Universidade de São Paulo
Instituto de Física

**Controle ótico e detecção de coerência de spin em sistemas
de multicamadas**

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**Optical control and detection of spin coherence in
multilayer systems**

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Dedication

It is with my deepest gratitude and warmest affection that I dedicated this thesis to:

My sweet family whose love, encouragement and prayers of day and night made me able to get such success and honor. Who always picked me off on time and encouraged me to go on every adventure, especially this one.

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Abstract

Since a decade, spintronics and related physics have attracted considerable attention due to the massive research conducted in these areas. The main reason for growing interest in these fields is the expectation to use the electron's spin instead of or in addition to the charge for the applications in spin-based electronics, quantum information, and quantum computation. A prime concern for these spins to be possible candidates for carrying information is the ability to coherently control them on the time scales much faster than the decoherence times.

This thesis reports on the spin dynamics in two-dimensional electron gases hosted in artificially grown III-V semiconductor quantum wells. Here we present a series of experiments utilizing the techniques to optically control the spin polarization triggered by either optical and electrical methods i.e. well known pump-probe technique and current-induced spin polarization.

We investigated the spin coherence in high mobility dense two-dimensional electron gas confined in GaAs/AlGaAs double and triple quantum wells, and, it's dephasing on the experimental parameters like applied magnetic field, optical power, pump-probe delay and excitation wavelength. We have also studied the large spin relaxation anisotropy and the influence of sample temperature on the long-lived spin coherence in triple quantum well structure. The anisotropy was studied as a function sample temperature, pump-probe delay time, and excitation power, where, the coherent spin dynamics was measured in a broad range of temperature from 5 K up to 250 K using time-resolved Kerr rotation and resonant spin amplification. Additionally, the influence of Al concentration on the spin dynamics of AlGaAs/AlAs QWs was studied. Where the composition engineering in the studied structures allows tuning of the spin dephasing time and electron g -factor.

Finally, we studied the macroscopic transverse drift of long current-induced spin coherence using non-local Kerr rotation measurements, based on the optical resonant amplification of the electrically-induced polarization. Significant spatial variation of the electron g -factor and the coherence times in the nanosecond scale transported away half-millimeter distances in a direction transverse to the applied electric field was observed.

Resumo

Há uma década, a spintrônica e outras áreas relacionadas vêm atraindo considerável atenção, devido a enorme quantidade de pesquisa conduzidas por elas. A principal razão para o crescente interesse neste campo é a expectativa da aplicação do controle do spin do elétron no lugar ou em adição à carga, em dispositivos eletrônicos e informação e computação quânticas. A possibilidade destes spins carregarem informação depende, primeiramente, da habilidade de controlá-los coerentemente, em uma escala de tempo muito mais rápida do que o tempo de decoerência.

Esta tese trata da dinâmica de spins em gases de elétrons bidimensionais, em poços quânticos de semicondutores III-V, crescidos artificialmente. Nós apresentamos uma série de experimentos, utilizando técnicas para o controle ótico da polarização de spin, desencadeadas por métodos óticos e eletrônicos, ou seja, técnicas conhecidas de bombeio e prova e polarização de spin induzida por corrente.

Nós investigamos a coerência de spin em gases bidimensionais, confinados em poços quânticos duplos e triplos de GaAs/AlGaAs e a dependência da defasagem com parâmetros experimentais, como campo magnético externo, potência ótica, tempo entre os pulsos de bombeio e prova e comprimento de onda da excitação. Também estudamos a grande anisotropia de relaxação de spin como função da temperatura da amostra, potência de excitação e defasagem entre bombeio e prova, medidos para uma vasta gama de temperatura, entre 5 K e 250 K, usando Rotação de Kerr com Resolução Temporal (TRKR) e Amplificação Ressonante de Spin (RSA). Além disso estudamos a influência da concentração de Al na dinâmica dos poços de AlGaAs/AlAs, para o qual a engenharia da composição da estrutura permite sintonizar o tempo de defasagem de spin e o fator g do elétron.

