit{k.p} theory requires only 8 bands [Pidgeon (1966), Weiler (1979)] (i.e. 4 bands each doubly degenerate with two spin directions): the three $p$-like $\Gamma_7$ and $\Gamma_8$ valence bands - heavy hole, light hole and spin-orbit split-off band - and the lowest $\Gamma_6$ $s$-like conduction band) because the energy separation between the $\Gamma_6$ band and the next conduction level is much greater than the fundamental gap $E_g$. Accurate modeling of GaAs on the other hand, where the higher gap is only two times bigger than $E_g$, requires 14 bands [Pfeffer (1990)] (i.e. also including the higher $p$-like $\Gamma_7$ and $\Gamma_8$ conduction bands). Fewer parameters are required for InAs and InSb and it is therefore interesting to investigate the comparison of experiment and theory for these materials, in order to try to resolve the discrepancy observed for GaAs in the literature. Sometimes the 8- and 14-band models are called 3- and 5-band models, because of three and five different energies of the bands, respectively.

In this work we have investigated the Larmor precession of electrons in the NGS materials (InAs and InSb) in the time-domain using a mid-infrared femtosecond pulsed pump-probe method, described in the chapter 3. The results have been used to extract the Lande gyromagnetic ratio, $g^*$, and spin lifetime, $\tau_s$, as a function of temperature from 77 K up to 293 K. We note that the NGS materials under
examination have large, negative g-factor, but that our experiment does not easily
provide the sign.

5.2 Experimental method for measurement of Lande g-value

The Experimental setup is similar to Figure 3.1; the only difference is adding an
external magnetic field, using a pair of permanent magnets mounted inside the
cryostat so that the field is in the sample plane (Voigt geometry). Temperature
dependency of the magnetic field, B(T), which is measured to be around 10%
decrease between 77 K and 293 K, can be in the same scale as our experimental errors,
and so it is neglected in our calculations.

Figure 5.1 shows a typical result with and without a magnetic field. It is obvious that
both situations have the same decay time and provide information about spin lifetime,
but information of precession can be found from oscillatory behaviour only in
presence of B.

![Figure 5.1](image)

**Figure 5.1** The effect of an external magnetic field on time evolution of the spin population.

The optical polarization is defined by Eqn 3.1. In the presence of an external
magnetic field it can be suitably fitted by an exponentially decaying sinusoid function:

\[
P_{opt} = A \exp\left(-t/\tau_s\right) \cos[\omega_L(t - t_0)]
\]

Eqn 5.1

where \(\tau_s\) is the spin lifetime, \(\omega_L\) is Larmor procession frequency:
\[ \omega_L = g^* \mu_B B / \hbar = 2\pi / T_L \]  
Eqn 5.2

Here \( T_L \) is the Larmor precession period, and \( B \) is magnetic field.

5.3 Results- spin precession and decay in non-zero magnetic field

5.3.1 InAs

By applying a Voigt geometry magnetic field of 0.288 T in the sample plane, the experimental result of the transient absorption for InAs (IC311), see Table 3.3, for wavelength 3.1 \( \mu \text{m} \) at 77 K is shown in Figure 5.2, where the spin lifetime is obtained \( \tau_s = 40 \pm 2 \) ps, and \( |g^*| \approx 12 \pm 0.1 \) (negative for InAs) achieved from Eqn 5.1 and Eqn 5.2.

![InAs ic311](image)

**Figure 5.2** The oscillation of spin population versus time delay, for \( B=0.288 \) T at \( T=77 \) K for 4-\( \mu \text{m} \) thick InAs (IC311), curve is fitted by Eqn 5.1.

Figure 5.3 shows the oscillating behaviour of time evolution of the spin population for InAs (IC311) in range of temperature between 77 K and 293 K. As it can be observed the oscillation frequency doesn’t show significant change at different temperatures. The \( g \)-factors calculated from the Larmor precession periods of the oscillations in the range of temperature among 77-293 K are also shown. From the experimental results we conclude that when temperature increases, spin life-time decreases, however the change in \( g \)-factor in this range of temperature and for low magnetic field \( B=0.288 \) T
is not very considerable. According to Oestreich et al. [Oestreich (1996)], for B<5T, the change in the g-factor versus B is approximately negligible for different temperatures.

![Diagram of InAs IC311](image)

**Figure 5.3** Left: The transient polarisation precession and decay (without showing the fitting) for InAs (IC311) at B=0.288 T, as a function of temperature for InAs at $\lambda \sim 3.1 \mu m$. The curves have been offset for clarity. Right: Electron absolute g-factor versus temperature from fits of Eqn 5.1 to transients at left.

### 5.3.1.1 Discussion of g-values in InAs in non-zero magnetic field

In an externally applied magnetic field the electron energy, effective mass and Lande g-factor represented by Eqn 2.19, Eqn 2.31 to Eqn 2.32 show that they are sensitive to the band gap, and give the approximate dependence of $m^*(E_g)$ and $g^*(E_g)$. The temperature dependence of the effective mass and g-factor, $m^*(T)$, and $g^*(T)$ (shown in Figure 5.4) can both be calculated from the Varshni form of $E_g(T)$ described in Eqn 5.3, assuming other band parameters are constant.

The other important parameter in calculation of $m^*$ and $g^*$ is the spin-orbit splitting, which is relatively independent of temperature since it represents an interaction that takes place deep within an atom [Koteles (1974)].

The only remaining parameter, which does not have a direct experimental access, is the interband momentum matrix element, whose temperature dependence has been assumed to be unimportant. According to the Cardona-Lawaetz model [Lawaetz
it is expected that the momentum matrix elements would be inversely proportional to the lattice spacing. Also, the less precise tight-binding calculations [Graf (1995)] show that $E_p$ depends on $1/d^2$ (d is the lattice constant and increases with increasing temperature) [Harrison (1988)]. Since the lattice constant of the materials studied here changes only by a few % from 4K to 300K, the fractional change in $E_p$ is small.

In Figure 5.4, we have calculated the band-edge value of $m^*(T)$ and $g^*(T)$ using both the three-band model (Eqn 2.31, Eqn.2.32, with $N_1=F=E=0$), and five-band model (Eqn. 2.33, Eqn. 2.34); comparison shows that the difference between the two models is not significant, confirming that higher bands have an insignificant effect. Our calculations show that introducing the extra conduction band in five-band model, decreases $(m_0/m^*)^{-1}$ by $\approx 10^{-3}$, and decreases $(g^*/g_0)^{-1}$ by $\approx 10^{-4}$, as shown in the figure. For samples with $\Delta \geq E_g$, such as InAs ($\Delta = E_g$), and InSb ($\Delta >> E_g$) the higher conduction bands make a negligible correction (although of course for wide gap semiconductors (WGSs) such as GaAs it should be considered).

As it can be seen, the results obtained by $k.p$ theory using temperature dependency of optical energy gap and our experimental data for absolute $g$-factor at different temperatures give reasonable agreement at low temperature, but very poor agreement at high temperature. The discrepancy at 293 K for InAs is about 55%, and the theory and experiment are divergent.
Figure 5.4 Temperature dependence of experimental left: absolute g factor for InAs (data taken from Figure 5.3), right: effective mass (data taken from [Stradling (1970)]). Also shown is the theory from three- and five-band k,p model using the parameters listed in Table 3.1; all band parameters are constant except for optical energy gap, $E_g$, which follows the Varshni form (see Eqn 5.3).

For effective mass, comparison between the results obtained by k,p theory using temperature dependency of optical energy gap and experimental data for effective mass for different temperatures from [Stradling (1970)], is also presented in Figure 5.4. It shows that theory and experiment have the same trend with only about 10% percent discrepancy in the magnitude with better agreement at lower temperatures. It has been reported that for InAs the percentage change in effective mass with temperature from magneto-phonon experiments is 7.5%, from dilational part (see next section) is 2%, and from optical energy gap is 12%; with InAs the estimated change from dilational part is about a factor of two less than that observed but the observed change in mass is clearly less than that found by the optical energy gap [Stradling (1970)].

Assuming we have exact experimental results, more consideration of k,p model is needed to explain this discrepancy between theory and experiment. There are some possibilities, which we can investigate to find out the reason of this discrepancy. We note that the k,p model has been defined for band edge parameters at low temperature ($k=T=0$), which are different of energies far from band edge, and higher temperatures. Temperature dependency of the other parameters except for $E_g$ has been ignored, which might be incorrect at higher temperatures. In the presence of non-parabolicity,
the results for energies far from band edge are different from band edge, so study of
the energy dependency of parameters such as g-factor might be useful. Furthermore,
we have considered the temperature dependence of band gap, $E_g$, as measured
optically, and it has previously been suggested that there are two contributions to this
– the dilational and vibrational contributions. It has been shown that the effective
mass is only sensitive to the dilational part, while the optical energy gap would
erroneously predict much larger changes in effective mass [Stradling (1970),
Shantharama (1984), Adachi (1992)]. It may be the case that the g-factor also follows
only the dilational bandgap. In the next sections we investigate these possible effects.

5.3.1.2 Temperature dependency of band parameters

5.3.1.2.1 Band gap

The temperature dependence of the band gap is assumed to follow the Varshni form
[Varshni (1967)]:

$$E_g = E_g(0K) - \frac{\alpha T^2}{T + \beta}$$

Eqn 5.3

It is known that the optical energy gap determined from optical measurements at the
fundamental edge [Panish (1959)] decreases with increasing temperature because of
lattice dilation and electron-phonon coupling. The temperature dependence of the
energy gap may be written by [Paul (1963)]:

$$\delta E_g = \delta T \left( \frac{\partial E_g}{\partial T} \right)_V + \delta V \left( \frac{\partial E_g}{\partial V} \right)_T$$

$$\frac{\partial E_g}{\partial T} = \left( \frac{\partial E_g}{\partial T} \right)_V + \left( \frac{\partial V}{\partial T} \right)_P \left( \frac{\partial P}{\partial T} \right)_V \left( \frac{\partial E_g}{\partial P} \right)_T$$

Eqn 5.4

where, the second term is the dilation part ($E_{\text{dil}}$) due to the change in the average
lattice constant, and the first term, vibration part ($E_{\text{vib}}$), is due to electron-phonon
interactions. In Eqn 5.4 $B$ is the volume expansion coefficient and $K$ is the
compressibility, and \( dE/dP \) is the pressure-induced band-gap shift (see Tables 3.1, 3.2). [Notice that, only in this section \( P \) is used as the symbol for pressure.]

The variation of thermal expansion can be written as [Hang (1991), Malikova (1996), Biernacki (1994), Lautenschlager (1985)]:

\[
\Delta E_{th}(T) = -3 \frac{1}{K} \left( \frac{\partial E}{\partial P} \right) \int_{T_0}^{T} \alpha_{th}(T')dT'
\]

Eqn 5.5

where \( \alpha_{th}(T) = B/3 \) is the linear thermal expansion coefficient. The coefficients of linear thermal expansion for InSb up to 300 K has been reported in [Gibbons (1958)], and for InAs for low and high temperatures have been accounted in [Sparks (1967)] and [Sirota (1959)], respectively. Additionally, the temperature dependency of lattice constant (the integral in Eqn 5.5) for InAs has been given in ref [Tsay (1972)]. The parameterised forms of \( \alpha(T) \) for InAs and InSb are shown in Figure 5.5.