Por fim, estudamos a deriva transversal macroscópica da longa coerência de spin induzida por corrente, através de medidas de Rotação de Kerr não-locais, baseadas na amplificação ressonante ótica da polarização eletricamente induzida. Observamos uma variação espacial significativa do fator g e do tempo de vida da coerência, na escala de nanosegundos, deslocada distâncias de meio milímetro na direção transversa ao campo magnético aplicado.

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Nomenclature

List of abbreviations

2DEG	Two-dimensional electron gas
AC	Alternating current
AMR	Anisotropic magnetoresistance
BS	Beam splitter
CB	Conduction band
CIP	Current-in-the-plane
CISP	Current-induced spin polarization
CPP	Current-perpendicular-to-the-plane
DP	Dyakonov-Perel mechanism
DQW	Double quantum well
F	Filter
FFT	Fast Fourier transform
GMR	Giant magnetoresistance
KR	Kerr rotation
LLs	Landau levels
LPE	Liquid phase epitaxy
MBE	Molecular beam epitaxy
MIS	Magnetointersubband
MOCVD	Metal organic chemical vapor deposition
MOSFET	Metal-oxide-semiconductor field-effect transistor

MOVPE	Metal organic vapor phase epitaxy
MR	Magnetoresistance
MRAM	Magnetic random access memory
OL	Objective lens
PEM	Photo-elastic modulator
RSA	Resonant spin amplification
SdH	Shubnikov-de-Haas
sFET	Spin field effect transistor
SHE	Spin Hall effect
SOC	Spin-orbit coupling
SOI	Spin-orbit interaction
SPSL	Short-period superlattices
TQW	Triple quantum well
TRKR	Time-resolved Kerr rotation
VB	Valance band
VPE	Vapor phase epitaxy
VTI	Variable temperature insert

List of symbols

A	Amplitude
a	Lattice constant
α	Rashba spin-orbit coupling parameter
B_{so}	Spin-orbit field
$B_{1/2}$	Full width at half maximum
$B_{\perp}, B_{\parallel}$	Components of B_{SO} perpendicular and parallel to external magnetic field
β_1	Linear spin-orbit constant due to the Dresselhaus bulk inversion asymmetry

β_3	Cubic spin-orbit constant due to the Dresselhaus bulk inversion asymmetry
c	Speed of light
D_c	Charge diffusion constant
D_s	Spin diffusion constant
E	Kinetic energy
$\vec{E}$	Electric field
E_g	Band gap energy
f	Frequency
g	Landé factor
h	Planck's constant
$\hbar$	Reduced Planck's constant
He	Liquid helium
I	Electric current
$\vec{J}$	Total angular momentum
$\vec{j}$	Current density
$\vec{K}$	Wave vector
L	Length
L	Orbital angular momentum
ℓ	Mean free path
ℓ_s	Spin transport length
$\vec{M}$	Magnetic moment
m	The electron rest mass
m^*	Effective mass
μ	Mobility
μ_B	Bohr magneton
n_s	Total electron density

$\vec{p}$	Momentum
R_{xx}	Longitudinal resistance
R_{xy}	Hall resistance
$R_{\uparrow\uparrow}$	Resistanc in parallel orientation of magnetization
$R_{\uparrow\downarrow}$	Resistanc in antiparallel orientation of magnetization
$\vec{S}$	Spin angular momentum
T	Temperature
T_2^*	Ensemble dephasing time
Δt	Pump-probe delay
t_m	Momentum relaxation time or transport scattering time
τ_p^*	Single-electron momentum scattering time
τ_q	Quantum life time
t_{rep}	Repetition period
τ_y	Relaxation time for electron spins oriented perpendicular to the QW growth direction.
τ_z	Relaxation time for electron spins oriented parallel to the QW growth direction.
τ_z/τ_y	Spin relaxation anisotropy
ν	Filling factor
ν_v	Verdet constant
V_{AC}	AC voltage
$\vec{v}_d$	Drift velocity
$\vec{v}_F$	Fermi velocity
w	Width
ω_c	Cyclotron frequency
ω_L	Larmor frequency
ρ	Resistivity tensor