![Figure 5.5](image)

---

It has been shown that the variation of energy gap with temperature is largely due to electron-phonon interaction rather than thermal expansion for some samples such as GaAs [Adams (1957)]; it can be also seen for InAs and InSb in Figure 5.6.
By considering the temperature dependency of the dilational part of energy gap, the difference between experimental and theoretical $g^*(T)$ and $m^*/m_0(T)$ would be decreased at higher temperatures. These results for InAs, which have been presented in Figure 5.7, show much improvement in agreement between experimental and theoretical results. By using the temperature dependency of the dilational part of energy gap, the difference in g-factor and effective mass by temperature is considerably decreased compared to the optical gap.
Note, the experimental g-value initially rises in magnitude with temperature and then falls. Interestingly, the thermal expansion coefficient also has a negative and then positive trend (Figure 5.5), though in the latter case the anomalous range is at much lower temperature. In a related material, Al$_x$Ga$_{1-x}$As alloy, in the temperature range from ~23 to ~95 K there is a competition between the negative thermal expansion (NTE) effect and the electron-phonon interaction [Loureno (2004)]. We speculate that there might be a link between the negative thermal expansion (NTE) and the rise and fall in g-value. Another speculation is that it might be because of the effect of high doping in the sample, so that the experiment doesn’t give the proper value of the band edge g-factor.

5.3.1.2.2 Temperature dependency of $E_p$ and $\Delta$ band parameters for InAs

We now consider the possible temperature dependency of other parameters in k.p model, such as spin-orbit splitting energy, and interband momentum matrix elements.

We take the experimental g-factor and the variation of $E_g$ from either the optical or dilational change, and rearrange the Eqn 2.32 to find $\Delta$. In this case we put $N_1=E=0$ and assume $E_p$ is constant. If we consider the temperature dependence of the spin-orbit splitting, $\Delta(T)$, from k.p three-band model, with temperature dependent optical energy gap, we find that $\Delta$ has an unacceptably large variation (Figure 5.8).

The experimental results for spin orbit splitting measurement have been mostly done for low temperature, though for other materials $\Delta$ has negligible experimental change with T. The values of $\Delta$ inferred from our measurement show nearly 50% variations with T and we therefore believe that a temperature variation of $\Delta$ along with the optical gap cannot be the explanation for the discrepancy in Figure 5.4. Using the dilational energy gap will decrease this variation to 20% still rather large.
Figure 5.8 **Left:** The temperature dependence of spin-orbit splitting energy ($\Delta$) inferred from our measured g-factors, using the three band model, for temperature dependent optical energy gap, and $E_p = 21$ eV. Also shown is the low temperature experimental value from modulated reflectivity [Landolt (1982)]. **Right:** comparison between $\Delta(T)$ using temperature dependency of optical gap and dilation gap.

Instead of allowing spin orbit splitting to vary with temperature, we might allow the $E_p$ to be temperature dependent. Although, in the k.p calculation the interband momentum matrix elements are assumed to be temperature independent, if we allow it to be a free parameter (and fixing $\Delta$ this time) and fit to our experimental g-factors, it decreases with temperature (Figure 5.9). It is seen that temperature dependence of $E_p$ using our experimental $g^*$ results is more than that found from the experimental $m^*/m_0$ results taken from [Stradling (1970)]. As already mentioned, theoretically $E_p$ depends on $1/d^2$ and changes no more than 0.1% from 0 to 293 K. This is not consistent with the large amount of change in the optical gap $E_p$ with temperature ($\sim 30\%$). With the dilational gap this variation is decreased ($\sim 12\%$) but it is still much larger than the lattice expansion.
5.3.1.3 Energy dependency of g-factor for InAs

We have measured the energy dependence of the g-factor by tuning the laser photon energy. The results are shown in Figure 5.10 for InAs at different temperatures. The figure also shows the energy dependency of absolute g-factor achieved by $8 \times 8 \textbf{k.p}$ (from Chapter 2 and Appendix C, and Figure D1 of Appendix D, and the predictions of Eqn 2.32). The theory lines were calculated using the optical and dilational bandgap with fixed $\Delta$ and $E_p$. Despite, having a few experimental data, the agreement between theory and experiment is satisfactory. The $8 \times 8 \textbf{k.p}$ results show that energy dependent g-factor obtained by Eqn 2.32 is reasonably acceptable. The better agreement between experiment and theory with dilational gap can be observed at low energy and high temperature (although we need more experimental data for RT at low energy, close to the band edge). It is obvious that g-factor changes with energy, an evidence for non-parabolicity of conduction bands in n-InAs sample. It should be noticed that non-parabolicity causes a reduction in the magnitude of $g^*$ with increasing energy above gap because of dephasing of spin precession. Previously, it has been reported that owing to the strong conduction band non-parabolicity in InAs, considerable care must be taken to measure the mass at the band edge rather than at the Fermi level [Washburn (1979)]. Because our sample is highly doped, the Fermi
energy is rather high. This might mean that even when our laser photon energy is tuned near the band edge, the average electron energy is near the Fermi energy. If this is the case, the data points at low excess energy and low temperature should be moved upwards. It is clear that this would improve the data by removing the knee which appears at low energy and low temperature, and may be important.

Figure 5.10 Energy dependency of absolute g-factor for InAs achieved by experiment, 8×8 k.p, and Eqn 2.32 (N_i=0) with optical and dilational gap at different temperatures, (note that for 8×8 k.p, dilational gap has been shown for 77 and 293 K). It is assumed that because of the high level of doping, the real absolute g-factor for low energy should be larger, results in similar slope with theories.
We show for comparison the theoretical temperature dependence of $g^*(E)$ on a single set of axes in Figure 5.11.

![Figure 5.11 Energy dependency of absolute g-factor for InAs achieved by 8 x 8 k.p (optical gap).](image)

5.3.2 InSb
The Larmor precession of electrons in the InSb (ME1655), Tables 3.4, has been investigated with the same experimental setup. By applying a magnetic field of 0.288T, typical transients for different temperatures are shown in Figure 5.12-left. The shapes of the transients are well described by a simple exponentially decaying sinusoid; the measured polarization precesses coherently at Larmor frequency while decaying with the spin lifetime. Figure 5.12-right shows the g-factors achieved from Larmor periods of InSb at different temperatures. As it can be seen, the difference in absolute g-factors at different temperatures is not significant.
Figure 5.12 Left: The transient polarisation precession and decay for B=0.288 (T) as a function of temperature for InSb at $\lambda \sim 5$ $\mu$m. The curves have been offset for clarity. Right: Temperature dependency of absolute g-factor for InSb (ME1655).

Notice that at each temperature the wavelength was tuned to the band edge. The experimental sensitivity was limited by the electron spin lifetime at short wavelength (3 $\mu$m), and also by the sensitivity of a cooled InSb detector (5 $\mu$m) at long wavelength (see Figure 5.13). Because of limitation of our cooled InSb detector for band-gap energy measurement of high-temperature InSb, we have used MCT (Mercury Cadmium Telluride) detector, for this sample, which for $\lambda > 5$ $\mu$m is more sensitive.

Figure 5.13 Typical laser spectrum; the black line shows the InSb band edge wavelength versus temperature.
5.3.2.1 Discussion of g-values in InSb in non-zero magnetic field

The temperature dependence of experimental and theoretical g-factor and effective mass, from the Varshni form of optical $E_g(T)$, assuming other band parameters are constant, can be seen in Figure 5.14. The parameters are from Table 3.2 (but $\alpha = 6 \times 10^{-4} \text{ eV K}^{-1}$, $\beta = 500 \text{ K}$). As it can be seen, for InSb the experimental and theoretical absolute value of the g factor versus temperature give reasonable agreement at low temperature, but very poor agreement at high temperature and the theory and experiment are divergent. The discrepancy at 293 K with optical gap is ~55%, and using the dilational energy gap will decrease to ~24%.

Figure 5.14 Left: Temperature dependence of experimental absolute g factor for InSb (data from Figure 5.12), right: Temperature dependence of experimental effective mass (data for effective mass is taken from [Adachi (1992)]). Also shown is the theory from three band k.p model using the parameters listed in Table 3.2; all band parameters are constant except for optical energy gap, $E_g$, which follows the Varshni form (blue lines), and the dilational part only (green lines).

It can also be seen that the theory and experiment effective mass show the same trend with only about ten percent discrepancy in the magnitude with better agreement at lower temperatures. It has previously been remarked that for InSb the percentage change in effective mass with temperature from magneto-phonon experiments is 9%, from dilational part is 7.5%, and from optical energy gap is 22%; i.e. the estimated dilational change in effective mass agrees well with the change deduced from the magneto-phonon experiments [Stradling (1970)]. By consideration of temperature
dependency of the dilational gap the difference between experimental and theoretical 
g*(T) and m*/m_0(T) are decreased at high temperatures. This can be seen in Figure 5.14.

5.3.2.2 Temperature dependency of band parameters for InSb

Again, we turn to the possibility of temperature dependence of the other parameters in the theory (spin-orbit splitting, Δ, and interband momentum matrix elements, E_p). Since the spin-orbit splitting for InSb is reported relatively independent of temperature, we consider the temperature dependence of E_p, calculated from lattice constant, our experimental g-factors and experimental m*/m_0 [Adachi (1992)], for temperature dependent optical and dilational gap, shown in Figure 5.15. It is seen that temperature dependence of E_p using our experimental g-factor results is stronger than that of experimental m*/m_0 results. As we mentioned the momentum matrix elements depends on 1/d^2, the temperature dependence of the lattice constants for our sample is much less (∼ 0.17%) and can not explain this amount of change in E_p with temperature (∼ 44% with g* and ∼ 14% with m* - with optical gap). Again, by considering the temperature dependency of dilational gap, this variation is reduced to 20% for g* and 6% for m*.

Recently, the strong decrease of P^2 and therefore E_p with temperature has been calculated for GaAs by not only considering the contribution of the lattice expansion to P^2 but by regarding the effect of fluctuations in the lattice spacing due to thermal phonons [Hubner (2007)]. The microscopic origin of decrease of P^2 is suggested to be the analogue to the shrinkage of the electronic band gap with temperature [Allen (1976)].
5.3.2.3 Energy dependency of g-factor in InSb

Typical transients for different excitation wavelengths at 77K with an applied magnetic field of 0.288T are shown in Figure 5.16. The spin lifetime as a function of the excess optical energy is also shown in the inset. The measured period of the oscillation becomes longer for excitation energies above the band gap illustrating the reduction of the absolute value of the effective g-factor with the electron energy due to non-parabolicity.

The macroscopic polarisation decays because spins of different quantum numbers $n$ or $k_B$ precess at different rates due to the non-parabolicity of conduction bands $g^*$, so the coherence is lost by dephasing. Increasing the pump photon energy increases the spread of electron energies and therefore reduces the coherence time, as shown in the inset of Figure 5.16. This is slightly different in the detail though not in concept from the usual reduction in spin coherence time with Fermi energy according to the DP model [D’Yakonov (1971), D’Yakonov (1972)] in zero field, because in that case the energy-dependent spin splitting responsible for the dephasing is the additional term proportional to $k^2$ due to inversion asymmetry, which increases with energy [Ivchenko (1977), Olesberg (2001)]. In our non-zero field case the energy dependent splitting...
arises from the non-parabolicity, and decreases with energy. In both cases increasing the energy spread results in faster decoherence.

![Figure 5.16](image)

**Figure 5.16** Measured spin precession as a function of time, taken using the circularly polarised pump-probe technique at 77 K and in an applied Voigt geometry magnetic field of 0.288 T. The optical excitation was of different wavelengths/photon energies: 5\(\mu\)m=250meV (blue squares), 4.3\(\mu\)m=290meV (red circles), and 3.2\(\mu\)m=390meV (green triangles). The spin lifetime taken from the decay of the envelope as a function of the excess optical energy is shown in the inset.

The effective g-factor obtained from the transients against excess optical energy above the band-gap is plotted in Figure 5.17. It shows the energy dependency of the g-factor for InSb at different temperatures, obtained experimentally and theoretically from Eqn 2.32 with dilational and optical change of bandgap. As it can be seen from the figure, Eqn 2.32 is reasonably consistent with the experimental data without any fitting parameters. Clearly the data of Figure 5.17 at high temperature (particularly at low energies) is in better agreement with the theory including only the dilational band gap, in accord with the previous work on effective mass.
Figure 5.17 Dependence of the absolute g-factor on photon energy at different temperatures for InSb, experimental data shown by red squares, the theoretical dependence from Eqn 2.32 ($N_i=0$) is shown as a green line (dilational change of bandgap) and blue line (full optical bandgap).
For comparison we illustrate the theoretical temperature dependence of $g^*(E)$, achieved by $8 \times 8 \mathbf{k.p}$ (optical gap) on a single set of axes in Figure 5.18.

![Figure 5.18](image)

**Figure 5.18** Energy dependency of absolute g-factor for InSb ME1655 achieved by $8 \times 8 \mathbf{k.p}$ model (optical part).

Also, for comparison absolute g-value obtained experimentally, and theoretically by Eqn 2.32 and $8 \times 8 \mathbf{k.p}$ model considering both optical and dilational gaps, for 77 and 293 K is presented in Figure 5.19. The results show that Eqn. 2.32 is in good agreement with $8 \times 8 \mathbf{k.p}$ model and satisfies NGS g(E) calculation.
Figure 5.19 Energy dependency of absolute g-factor for InSb ME1655 achieved by experiment, Eqn 2.32 and 8x8 k.p model – including dilational and optical gap for 77, 293 K.

As it can be seen the 8x8 k.p model with no adjusted parameters is quite consistent with the experimental data at low energy.

It is apparent that particularly for higher temperature and lower energy, calculation by dilational gap is closer to the experimental data.

It should be mentioned that using a linear energy dependence for $g^*$ is a poor approximation, and the 8-band theory departs 15% from such a linear dependence $kT$ above the band edge at 300 K.
5.3.3 QW and Comparison with bulk

For our QW samples with the same experimental set-up, we have measured g-factor. Figure 5.20 represents an oscillatory behaviour of time evolution of spin polarization for 30 nm QW sample ADJ1129 (Table 3.5). The g-factor is calculated from the Larmor period. The spin life-times of 10 nm (ADJ1127) and 20 nm QW (ME1831) were too short to be able to see the oscillation and g-factor measurement.

**Figure 5.20** The oscillatory behaviour of time evolution of spin polarization for 30 nm QW sample (ADJ1129, Table 3.5), at 77 k and 3 \( \mu \)m wavelength.

Since in quantum wells there are two materials with different band gap, considerable care must be taken in g-factor and spin lifetime measurement at different photon energies, because we might record signal from different regions of the sample (barrier and well) for different wavelengths. This is particularly a problem when, as in our case, the laser spectrum is very broad (Figure 5.13). Because the band-edge g-factor of the various materials in the structure may be calculated, we can identify from the g-factor measurement which part of the structure we are probing. It is therefore interesting to investigate g-values in the quantum well samples used in the previous chapter for spin lifetimes.
The temperature dependence of the g-factor in different materials (InSb wells, AlInSb barriers), along with bulk InSb, is presented in Figure 5.21. One can see that the g-value does not depend much on well width, and this is likely to be because $g^*$ is more sensitive to the photon energy than to the bandgap. For g-factor measurement of the barrier, the wavelength was around 3μm, however for the QW the wavelength was ~5 μm. The effect of 2-Dimensional behaviour on $g^*$ is complex theoretically, and we did not investigate it here. However, we argue that the g-value of the bulk barrier material can be calculated and the g-value of the bulk material from which the well is made may also be calculated. The g-value of the 2D electrons in the well must be intermediate between these two.

The g-factor measured at 3μm corresponds just to that expected for the barrier material, and that measured at 5μm is intermediate between the barrier and InSb, and must therefore be associated with the well. Measurements performed at 4μm (not shown); although apparently below the band-gap energy of the barrier (3.7μm), give a g-value that corresponds to the barrier due to the broad spectrum of the laser. Spin lifetime values determined at this wavelength would therefore have a significant effect of the barrier and should not be associated with the well.

Figure 5.21 Comparison of temperature dependence of absolute g-factor for bulk, well, and barrier. As it is clear 30- and 40- nm QWs show similar results, and the largest and smallest g-factors belong to the bulk and barrier, respectively. The g-factors do not show much difference with temperature, as we have already observed that periods of the Larmor oscillations do not depend on the temperature very much.
5.3.4 Conclusion

Coherent Larmor precession in strongly spin-orbit coupled semiconductors such as bulk InAs and InSb has been studied up to 293 K. In presence of very small magnetic field even at room-temperature, the inferred Zeeman spin-splitting is strongly non-parabolic. The large spin-orbit coupling mixed across a small gap produces a theoretical g-value that is large and negative with a strong energy dependence that agrees well with experiment.

In study of temperature dependency of the energy gap, it has been shown that the dilational part reduces the difference between k.p theory and experiment.

The possibility of temperature dependence of interband matrix element has been also considered, but much bigger changes are required than theoretically expected, unless the effects of thermal phonons at high temperature are also included.

These assumptions might also solve the apparent anomaly [Oestreich (1995), Oestreich (1996), Lai (2006), Hohage (2006)] in the temperature dependence of g-factor between experiment and k.p theory previously reported for GaAs. For GaAs the non-parabolicity is usually taken to be small because the dependence of the effective mass on electron energy is small. However, the g-value, although small in magnitude, is significantly non-parabolic and changes from -0.44 at the band edge for low temperature to zero at an excess energy of 80 meV. A thermal distribution of electrons at 300 K can therefore have an average g-value that is measurably different from the band-edge value, so that while the band-edge g-factor increases in magnitude with temperature, the thermal average g-value may decrease. Moreover, the authors have not considered the dilational change in the band gap with temperature, which certainly will reduce the difference between theory and experiment.

Furthermore, the g-factor has been calculated for well and barrier in AlInSb QW samples. Comparison with bulk shows that QWs have smaller absolute g-factor than bulk, because the g-factor depends on the band gap energy $E_g$ (see Eqn 2-32), and for QWs the energy levels are higher than in the bulk due to the confinement energy.

The coherent spin precession observed at room temperature in modest magnetic field shows that InSb holds promise for future spin physics and applications.
6 Experiment on donor dopant relaxation

6.1 Introduction and motivation
Previously in this thesis we described spin polarisation lifetime measurements, and spin polarisation precession measurements. All of the mid-infrared time resolved techniques used were aimed at spintronic and quantum information physics and applications. In this chapter we describe a far-infrared population lifetime measurement, also with quantum information goals.

We report on a remarkable feature of dynamics in the excited Rydberg states of hydrogenic Group V donor atoms in silicon. We have used a three beam balanced pump-probe measurement technique [Hughes (1995), Findlay (1998)], at the Dutch free electron laser, FELIX to measure directly the population lifetimes ($T_1$) of Rydberg states of the impurities P and As in Si. Comparing these lifetimes and the corresponding linewidths we find that, at low temperature the only decoherence mechanism for transitions between Rydberg levels is lifetime broadening due to the phonon induced decay of these levels. The Si host does not introduce any other sources of decoherence, and thus in this respect Si is like the vacuum. Thus, our experiments reveal that impurities in Si are like atoms in traps, and we expect that all the coherent manipulation of atomic states in traps can be reproduced in impurities in Si. In particular, the suggestions that silicon doped with group V atoms can provide a route to a practical quantum computer [Kane (1998)], and that group V donor atoms in silicon are a promising far infrared (THz) laser materials [Pavlov (2000), Borak (2005), Pavlov (2006), Hubers (2005)] are supported by these findings.

6.2 Free Electron Laser for Infrared eXperiment (FELIX)
Free electron laser (FEL) generates tuneable, coherent and high power radiation in a wavelength range from millimetre to x-ray. It uses a relativistic electron beam as the lasing medium. Main components of a FEL are an accelerator and a very long undulator. The accelerated electrons pass through a periodic transverse magnetic field, produced by arranging magnets with alternating poles along the beam path (undulator), and produce synchrotron radiation (Figure 6.1).
With a FEL continuous wavelength scans over a wide spectral range as well as high peak power experiments are possible. It provides the opportunity to control characteristics such as wavelength, bandwidth, pulse energy, and duration. The FELIX facility provides two lines 2.5-35 μm or 25-250 μm in wavelength. There are two accelerators with two ranges of electron energies of up to 25 MeV and 45 MeV. Radiation wavelength can be changed by varying the magnetic field of the undulator or by varying the electron energy.

As the name suggests, 'free'-electrons play a key role in an FEL, in contrast to conventional lasers where the electrons are strongly bound to atoms or molecules that are either in the gas-, liquid- or condensed phase. Only in this sense the electrons in an FEL are free though, for another key ingredient is a periodically varying magnetic field structure, the undulator, which induces a periodic deflection of the trajectory of the beam of electrons.

A relativistic beam of electrons, produced by an accelerator, is injected into a resonator, consisting of two high-reflectivity mirrors, around the undulator. The magnetic field of the undulator is perpendicular to the direction of the electron beam and periodically changes polarity a (large) number of times along its length. This will cause a periodic deflection of the electrons while traversing the undulator. The transverse motion is quite analogous to the oscillatory motion of electrons in a stationary dipole antenna and hence will result in the emission of radiation with a frequency equal to the oscillation frequency. The overall motion of the electrons in
the undulator resembles the motion of oscillating electrons in a dipole antenna moving close to the speed of light. This high velocity results in a strong Doppler shift: the frequency of the radiation emitted in the forward direction is typically up-shifted by a factor $\gamma^2$, where $\gamma$, the Lorentz factor, is a measure of the electron energy. [For FELIX this upshift is in the range of 300 to 15,000. Given the 65 mm-period of the undulator, this brings the radiation wavelength in the IR range.] This radiation, referred to as spontaneous emission, is usually very weak though. This is a consequence of the fact that the electrons are typically spread out over an interval that is much larger than the radiation wavelength and will therefore not emit coherently. But on successive roundtrips in the resonator, this weak radiation will be amplified (gain mechanism) by fresh electrons, until saturation sets in at a power level which is typically 10 million to 100 million times that of the spontaneous emission. Except for some dynamical effects such as slippage, the time structure of the radiation will mimic that of the electron beam.

The laser beam of FELIX consists of very short micro pulses, each of an adjustable width between 0.25 ps and 10 ps. The separation between those micropulses is either 1 or 40 ns. The micropulses form together a train that creates the macropulse. The duration of a macropulse ranges up to 10 $\mu$s (Figure 6.2). Repetition rate of the macropulses is either 5 Hz or 10 Hz.

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**Figure 6.2** Temporal structure of the laser beam
6.3 Experimental setup (Far-infrared pump-probe)

The schematic pump-probe setup has been shown in Figure 6.3. From the pump pulse (FELIX) a probe beam and a reference beam are split by two beam-splitters.

Figure 6.3 a) Schematic of pump-probe setup; b) Experimental pump-probe setup, the right photo shows the inside of the left one; the optical setup is under nitrogen gas to protect optical components from water and other atmospheric absorption.
The reference beam goes an extra 6 meters distance, so it has 20 ns delay respect to the probe beam. Then, reference and probe beams follow the same optical path, transmit through the sample at the same position and are detected by a Ga:Ge detector. The three beams (pump, probe, reference) are focused on the sample using a parabolic mirror.

Effective pump and probe energy fluences per micropulses are estimated to be 350 and 10 $\mu J/cm^2$. The relative transmittance of the probe was measured as a function of optical delay between pump and probe pulses. When pump is incident onto the sample, electrons are excited to the upper states from the ground state. When the probe beam arrives, electrons still have occupied higher states; therefore the probe is transmitted, while the electrons decay back to the ground state, the probe transmission is reduced. Then after a long time i.e. 20 ns after probe arrival, when this process has been finished, the reference beam arrives. The difference between the probe and reference beam intensities is measured by the detector.

### 6.4 Samples
The two samples investigated were float-zone grown Si wafers of thickness 400-μm and doped with P or As to donor concentrations of $N_0 \sim 2 \times 10^{21}$ m$^{-3}$. The silicon was 'natural', i.e. it has not been isotopically purified (and is therefore expected to have inhomogeneously broadened spectral lines). We focus first on Si:P but note that Si:As is potentially more useful because the energy gap between Rydberg states is larger (see Figure 6.4).
Figure 6.4 Optical transitions from the lowest energy states of the hydrogen like donor impurities in silicon.

6.5 Results

Figure 6.5 shows the measured probe transmission change as a function of time delay with respect to the pump pulse for the 2p₀ line (the 1s(A₁) → 2p₀ transition) at 34.1 meV in P-doped Si. The rise of the leading edge indicates the pulse duration, which was ~10 ps in this case. For all pump powers the decay can be adequately fitted by a straight line on a log-linear scale, and hence described by a single exponential lifetime. Using frequencies away from a Rydberg transition all show very weak signal that simply follows the pulse shape.

The 1s(A₁)-2p₀ absorption cross-section was taken from the measured small-signal transmission spectrum and found to be 0.6x10⁻²⁰ m². The peak focused photon flux was in the range 1.1x10²⁸ to 1.9x10³¹ m⁻²s⁻¹ and was therefore enough to fully bleach transitions with lifetime longer than ~10ps (i.e. any lifetime longer than the pulse duration, which was also ~10ps).
Figure 6.5. The transmission change of the probe pulse induced by the pump, as a function of the time delay between them, observed in the Si:P sample for the 1s(A1) → 2p₀ transition at sample temperature of 10 K and a laser photon energy of 34.1 meV. Laser pump powers used corresponded to micropulse energies ranging from 0.86 to 0.001 µJ (some of which are shown in the figure). The transient signals all show a single exponential decay with lifetime $208 \pm 10$ ps.

The measured lifetime of the 2p₀ state in Si:P is 208 ps which corresponds to a lifetime broadened linewidth of 0.025 cm⁻¹. The actual linewidth may be larger (but not smaller) than this value, due to dephasing processes. The lowest measured value for the linewidth reported for this transition, by Karaiskaj et al. [Karaiskaj (2003)] is 0.034 cm⁻¹, which was obtained in an isotopically pure $^{28}$Si sample, is only slightly larger. We therefore believe that in that case, the sample was very close to being free of dephasing processes, and in contrast to the behaviour of defects in many other hosts, the dominant broadening mechanism for donors in Si is lifetime broadening. Karaiskaj et al. [Karaiskaj (2003)] suggested that even narrower linewidths will be found in chemically, as well as isotopically purified Si. Donors in Si should then behave like stationary atoms in traps, and, at low temperature show little or no inhomogeneous broadening.
Figure 6.6 shows the absorption spectrum and lifetimes for our P and As-doped Si sample, as measured by FTIR with a resolution of 0.25 cm\(^{-1}\) (0.03 meV) at 10 and 50 K.

Figure 6.6 a) The absorption spectrum for P-doped Si measured by FTIR spectroscopy with 0.03 meV resolution. Above 30 meV the sample temperature was 10 K (filled symbols); below 30 meV it was 50 K (open symbols). The lifetimes of the indicated states, determined from pump-probe signals (such as Figure 6.5) are also shown; b) as a) for Si:As (with the exception that the low energy experiments were at 60 K); c) The one-phonon density of states of Si taken from Ref. [Flensburg (1999)].
6.6 Discussion

The well-known Lyman transitions $1s(A_1) \rightarrow np_0, np_\pm$ between 34 and 45 meV are clearly seen in the absorption spectra of Figure 6.6. In the lower energy range from 18 to 34 meV absorption lines due to $1s(E), 1s(T_2) \rightarrow np_0, np_\pm$ transitions can be seen, but only at higher temperatures, where the initial $1s(E)$ and $1s(T_2)$ states are thermally populated. It is apparent that our measured lifetimes are generally higher for Si:P than for Si:As, and that the shortest value being obtained for the $1s(A_1) \rightarrow 2p_0$ transition of Si:As which nearly coincides with a peak in the LA density of states.

Below 42.5 meV, a probe signal was only detected when the free-electron laser was tuned to impurity transition energy. For the transitions from $1s(A_1)$ to levels above $3p_\pm$, the spectral width of FELIX overlaps with many closely spaced energy transitions and the continuum and therefore the decay times for this state and above may be a mixture of different transitions complicating the results. The energy of the $3p_0, 3p_\pm$ states are close to the maximum of the longitudinal acoustic (LA) phonon density situated at an energy of 42.7 meV. We expect an increased relaxation rate for these states as observed for the $3p_\pm \rightarrow 1s(A_1)$ transition. The lifetime of the $2p_\pm \rightarrow 1s(A_1)$ transition is intermediate between those of the $2p_0$ and $3p_\pm \rightarrow 1s(A_1)$ transitions. This is because the $3p_0, 3p_\pm$ are very close to $2p_\pm ([E_{2p_\pm} - E_{3p_0}]/k_B \sim 10$ K), so at the temperatures of the measurements, the $2p_\pm$ electrons can cascade upwards to these states by absorption of equilibrium phonons, the reverse of the cascade phonon-assisted relaxation process for highly excited electrons.

In Si:As, the energy of the ground state $1s(A_1)$ is shifted to lower energies by $\sim 10$ meV. The lifetimes of $2p \rightarrow 1s(A_1)$ transitions in Si:As are therefore different because of the different overlap with the one-phonon density of states. Whereas in Si:P the $3p$ transitions are the fastest because of resonance with the LA phonons, in Si:As, it is the $2p_0$ which is most closely resonant. The electrons in the $2p_\pm$ state follow a cascade relaxation process through $2p_0$, and have a slightly longer lifetime.

Further insight into the relaxation dynamics of the donor center can be gained by studying the temperature dependence of the measured lifetimes (Figure 6.7). For the $2p_0 \rightarrow 1s(A_1)$ transition we can see the lifetime is increasing up to 50 K and decreases.
at higher temperatures. At a temperature of ~45 K, when the $1s(E)$ is thermally populated, we were able to measure the lifetime of $2p_0 \rightarrow 1s(E)$ transition.

![Figure 6.7](image.png)

**Figure 6.7** The temperature dependence of the lifetimes for $2p_0$, $2p_\pm \rightarrow 1s(A_{1})$ transitions at 34.1 and 39.2 meV, respectively in P-doped Si. The solid and dashed lines are fits of Eqn 6.1 to the $2p_0$ and $2p_\pm \rightarrow 1s(A_{1})$ transitions, respectively. The inset shows the scheme of optical transitions in Si:P material (as described by Eqn 6.1).

It can be seen from Figure 6.7 that the measured lifetime initially increases, which can be only explained by thermally shifting the electrons into states where the relaxation is slower. This would be expected if the excited electrons decay to the ground state via the intermediate states of the continuum. The recombination part of this pathway has a long lifetime at low concentrations of free-electrons, because the electrons spend a long time spatially separated from the ion. Only when the donors become almost completely ionised at higher temperatures does the observed absorption recovery time drop. We can describe this temperature dependence with a phenomenological equation for the effective relaxation time:

$$\frac{1}{\tau_{\text{eff}}(T)} = \frac{1}{T_1} - R_{a} e^{-\Delta E_a/kT} + R_{b} e^{-\Delta E_b/kT}$$

Eqn 6.1
The first term on the right describes the direct population relaxation from \(2p_x \rightarrow 1s(A_1)\). The second term, for the case of ionized acceptors at finite temperature [Cuthbert (1967)], comes about because raising the temperature increases the number of equilibrium free electrons created via activation from the \(2p\) states to the bottom of the conduction band. These free electrons then spend an increasing fraction of their time away from the donor centers and the upshot is an increased effective lifetime as measured in our absorption experiment that senses a recovery of the \(1s \rightarrow 2p\) signal only when the \(1s\) state is reoccupied, which is of course less likely when the original electrons are far from the donors. At higher temperature, the Boltzmann tail of the free electron distribution can have enough energy to enable thermal excitation from the \(2p\) state sufficiently far into the conduction band for subsequent recombination via emission of the strong transverse optical phonons near 60 meV (see Figure 6.6c). This gives rise to the third, thermally activated term. The inset of Figure 6.7 illustrates these effects.

The adjustable parameters of Eqn 6.1 may therefore be interpreted as follows: \(\Delta E_a\) is the ionization energy for \(2p\) electrons, \(\Delta E_b = E_{TO} - E_{21}\) is the activation energy for optical phonon emission where \(E_{21}\) is the energy of \(2p_0 \rightarrow 1s(A_1)\) transitions, \(1/R_b = 1/R_{TO}\) is the optical phonon emission lifetime. The solid line in Figure 6.7 corresponds to the following values of the fitting parameters: \(T_1 = 215 \pm 10\) ps, \(\Delta E_a = 11.8 \pm 1.1\) meV, \(\Delta E_b = 32.1 \pm 2.1\) meV and \(1/R_b = 1.7\) ps. The energy values for the excited state and the ground state involved in the relaxation process for the \(2p_0 \rightarrow 1s(A_1)\) transition are in good agreement with \(\Delta E_a = 11.5\) meV, \(E_{TO}-E_{21} \sim 30\) meV. The temperature dependence of the lifetime of the \(2p_x\) is similar to that of the \(2p_0\) (Figure 6.7) because the transition energy is similar, though the ionization energy is smaller by a factor of two giving rise to the steeper initial increase.

### 6.7 Conclusion

Our conclusion is that recombination is slower than the direct process of \(1s-2p\) population relaxation (by investigation of the temperature dependence) which confirms that it is not important in the low temperature measurements.
We demonstrate that transitions between hydrogen-like shallow donor impurity states in silicon show only lifetime broadening. Our time-resolved studies of the population dynamics of these states, obtained using a free-electron laser, yield lifetimes for the $2p_0$, $2p_\pm$ states ranging from $\sim100$ to 240 ps in the temperature range of 10 K to 50 K. At low temperature, lifetime broadening is the dominant decoherence mechanism, showing that coherent THz manipulation on a $\sim100$ ps timescale is possible.

We have shown that, at low temperatures, the dominant linewidth broadening for excited states of carefully prepared impurities in Si is due to direct phonon decay to lower levels, rather than dephasing collisions or inhomogeneous broadening. The lifetimes $\sim200$ ps are long enough that we can expect coherent manipulation of these impurities to be practical, as required by some quantum information processing schemes.
7 Conclusion & future work

7.1 Conclusion
Several materials in the form of films, quantum wells (QW), and quantum dots (QD) have been used in the pump-probe experiment at mid- and far- infrared regions at different temperatures to study spin dynamics and donor transitions in the samples used.

Time-resolved pump-probe measurements using an ultrafast Ti:sapphire laser have been employed to study spin lifetime and spin precession at mid-infrared wavelengths for different narrow gap semiconductor (NGS) samples including thick InAs film, thick and thin InSb films, different n-InSb-AlInSb QWs, and PbSe and HgTe QDs.

The experimental results show that with increasing temperature, spin lifetime and absolute g-factor decrease. The experimental temperature dependent g-factor results have been compared with k.p theory. The experimental and theoretical temperature dependent g-factor becomes divergent at high temperature. It has been shown that when the temperature dependency of the dilational energy gap is considered rather than the optical gap, this deviation is enormously modified.

The temperature dependency of other parameters in the k.p model such as momentum matrix elements is also considered as a result of the lattice expansion and thermal phonon effects at high temperature.

In non-parabolic conduction bands, the g-factor is energy dependent and its magnitude at the band edge is different from energies far from the band edge. The experimental results of energy dependent g-factor have been confirmed by the three-band model and $8 \times 8$ k.p theory.

By including these effects, which are important at particular circumstances such as working at high temperature and far from the band-edge, we believe that the k.p theory can provide a powerful theoretical tool for comparison with experimental results.
The spin dynamic results have shown that in the range of temperature of 77 to 293 K, the dominant spin relaxation mechanism is, in most cases, D’yakonov-Perel’ (DP). Unlike Elliot-Yafet (EY) mechanism, when DP mechanism dominates, applying an external electric or magnetic field is a useful tool for spin manipulation.

Although, InAs bulk shows longer spin lifetime, comparison between results of spin lifetime for InSb thin films, with previous results for InAs thin films, shows that existence of an accumulation layer in InAs surface with very short spin lifetime and high current is a disadvantage of this material. While InSb has a depletion surface layer with longer spin lifetime and lower current, so we infer that InSb can be a better candidate for spintronics applications (such as spin-FET [Datta (1990)]).

Spin dynamic study of different QW samples shows that the spin lifetime depends on different samples’ characters such as well-width, mobility, and carrier concentration (200 fs for highly doped narrow QWs to 4 ps for undoped wide QWs). Also, the g-factor of a QW is -33, which is less than that of bulk (-50). The DP relaxation mechanism dominates for high mobility for the whole temperature range. For low mobility at low temperature the EY is dominant.

Preliminary study on spin lifetime and g-factor measurement on QD samples has been done. Only some results of the former have been presented here. The quality of the QD samples were used (PbSe and HgTe) was not quite satisfactory. We believe that more experimental and theoretical investigation should be done on QD samples with good quality, to achieve a more precise conclusion.

Finally, a pump-probe experiment using a free electron laser has been used at far-infrared wavelengths to study the donor transitions in Si samples. The transitions between hydrogen-like shallow donor impurity states (P, As) in silicon show only lifetime broadening. Time-resolved study of population dynamics of these states yields lifetimes from 100 to 240 ps in the temperature range of 10 K to 50 K. The coherence manipulation of these impurities required for quantum information processing is applicable due to the long lifetime.
7.2 Future work

Since spin dynamic study plays an important role in spintronics and the applications are in fast progress, it is important to do more experimental and theoretical investigation about spin lifetime and spin precession measurement for different samples including narrow-, medium-, and wide-gap materials in various structure such as bulk and nanostructures, with different characters (such as dimensions, mobility, and carrier concentration). These studies are quite useful to study the effect of different characters separately, also to recognize the dominant relaxation mechanism, and finally to identify the best materials with specific parameter for applications.

The range of temperatures can be extended to have a better understanding of the predominant phenomena at different temperatures. Particularly, most of the previous experiments have been performed at low temperature and they can provide a helpful tool for comparison with the new results.

During our experiment for g-factor measurement a constant external magnetic field has been used. It would be valuable to consider the effect of different magnitudes and directions of magnetic fields. Moreover, applying an external electric field would also be useful for more study about spin dynamics and its manipulation.

Because QDs have been considered extensively for different applications, it would be quite useful if more experiments are performed on different materials of good quality, with different parameters such as various sizes.

The plan for silicon donors would be to use the same setup of g-factor measurement using free electron laser in Holland, to investigate the possibility of coherent control of spin-polarised transitions between hydrogen like shallow donor impurity states.
A Appendix: Development of k.p theory

For finding the full Hamiltonian, there have been two different approaches. In the first approach interband matrix elements of the k.p perturbation Hamiltonian, and also spin-orbit Hamiltonian are found. In the Kane model [Kane (1957)] the k.p interactions within the valence conduction band complex are considered in an exact manner, while the interactions with remote bands are accounted for in second order perturbation theory. A second method was that of Luttinger [Luttinger (1955a)]. He used all allowed of matrix elements of k and k×k among the valence band states (Γ₈). His parameters were combination of those of Kane, but included an additional parameter q, which is nonzero only in the presence of spin-orbit splittings of the intermediate states. Luttinger-Kohn (LK) Hamiltonian [Luttinger (1955b)] considers degenerate bands and spin-orbit splitting. In the LK model, only the interactions within the valence band are treated exactly, while second order perturbation theory is used for the interactions with all other bands, including those with the conduction band. Luttinger’s results were extended by Roth et al. [Roth (1959)] to include Γ₇ split-off band. A similar approach taking into account the presence of a magnetic field, known as the Pidgeon and Brown (PB) model [Pidgeon (1966)] included the Γ₆ conduction band, combining the results of Kane [Kane (1957)] and of Roth et al [Roth (1959)]. They neglected the effects of inversion symmetry in their model. Weiler et al [Weiler (1978)] have used the second approach to obtain a complete set of parameters for the coupled Γ₆, Γ₇, and Γ₈ bands, and found three new parameters in addition to those of Kane and Roth et al. with the same origin as Luttinger’s parameter q. An alternative approach employs the band-decoupling scheme of Luttinger and Kohn [Luttinger (1955b)] via three-level model with higher orders of the k.p perturbation.

Another development can be achieved by going from the three-level model to the five-level model (14 x 14 Hamiltonian), including the higher conduction band [Weisbuch (1977), Pfeffer (1990), Pfeffer (1996)].
B Appendix: Time independent perturbation theory

Generally, k.p is a method which uses perturbation theory to relate the information of energy levels at k to the effective mass. Therefore, for theoretical consideration we need to mention some basic concepts of Quantum mechanics about Schrodinger equation, first. We start with Hamiltonian in time-independent perturbation theory:

\[ H = H^{(0)} + H' \]  \hspace{1cm} \text{Eqn B1}

where \( H' \) is a small perturbation, and \( H^{(0)} \) is unperturbed Hamiltonian with known solutions:

\[ H^{(0)} \phi^{(0)}_n = E^{(0)} \phi^{(0)}_n \]  \hspace{1cm} \text{Eqn B2}

with \( n = 1, 2, 3, \ldots \), \( E^{(0)} \) is eigen-energy, and \( \phi^{(0)}_n \) is eigen-function and forms an orthonormal basis. When the basis states are well chosen, then we can find the eigen-functions and eigen-energies of \( H \) to the first and second order using

\[ \psi_n = \phi^{(0)}_n + \sum_{m \neq n} \frac{H'_{mn}}{E^{(0)}_n - E^{(0)}_m} \phi^{(0)}_m, \]  \hspace{1cm} \text{Eqn B3}

\[ E_n = E^{(0)}_n + H'_{nn} \]  \hspace{1cm} \text{Eqn B4}

and

\[ \psi_n = \phi^{(0)}_n + \sum_{m \neq n} \frac{H'_{mn}}{E^{(0)}_n - E^{(0)}_m} \phi^{(0)}_m + \sum_{m \neq n} \left[ \sum_{k \neq \mu} \left\{ \frac{H'_{mk} H'_{k\mu}}{(E^{(0)}_k - E^{(0)}_m)(E^{(0)}_k - E^{(0)}_n)} - \frac{H'_{mn} H'_{mn}}{(E^{(0)}_n - E^{(0)}_m)^2} \right\} \phi^{(0)}_\mu - \frac{|H'_{mn}|^2}{2(E^{(0)}_n - E^{(0)}_m)^2} \phi^{(0)}_n \right]. \]  \hspace{1cm} \text{Eqn B5}

\[ E_n = E^{(0)}_n + H'_{nn} + \sum_{m \neq n} \frac{|H'_{mn}|^2}{E^{(0)}_n - E^{(0)}_m}. \]  \hspace{1cm} \text{Eqn B6}

with \( H'_{nm} = \int \phi^{(0)*}_m H' \phi^{(0)}_n d^3r \).
**H$_{SO}$- Hamiltonian with spin-orbit interaction**

The initial one-electron eigen-value problem with spin-orbit interaction (no external magnetic field) infers:

$$\left[ \frac{p^2}{2m_0} + V(r) + \frac{\hbar}{4m_0^2c^2} (\sigma \times \nabla V) \cdot p \right] \Psi = E \Psi$$  \hspace{1cm} \text{Eqn B7}

The solution of Eqn B7 in the form of Luttinger and Kohn (LK) [Luttinger (1955b)] is:

$$\Psi^m_k(r) = \exp(i k \cdot r) \sum_i c^m_i(k) u_i(r).$$  \hspace{1cm} \text{Eqn B8}

The summation is over all bands and the index $m$ shows the band of interest. The LK periodic amplitudes satisfy Eqn B7 at a band's extremum (at $k=0$),

$$\left[ \frac{p^2}{2m_0} + V(r) + \frac{\hbar}{4m_0^2c^2} (\sigma \times \nabla V) \cdot p \right] u_i = E^0_i u_i$$  \hspace{1cm} \text{Eqn B9}

where $E^0_i$ is the edge energy of the $i$th band. The LK functions are orthonormal: $\langle u_r | u_i \rangle = \delta_{i,r}$, where the integration is over the unit cell. Placing Eqn B8 in Eqn B7 multiplying on the left by $u_r^*$, and integrating over the unit cell, gives:

$$\sum_i \left[ \left( E^0_i + \frac{\hbar^2 k^2}{2m_0} - E \right) \delta_{i,r} + \frac{\hbar}{m_0} k \cdot p_{i,r} + H^0_{i,r} \right] c^m_i = 0.$$  \hspace{1cm} \text{Eqn B10}

The index $i'=1, 2, 3, \ldots$ runs over the bands, and $p_{i,r}$ are interband matrix elements,

$$p_{i,r} = \langle u_r | p + \frac{\hbar}{4m_0c^2} (\sigma \times \nabla) | u_i \rangle.$$  \hspace{1cm} \text{Eqn B11}

The second term which appears to lead to linear $k$ terms in the energy $E(k)$ disappears for $l = l'$ [Bir (1974)].
**H_B- Hamiltonian with external magnetic field, no spin-orbit interaction**

The interaction potential energy of spin in presence of an external magnetic field is \(-\mathbf{\mu}_s \cdot \mathbf{B}\), where \(\mathbf{\mu}_s = -es/m_0\), therefore, in quantum mechanics the Hamiltonian is \(H_B = e\mathbf{S} \cdot \mathbf{B}/m_0\) or \(H_B = \mu_S \mathbf{B}\), where \(\mathbf{S} = \hbar \sigma/2\) and \(\mu_S = e\hbar/2m_0\).

Classically, by applying a magnetic field, electrons move under Lorentz force, \(\mathbf{F} = -ev \times \mathbf{B}\), and related to vector potential \(\mathbf{B} = \mathbf{V} \times \mathbf{A}\). Because \(\mathbf{V} \times \mathbf{E} = -\partial \mathbf{B}/\partial t = -\partial (\mathbf{V} \times \mathbf{A})/\partial t\), we have \(\mathbf{E} = -\partial \mathbf{A}/\partial t\), so for motion equation \(\partial \mathbf{p}/\partial t = -e\mathbf{E}\), we obtain \(\hbar \partial \mathbf{k}/\partial t = e\partial \mathbf{A}/\partial t\) or \(\hbar \mathbf{k}(\mathbf{B}) = \hbar \mathbf{k}(0) + e\mathbf{A}\). It means that in presence of an external magnetic field we can replace the momentum operator by \(\mathbf{p} + e\mathbf{A}\). Therefore, Hamiltonian equation is:

\[
H\Psi = \{[\mathbf{p} + e\mathbf{A}(\mathbf{r})] / 2m_0 + \mathbf{V}(\mathbf{r})\}\Psi
\]

Eqn B12

where \(\mathbf{p} = \hbar \mathbf{V}/i\) is the momentum operator, \(m_0\) is the free electron mass, and \(\mathbf{V}(\mathbf{r})\) is the periodic potential energy of the electron in the crystal. With choosing a \(\mathbf{B}\) in the \(z\)-direction, and \(\mathbf{A} = Bx\hat{y}\), the Hamiltonian becomes:

\[
\left[ \frac{\hbar^2}{2m_0} \nabla^2 - \frac{ie\hbar Bx}{m_0} \frac{\partial}{\partial y} + \frac{(eBx)^2}{2m_0} \right] \Psi = E\Psi
\]

Eqn B13

Then, we can write the wavefunction as:

\[
\Psi(x, y) = u(x) \exp(ik_y y)
\]

Eqn B14

Substitution of the wavefunction in the Eqn B13 gives:

\[
\left[ \frac{\hbar^2}{2m_0} \frac{\partial^2}{\partial x^2} + \frac{1}{2} m_0 \omega_c^2 \left[ x + \frac{\hbar k_y}{eB} \right]^2 \right] u(x) = Eu(x)
\]

Eqn B15

which is the Schrodinger equation for a one-dimensional harmonic oscillator with the magnetic field dependent cyclotron frequency, \(\omega_c = eB/m_0\), and the centre of the parabolic
potential at \( x_k = -\hbar k / eB \). Solution of Eqn B15 yields a quantized set of energy levels known as Landau levels:

\[
E_n = \left( n + \frac{1}{2} \right) \hbar \omega_c
\]

Eqn B16

The wave-functions are the usual Hermit polynomials:

\[
\Psi(x, y) = \sqrt{\frac{1}{2^n n! \sqrt{l_B}}} \exp \left[ -\frac{(x - x_k)^2}{2l_B^2} \right] \frac{H_n}{l_B} \frac{x - x_k}{l_B} \delta_{k_y} \]

Eqn B17

with the magnetic length, or Larmor radius, \( l_B = \sqrt{\hbar / eB} \).

**H\text{Total}** Hamiltonian with spin-orbit interaction and external magnetic field

The initial one-electron eigenvalue problem with spin-orbit interaction in the presence of an external magnetic field gives:

\[
\left[ \frac{p^2}{2m_0} + V(r) + \frac{\hbar}{4m_0 c^2} (\sigma \times \nabla V) \cdot p + \mu_B \mathbf{B} \cdot \sigma \right] \Psi = E \Psi
\]

Eqn B18

with \( p \rightarrow p + eA(r) \). Due to the presence of a magnetic field the eigen-value problem Eqn B18 is not periodic. The solutions are considered in the form of:

\[
\Psi = \sum_l h_l(r) u_l(r),
\]

Eqn B19

\( u_l(r) \) are the LK periodic functions satisfying Eqn B9, and \( h_l(r) \) are the slowly varying envelope functions. Inserting Eqn B19 into Eqn B18, using Eqn B9, multiplying on the left by \( u_l^* \), and integrating over the unit cell, gives:

\[
\left[ \frac{p^2}{2m_0} + E_{l0} - E \right] \delta_{ll} + \frac{1}{m_0} p_{l\sigma} \cdot p + \mu_B B \cdot \sigma l + H_{l0}^{30} \right] h_l = 0.
\]

Eqn B20
where \( \sigma_{mn} = \langle \sigma | \sigma | u_i \rangle \). Eqn B20 represents set of coupled differential equations for the envelope functions \( h_i(r) \). Taking the set of basis of LK functions, the resulting eigen-value problem is written in matrix form of Eqn B20, it has been done for five band model of middle gap semiconductors [Pfeffer (1990), Pfeffer (1996)].

Each envelope function of the set Eqn B20 is developed into a series:

\[
h_i(r) = \exp(ik_z z) \sum f_{nm} \phi_m, \tag{Eqn B21}
\]

where \( \phi \) are the harmonic oscillator functions and \( f_{nm} \) are numerical coefficients. For \( k_z = 0 \), and for three band model the set Eqn B20 factorizes into two \( 4 \times 4 \) sets (for two spin directions). For band electron, free electron mass, \( m_0 \) is replaced by effective mass, \( m^* \). In the presence of spin-orbit splitting and external magnetic field the electron energy is given by:

\[
E_{n,k}^*(B) = E_n + \frac{\hbar^2 k^2}{2m^*} + (n + \frac{1}{2}) \frac{\hbar eB}{m^*} \pm \frac{1}{2} g^* \mu_B B \tag{Eqn B22}
\]

where \( k = k_z \) is the component of momentum parallel to the magnetic field \( B \), and the quantum numbers \( \pm \) refer to spin.
C Appendix 8x8 k.p matrices

Table C1 The k.p Weiler Hamiltonian for a zinc-blende semiconductor; the upper triangle is complex conjugate of the lower one [Weiler (1981)]. For simplicity, the terms in matrix Hamiltonian are given in atomic units $\hbar = m = 1$. The $8 \times 8$ matrix is Hermitian, and split into $4 \times 4$ blocks i.e.

$$\hat{H} = \begin{bmatrix} \hat{H}_{aa} & \hat{H}_{ab} \\ \hat{H}_{ba} & \hat{H}_{bb} \end{bmatrix},$$

where

$$\hat{H}_{aa} =$$

<table>
<thead>
<tr>
<th>$E_g$</th>
<th>$+(F + \frac{1}{2})k^2$</th>
<th>$+(N_1 + \frac{1}{2})H_z$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{1}{\sqrt{2}}P_{k}^{-}$</td>
<td>$-\frac{1}{2} \gamma_1 k^2$</td>
<td>$+ \frac{1}{2} \gamma_1 F_3^1$</td>
</tr>
<tr>
<td>$+ \frac{1}{\sqrt{2}}GF_4^+$</td>
<td>$- \frac{1}{2} \gamma_1 k^2$</td>
<td>$+ \frac{1}{2} \gamma_1 F_3^1$</td>
</tr>
<tr>
<td>$+ \frac{1}{2} N_{3}H^+$</td>
<td>$- \frac{1}{2} \gamma_1 k^2$</td>
<td>$+ \frac{1}{2} \gamma_1 F_3^1$</td>
</tr>
</tbody>
</table>

| $\frac{1}{\sqrt{6}}P_{k}^{z}$ | $\frac{1}{2} \gamma_2 F_3^2$ | $- \frac{1}{2} \gamma_1 k^2$ |
| $+ \frac{1}{\sqrt{6}}GF_{z}^+$ | $\frac{1}{2} \gamma_2 F_3^2$ | $- \frac{1}{2} \gamma_1 k^2$ |
| $+ \frac{1}{2} N_{3}H^{-}$ | $\frac{1}{4} (\kappa + \frac{1}{4} q)H_z$ | $\frac{1}{4} (\kappa + \frac{1}{4} q)H_z$ |

| $\frac{1}{\sqrt{3}}P_{k}^{\prime}$ | $- \frac{1}{2} \gamma_1 F_3^2$ | $- \frac{1}{2} \gamma_1 k^2$ |
| $- \frac{1}{\sqrt{3}}GF_{k}^+$ | $- \frac{1}{2} \gamma_1 F_3^2$ | $- \frac{1}{2} \gamma_1 k^2$ |
| $+ \frac{1}{2} C'k_z$ | $- \frac{1}{2} \gamma_1 k^2$ | $+ (\kappa^+ + \frac{1}{2}) H_z$ |

| $\frac{1}{\sqrt{3}}G'F_4$ | $+ \frac{1}{2} C'k_z$ | $+ \frac{1}{2} C'k_z$ |
| $- \frac{1}{\sqrt{3}}G'F_4$ | $+ \frac{1}{2} C'k_z$ | $+ \frac{1}{2} C'k_z$ |

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\[
\hat{H}_{\text{so}} =
\begin{align*}
E_z \\
+ (F + \frac{1}{2})k^2 \\
- (N_1 + \frac{1}{2})H_z \\
\frac{1}{\sqrt{6}} Pk^- \\
+ \frac{1}{\sqrt{6}} GF_4^+ \\
- \frac{\sqrt{3}}{2} N_1 H^+ \\
- \frac{1}{2} \gamma_1 k^2 \\
+ \frac{1}{2} \gamma_2 F_3^1 \\
- Ck_z \\
\frac{1}{\sqrt{3}} P'k^- \\
+ \frac{1}{\sqrt{3}} GF_4^+ \\
- \frac{1}{2} (\kappa'' + 1)H_z \\
\frac{1}{\sqrt{2}} \gamma_1' F_3^2 \\
- \frac{3}{2} \gamma_2' F_4^z \\
\frac{1}{2} C'k_z \\
\frac{1}{\sqrt{2}} \gamma_1' F_3^2 \\
- \frac{3}{2} \gamma_2' F_4^z \\
\frac{1}{2} C'k_z \\
\frac{1}{\sqrt{2}} \gamma_1' F_3^2 \\
- \frac{3}{2} \gamma_2' F_4^z \\
\frac{1}{2} C'k_z \\
\frac{1}{\sqrt{2}} \gamma_1' F_3^2 \\
- \frac{3}{2} \gamma_2' F_4^z \\
\frac{1}{2} C'k_z \\
- \Delta \\
- \frac{1}{2} \gamma_1' k^2 \\
- (\kappa' + \frac{1}{2}) H_z
\end{align*}
\]
\[
\begin{array}{|c|c|c|c|
\hline
\text{Column 1} & \text{Column 2} & \text{Column 3} & \text{Column 4} \\
\hline
\frac{1}{3} \sqrt[3]{P k} & \frac{1}{3} \sqrt[3]{P k} & \frac{1}{3} \sqrt[3]{P k} & \frac{1}{3} \sqrt[3]{P k} \\
\hline
N_{2} F_{3}^{+} + N_{3} H_{2} & N_{2} F_{3}^{+} + N_{3} H_{2} & N_{2} F_{3}^{+} + N_{3} H_{2} & N_{2} F_{3}^{+} + N_{3} H_{2} \\
\hline
N_{2} F_{3}^{+} + N_{3} H_{2} & N_{2} F_{3}^{+} + N_{3} H_{2} & N_{2} F_{3}^{+} + N_{3} H_{2} & N_{2} F_{3}^{+} + N_{3} H_{2} \\
\hline
N_{2} F_{3}^{+} + N_{3} H_{2} & N_{2} F_{3}^{+} + N_{3} H_{2} & N_{2} F_{3}^{+} + N_{3} H_{2} & N_{2} F_{3}^{+} + N_{3} H_{2} \\
\hline
\end{array}
\]
Table C2 The $8 \times 8$ $k.p$ operator Hamiltonian of Table C1, where the $k_x$, $k_y$ dependence is rewritten in terms of the raising and lowering operators for non-zero magnetic field. The magnetic field $B$ enters via the free electron cyclotron energy $s = e B / m_0$ (in atomic units $B = s$).

<table>
<thead>
<tr>
<th>$E^*$</th>
<th>$P \sqrt{s} a^+$</th>
<th>$- \frac{P}{\sqrt{3}} \sqrt{s} a$</th>
<th>$- \frac{2s}{\sqrt{3}} P'a^+$</th>
<th>$- \frac{2}{\sqrt{3}} P'k_z$</th>
<th>$- \frac{2}{\sqrt{3}} s G(a'^2 - a^2)$</th>
<th>$\frac{1}{\sqrt{3}} P'k_z$</th>
<th>$\frac{1}{\sqrt{3}} s G(a'^2 - a^2)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(\frac{F}{2} + \frac{1}{2}(s(2n+1) + k_z^2)) + (N_1 + \frac{1}{2}) s$</td>
<td>$\frac{1}{2} \gamma_1 [s(2n+1) + k_z^2]$</td>
<td>$\frac{2}{\sqrt{3}} \sqrt{s} G k_z a^+$</td>
<td>$\frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{2}{\sqrt{3}} s G k_z a^+$</td>
<td>$0$</td>
<td>$- \frac{2}{\sqrt{3}} s G k_z a^+$</td>
<td>$0$</td>
</tr>
<tr>
<td>$+ (N_1 + \frac{1}{2}) s$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
</tr>
<tr>
<td>$P \sqrt{s} a$</td>
<td>$\sqrt{s} a^+$</td>
<td>$\sqrt{s} a^+$</td>
<td>$\sqrt{s} a^+$</td>
<td>$\sqrt{s} a^+$</td>
<td>$\sqrt{s} a^+$</td>
<td>$\sqrt{s} a^+$</td>
<td>$\sqrt{s} a^+$</td>
</tr>
<tr>
<td>$+ 2 \sqrt{s} G k_z a^+$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$+ \frac{1}{2} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
</tr>
<tr>
<td>$\frac{2s}{\sqrt{3}} P'a^+$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
</tr>
<tr>
<td>$- 2 \sqrt{s} G k_z a^+$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
<td>$- \frac{1}{\sqrt{2}} \gamma_2 [2k_z^2 - s(2n+1)]$</td>
</tr>
</tbody>
</table>

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<table>
<thead>
<tr>
<th>( E_p )</th>
<th>( \sqrt{3} P_k )</th>
<th>( -\sqrt{3} P_a )</th>
<th>( 0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \sqrt{3} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( N_{a_2} \left( s \left( 2n + 1 \right) \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
</tr>
<tr>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
</tr>
<tr>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
</tr>
<tr>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
<td>( \frac{1}{\sqrt{3}} \left( \frac{s \alpha^* - \mu^a}{G + \frac{1}{3} \alpha^*} \right) )</td>
</tr>
</tbody>
</table>
Table C3 The Pidgeon-Brown matrices, i.e. the result of operating the matrix of table 2 on the harmonic oscillator basis and considering only noncoupling terms with \( k_z = 0 \). This is equivalent to the assumption that all terms proportional to the inversion symmetry are negligible \((G = C = C' = q = N_1 = N_2 = N_3 = 0)\), and that the valence band warping term \( \mu \) is also negligible. In this case the off-diagonal 4 x 4 blocks disappear, i.e. \( H = \begin{bmatrix} H_{aa} & 0 \\ 0 & H_{bb} \end{bmatrix} \),

where \( H_{aa} = \)

<table>
<thead>
<tr>
<th>( E_z + (P + \frac{1}{2})s(2n+1) + (N_1 + \frac{1}{2})s )</th>
<th>( P\sqrt{sn} )</th>
<th>( \frac{-1}{\sqrt{3}} P \sqrt{s(n+1)} )</th>
<th>( \frac{2}{\sqrt{3}} P' \sqrt{s(n+1)} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( P\sqrt{sn} )</td>
<td>( (-\frac{1}{2} \gamma_1 - \frac{1}{2} \gamma_2)(2n+1)s )</td>
<td>( -\frac{3}{2} (\kappa + \frac{9}{4} q)s )</td>
<td>( \bar{\gamma} \sqrt{3n(n+1)s} )</td>
</tr>
<tr>
<td>( \frac{-1}{\sqrt{3}} P \sqrt{s(n+1)} )</td>
<td>( -\bar{\gamma} \sqrt{3n(n+1)s} )</td>
<td>( \frac{1}{\sqrt{2}} \gamma_1's(2n+3) )</td>
<td>( -\frac{1}{\sqrt{2}} (\kappa' + 1)s )</td>
</tr>
<tr>
<td>( \frac{2}{\sqrt{3}} P' \sqrt{s(n+1)} )</td>
<td>( -\bar{\gamma}' \sqrt{6n(n+1)s} )</td>
<td>( \frac{1}{\sqrt{2}} \gamma_2's(2n+3) )</td>
<td>( -\Delta - \frac{1}{2} \gamma_2's(2n+3) )</td>
</tr>
</tbody>
</table>

---

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\[
H_{bb} = \\
\begin{array}{|c|c|c|}
\hline
E_z + (F + \frac{1}{2}) s(2n+3) & \frac{1}{\sqrt{3}} P \sqrt{s(n+1)} & \frac{2}{\sqrt{3}} P' \sqrt{s(n+1)} \\
- (N_t + \frac{1}{2}) s & -P \sqrt{s(n+2)} & -\frac{1}{\sqrt{2}} (\kappa' + \frac{1}{2}) s \\
\hline
\frac{1}{\sqrt{5}} P \sqrt{s(n+1)} & -\frac{1}{2} (\gamma_1 - \gamma_2) s(n+1) & \frac{1}{\sqrt{2}} \gamma_2 s(n+1) \\
-\frac{1}{2} (\kappa + \frac{1}{4}) s & -\frac{1}{2} (\gamma_1 - \gamma_2) s(n+1) & -\frac{1}{\sqrt{2}} \gamma_2 s(n+1) \\
\hline
-\gamma_1 s(n+2) & -\gamma_2 s(n+1)(n+2) & \frac{1}{2} \gamma_2 s(n+1)(n+2) \\
\hline
\frac{1}{\sqrt{3}} P' \sqrt{s(n+1)} & -\gamma_2 s(n+1) & \frac{1}{\sqrt{2}} \gamma_2 s(n+1) \\
-\frac{1}{\sqrt{2}} (\kappa' + \frac{1}{2}) s & -\frac{1}{\sqrt{2}} (\kappa' + \frac{1}{2}) s & \frac{1}{\sqrt{2}} \gamma_2 s(n+1) \\
\hline
\end{array}
\]
Table C4 The terms that must be added to Table C1 if non-coupling $k_z \neq 0$, but still assuming all terms that couple harmonic oscillators are negligible ($G = C = C' = q = N_1 = N_2 = N_3 = \mu = 0$).

<table>
<thead>
<tr>
<th>$(F + \frac{1}{2})k_z^2$</th>
<th>0</th>
<th>0</th>
<th>0</th>
<th>0</th>
<th>$-\frac{2}{\sqrt{3}} P k_z$</th>
<th>0</th>
<th>$-\frac{1}{\sqrt{3}} P' k_z$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>$(-\frac{1}{2} \gamma_1 + \gamma_2)k_z^2$</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>$\sqrt{6n} \gamma_3 k_z$</td>
<td>0</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>$(-\frac{1}{2} \gamma_1 - \gamma_2)k_z^2$</td>
<td>$-\sqrt{2} \gamma_2' k_z^2$</td>
<td>$-\frac{2}{\sqrt{3}} P k_z$</td>
<td>0</td>
<td>$-\sqrt{6(n+2)} \gamma_3' k_z$</td>
<td>$3\sqrt{(n+1)} \gamma_3' k_z$</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>$-\sqrt{2} \gamma_2' k_z^2$</td>
<td>$-\frac{1}{2} \gamma_1' k_z^2$</td>
<td>$-\frac{1}{\sqrt{3}} P' k_z$</td>
<td>$3\sqrt{(n+1)} \gamma_3' k_z$</td>
<td>$-\sqrt{3(n+2)} \gamma_3' k_z$</td>
<td>0</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>$-\frac{2}{\sqrt{3}} P k_z$</td>
<td>$-\frac{1}{\sqrt{3}} P' k_z$</td>
<td>$(F + \frac{1}{2})k_z^2$</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$-\frac{2}{\sqrt{3}} P k_z$</td>
<td>$\sqrt{6n} \gamma_3 k_z$</td>
<td>0</td>
<td>$3\sqrt{(n+1)} \gamma_3' k_z$</td>
<td>0</td>
<td>$(-\frac{1}{2} \gamma_1 - \gamma_2)k_z^2$</td>
<td>0</td>
<td>$\sqrt{2} \gamma_2' k_z^2$</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>$-\sqrt{6(n+2)} \gamma_3' k_z$</td>
<td>$-\sqrt{3(n+2)} \gamma_3' k_z$</td>
<td>0</td>
<td>0</td>
<td>$(-\frac{1}{2} \gamma_1 + \gamma_2)k_z^2$</td>
<td>0</td>
</tr>
<tr>
<td>$-\frac{1}{\sqrt{3}} P' k_z$</td>
<td>$-\sqrt{3n} \gamma_3' k_z$</td>
<td>$3\sqrt{(n+1)} \gamma_3' k_z$</td>
<td>0</td>
<td>0</td>
<td>$\sqrt{2} \gamma_2' k_z^2$</td>
<td>0</td>
<td>$-\frac{1}{2} \gamma_1 k_z^2$</td>
</tr>
</tbody>
</table>

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Table C5 Including all coupling terms including non-zero \( k_2, G, C, C', q, N_1, N_2, N_3 \), and also \( \mu \neq 0 \). Only, the lower triangle is shown here. Note that this matrix is not Hermitian.

<table>
<thead>
<tr>
<th>( n )</th>
<th>( 0 )</th>
<th>( 2\sqrt{(n+1)sGk_z\phi_{n+1}} )</th>
<th>( -\sqrt{3(n-1)(n-2)\mu s\phi_{n-1} + Ck_z\phi_{n-1}} )</th>
<th>( \sqrt{6(n-1)(n-2)\mu s\phi_{n-1}} + \frac{1}{\sqrt{2}}Ck_z\phi_{n-1} )</th>
<th>( 0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 2\sqrt{\frac{n}{3}}Gk_z\phi_{n-1} )</td>
<td>( -\sqrt{3(n-1)(n-2)\mu s\phi_{n-1} + Ck_z\phi_{n-1}} )</td>
<td>( 0 )</td>
<td>( 0 )</td>
<td>( 0 )</td>
<td></td>
</tr>
<tr>
<td>( -2\sqrt{\frac{2n}{3}}G'k_z\phi_{n-1} )</td>
<td>( \sqrt{6(n-1)(n-2)\mu s\phi_{n-1}} + \frac{1}{\sqrt{2}}C'k_z\phi_{n-1} )</td>
<td>( -\sqrt{\frac{2}{3}G + (\sqrt{3}N_2)s\sqrt{n(n+1)}\phi_{n-1}} + \sqrt{\frac{2}{3}G - (\sqrt{3}N_2)}s\sqrt{(n+1)(n+2)}\phi_{n+1} )</td>
<td>( -\sqrt{\frac{2}{3}G + (\sqrt{3}N_2)s\sqrt{n(n+1)}\phi_{n-1}} + \sqrt{\frac{2}{3}G - (\sqrt{3}N_2)}s\sqrt{(n+1)(n+2)}\phi_{n+1} )</td>
<td>( 0 )</td>
<td></td>
</tr>
<tr>
<td>( N_2[2k_2^2 - s(2n-1)]\phi_{n-1} + N_3s\phi_{n-1} )</td>
<td>( -\sqrt{\frac{2}{3}G + (\sqrt{3}N_2)s\sqrt{n(n+1)}\phi_{n-1}} + \sqrt{\frac{2}{3}G - (\sqrt{3}N_2)}s\sqrt{(n+1)(n+2)}\phi_{n+1} )</td>
<td>( -\frac{1}{\sqrt{3}}Gs\sqrt{n(n+1)}\phi_{n-1} - \sqrt{(n+2)(n+3)}\phi_{n+3} )</td>
<td>( -\frac{1}{\sqrt{3}}Gs\sqrt{n(n+1)}\phi_{n-1} - \sqrt{(n+2)(n+3)}\phi_{n+3} )</td>
<td>( 0 )</td>
<td></td>
</tr>
</tbody>
</table>

Continued overleaf
<table>
<thead>
<tr>
<th>Term</th>
<th>Term</th>
<th>Term</th>
<th>Term</th>
<th>Term</th>
<th>Term</th>
</tr>
</thead>
<tbody>
<tr>
<td>(-N_s (2k_s^2 - s(2n+1))\phi_n + N_s s\phi_n)</td>
<td>(-\frac{\sqrt{3}}{2} C \sqrt{n} s \phi_n)</td>
<td>(-\frac{\sqrt{2}}{2} C' \sqrt{(n+1)} s \phi_n)</td>
<td>(-\frac{1}{2} C' \sqrt{(n+1)} s \phi_n)</td>
<td>(2\sqrt{2} s(n+1) G k_2 \phi_n)</td>
<td>(-\sqrt{3(n+1)} \mu s \phi_{n+2})</td>
</tr>
<tr>
<td>(-\frac{1}{\sqrt{3}} G \sqrt{n(n-1)} \phi_{n-2})</td>
<td>(\frac{1}{2} C' \sqrt{(n-1)} s \phi_{n-2})</td>
<td>(\sqrt{3} C' \sqrt{(n+2)} s \phi_{n+2})</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>(-\frac{1}{\sqrt{3}} G \sqrt{n(n-1)} \phi_{n-2})</td>
<td>(\frac{1}{2} C' \sqrt{(n-1)} s \phi_{n-2})</td>
<td>(\sqrt{3} C' \sqrt{(n+2)} s \phi_{n+2})</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>(-\frac{1}{\sqrt{3}} G \sqrt{n(n-1)} \phi_{n-2})</td>
<td>(\frac{1}{2} C' \sqrt{(n-1)} s \phi_{n-2})</td>
<td>(\sqrt{3} C' \sqrt{(n+2)} s \phi_{n+2})</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>(-\frac{1}{\sqrt{3}} G \sqrt{n(n-1)} \phi_{n-2})</td>
<td>(\frac{1}{2} C' \sqrt{(n-1)} s \phi_{n-2})</td>
<td>(\sqrt{3} C' \sqrt{(n+2)} s \phi_{n+2})</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>
D Appendix: Program for computing the 8x8 k.p matrix

The computer program has been written in Fortran language to compute the 8×8 k.p matrix for \( k_z = 0 \) with changing magnetic field B via parameter s (\( s = eB/m_0 \)), see Table C2. We can also consider different \( k_z (k_z \neq 0) \), by adding the elements from Table C4 when we keep the B constant similar to our experimental conditions in a permanent magnetic field, but the result is the same as the previous one. We can also keep other coupling terms besides of \( k_z \) including C, C', G, N_1, N_2, N_3, q, \( \mu \neq 0 \), but the procedure is long and complicated without considerable difference in the result.

Program InSb PB

C compute the PB landau levels

PARAMETER (Nbands=8)
REAL*8 H(Nbands,Nbands)
REAL*8 E(Nbands), V(Nbands,Nbands), Space(Nbands)
REAL*8 Energy(1000)
C IMPLICIT DOUBLE PRECISION (A-H,O-Z)
C implicit integer (i-n)
integer count, i
real*8 Nmax, N
real*8 kz, kzmax
real*8 Eg, Ep, Delta
gaml, gaml2, gaml3, kapl, gamlbar
real*8 gaml, gam2, gam3, kap, gambar, mu
real*8 F, Nl
real*8 mc, mlh, mhh, g
real*8 B, s, Bmax, points
real*8 r2, Eupper, Elower
real*8 Eg0, alpha, beta, T

external F02ABF

open(3, file='temp.dat')

C InSb params
Eg0= 0.2352d0
Ep= 23.1d0
Delta=0.803d0
F=-0.5d0
Nl=-0.65d0
gaml=3.1d0
gamlbar=-0.20d0
mu=0.55d0
kap=-1.3d0
C InSb params 300K
alpha=0.32d-3
beta=170d0
T=300d0
Eg=Eg0-alpha*T**2/(T+beta)
write (*,*) 'Eg0=' ,Eg

params
Eg= 1.519d0
Ep= 27.86d0
Delta=0.341d0
F=-1.7d0
N1=-0.0495d0
gaml=0d0
gambar=0d0
mu=0d0
kap=0d0
gaml1 = gaml+Ep/(3d0*Eg)
gamlbar = gambar+Ep/(6d0*Eg)
gaml2 = gamlbar-mu
gaml3 = gamlbar+mu
gapl = kap+Ep/(6d0*Eg)
mc = 1d0/(1d0+2d0*F+Ep/3d0*(2d0/Eg+1d0/(Eg+Delta)))
mlh = 1d0/(gaml1+2d0*gamlbar)
mhh = 1d0/(gaml1-2d0*gamlbar)
g = 2d0*(1d0+2d0*N1-Ep/3d0*(1d0/Eg-1d0/(Eg+Delta)))
write(*,*), 'mc, mlh, mhh, g'
write(*,*), mc, mlh, mhh, g
write(*,*), 'gaml1, gaml2, gaml3, gapl'
write(*,*), gaml1, gaml2, gaml3, gapl
write(*,*), 'gam2,3->gambar'
gam2=gambar-mu
gam3=gambar+mu

write (3,55) 'B','E1','E2','E3','E4','E5','E6','E7','E8'
+ 'HH-2b',
+ 'SO1a','LSO1a','HH-1b','CB-1b',
+ 'SO0','SO0','LS00','LH00','HH00','CB00','CB00',
+ 'SO1','SO1','LSO1','LH1','HH1','HH1','CB1','CB1',
+ 'SO2','SO2','LSO2','LH2','HH2','HH2','CB2','CB2'
1 format(50A14)

Eupper=3d0
Elower =0.1d0
Nmax = 2d0
points = 200d0
r2 = dsqrt(2d0)
kzmax = dsqrt(2d0*mc*0.2d0)
write (*,*) kzmax
write (*,*) kz=kzmax/10d0
kz=0d0
write (*,*) kz
Bmax=20d0
B=10d0

DO 10 kz = kzmax/points , kzmax, kzmax/points
DO 10 B = Bmax/points, Bmax, Bmax/points
   s = B/(2.35d5)*27.2d0
   c write (*,*)'s=',s
   c NB B in T, s in Ev (not Hartrees)
   n=-2d0
   c for n=-2, only state 4
   H(4,4)=-s*((gam1+gam2)*(n-2.5d0)-1.5d0*kap)
   count = 1
   Energy(1)=H(4,4)
   if ( (H(4,4) .GE. Elower) .and. (H(4,4) .LE. Eupper)) then
      Energy(count)=H(4,4)
      count = count+1
   endif
   CALL CLEAR(H,E,V,Nbands)
   n=-1d0
   c for n=-1, only rows 5,7,2,4
   c cb diagonals
   H(2,2)=((2d0*F+ld0)*(n+1.5d0)-(N1+0.5d0))*s+Eg
   c cb to vb coupling, same set
   H(4,2)=-dsqrt((n+2d0)*Ep*s/2d0)
   c vb diagonals
   H(4,4)=-s*((gam1+gam2)*(n-2.5d0)-1.5d0*kap)
   H(5,5)=-s*((gam1-gam2)*(n+1.5d0)-0.5d0*kap)
   H(7,7)=-s*(gam1*(n+1.5d0)-(kap+0.5d0))-delta
   c vb to vb coupling, same set
   H(7,5)=s*r2*(gam2*(n+l.5d0)-(kap+1d0)/2d0)
   CALL F02ABF(H,Nbands,Nbands,E,V,Nbands,Space,Ifail)
   do 31 i = 1,Nbands
      if ( (E(i) .GE. Elower) .and. (E(i) .LE. Eupper)) then
         Energy(count)=E(i)
         count = count+1
      endif
   1 continue
   CALL CLEAR(H,E,V,Nbands)
   n=Od0
   c for n=0 only rows 1,5,7,2,6,4,8 (row 3 missing)
c2345678901234567890123456789012345678901234567890123456789
   0
c cb diagonals
   H(1,1)=((2d0*F+1d0)*(n+0.5d0)+(N1+0.5d0))*s+Eg
   H(2,2)=((2d0*F+1d0)*(n+1.5d0)-(N1+0.5d0))*s+Eg
c cb to vb coupling, same set
H(5,1)=dsqrt ((n+1d0)*Ep*s/6d0)
H(7,1)=dsqrt ((n+1d0)*Ep*s/3d0)
H(6,2)=dsqrt ((n+1d0)*Ep*s/6d0)
H(4,2)=dsqrt ((n+2d0)*Ep*s/2d0)
H(8,2)=dsqrt ((n+1d0)*Ep*s/3d0)

H(7,5)= s*r2*(gam2*(n+1.5d0)-(kap+1d0)/2d0)
H(6,4)= s*gambar*dsqrt (3d0*(n+1d0)*(n+2d0))
H(8,4)= s*gambar*dsqrt (6d0*(n+1d0)*(n+2d0))
H(8,6)= -s*r2*(gam2*(n+0.5d0)+(kap+1d0)/2d0)

CALL F02ABF(H,Nbands,Nbands,E,V,Nbands,Space,Ifail)

do 32 i = 1,Nbands
if ( (E(i) .GE. Elower) .and. (E(i) .LE. Eupper)) then
  Energy(count)=E(i)
  count = count+1
endif
1 continue

do 77 n=1d0,Nmax
CALL CLEAR(H,E,V,Nbands)

H(1,1)=((2d0*F+1d0)*(n+0.5d0)+(N1+0.5d0))*s+Eg
H(2,2)=((2d0*F+1d0)*(n+1.5d0)-(N1+0.5d0))*s+Eg

H(3,1)= dsqrt ( n *Ep*s/2d0)
H(5,1)=dsqrt ((n+1d0)*Ep*s/6d0)
H(7,1)=dsqrt ((n+1d0)*Ep*s/3d0)
H(6,2)=dsqrt ((n+1d0)*Ep*s/6d0)
H(4,2)=dsqrt ((n+2d0)*Ep*s/2d0)
H(8,2)=dsqrt ((n+1d0)*Ep*s/3d0)

H(3,3)=-s*((gambar+gam2)*(n-2.5d0)-1.5d0*kap)
H(4,4)=-s*((gambar+gam2)*(n-1.5d0)-0.5d0*kap)
H(5,5)= s*((gambar+gam2)*(n+0.5d0)+0.5d0*kap)

H(7,7)=-s*(gambar*(n+1.5d0)-(kap+0.5d0))-delta
H(8,8)=-s*(gambar*(n+0.5d0)+(kap+0.5d0))-delta
SUBROUTINE CLEAR (H, E, V, N)
C clear arrays
REAL*8 E(N), V(N,N)
REAL*8 H(N,N)
INTEGER N, I, J
DO 1 I=1, N
  E(I) = 0.0
  DO 1 J=1, N
     V(I,J) = 0.0
     H(I,J) = 0.0
1 CONTINUE
END
Figure D1 shows a typical solution of the $8 \times 8$ k.p Fortran program, giving the Landau energies by changing the magnitude of the external magnetic field $B$ for the conduction band, heavy hole and light hole of the valence band. It is clear that by changing the $B$, energy splittings change. From these results for $n^{th}$ Landau level with two spin directions, we can estimate the average and difference energies, for example, in the conduction band. Then by using $E(n_+)-E(n_-)=g^* \mu_B B$, for each amount of $B$, g-factor can be achieved versus average energies ($E_{av}=[E(n_+)+E(n_-)]/2$).

In our experiment we have used $B=0.288$ T, the diagrams of $g^*(E)$ for InAs and InSb at different temperatures can be seen in Figures 5.11, and 5.18.
Figure D1 Typical solution of the $8 \times 8$ k.p Fortran program, giving the Landau energies versus magnetic field for the conduction band, heavy hole and light hole of the valence band.

In this case maximum landau level, used in the program is $n=2$. 

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