Θ_F	Faraday rotation angle
Θ_K	Kerr rotation angle
φ	Phase
Δ_{ij}	Occupied subband separation energies (i,j = 1,2,3)
λ	Wavelength
λ_o	de-Broglie wavelength of a free electron
$\vec{\sigma}$	Electron density
σ^+	Right circularly polarized light
σ^-	Left circularly polarized light

Physical Constants

c	Speed of light in a vacuum	$2.998 \times 10^{-34} \text{ m/s}$
e	Electron charge	$1.60217662 \times 10^{-19} \text{ C}$
h	Planck's constant	$6.62607 \times 10^{-34} \text{ Js}$
$\hbar$	Reduced Planck's constant	$1.054571800(13) \times 10^{-34} \text{ Js}$
m	Electron mass	$9.10938356 \times 10^{-31} \text{ Kg}$
μ_B	Bohr magneton	$9.274 \times 10^{-24} \text{ J/T}$

Chapter 1

Introduction and motivation

The 1956 Nobel Prize in physics was given jointly to W. B. Shockley, J. Bardeen, and W. H. Brattain for their researches on semiconductors and their remarkable discovery of transistor effect, which has revolutionized the field of electronics and paved the way for future technology. Today, no electronic device would work without transistor which is the workhorse of modern technology, and the tremendous impact it has on the information age cannot be underestimated. Before the transistor the technology available at the time was based on vacuum tubes which were consuming a lot of power, giving off too much heat, and were unreliable. With this invention, the vacuum-tube-based computers that occupied an entire room have reduced to slender computers of the size of few cm^2 and have advanced at an astonishing rate. This rapid improvement has played a vital role in driving the global economy and have changed the way we store and process information.

The miniaturization obeying Moore's Law [1], which states that the number of transistors on a circuit would roughly double every 18 months, has lead to an exponential increase in the packing density of a microprocessor. This empirical law (was properly valid till 1980) remained prescient to the last fifty years cannot be sustained much longer. However, in the semiconductor industry, problems arises as the dimension of transistors are getting smaller and smaller (nanometric size) to integrate more transistors on the same area of a chip. One of the problems is device heating, caused by the higher resistance of device due to a smaller size. The temperature of the device can reach a certain regime where its failure can occur. On the other hand, much energy is consumed. However, if we further continue to shrink the dimension of transistor we are crossing the quantum size boundaries where the quantum mechanical phenomena become important, and the flow of electrons can no longer be controlled. It is, therefore, a primary task to find physical effects that might overcome these fundamental limits. One alternative to these problems is to encode the information using the quantum characteristic of the electron: its spin.

1.1 A journey toward spintronics

One of the most striking and novel discoveries in 1799/1800 was the invention of first electrical battery by Alessandro Volta [2]. In 1780, Galvani, the Italian physician, and anatomist was experimenting with dissected frog legs, discovered that the muscles of a frog would contract when touched by a metallic object. He believed the energy that drives this contraction was being generated by the frog muscles and called it animal electricity [3, 4]. However, Volta disagreed, believing that the frog muscles were just reacting to the electricity and this phenomenon was created by two different metals joined mutually by a moist intermediary, not by frog muscles. To prove Galvani wrong, Volta initiated a series of experiments by stacking different metallic discs on top of each other, separated by brine-soaked cardboard or cloth. He built a pile, consisted of many discs, nowadays known as the Voltaic pile. By connecting a wire to both ends of the pile, a steady current flow was detected. This invention made possible many new experiments and led to pronounced discoveries, such as the first electrolysis of water and the investigation of electricity.

In 1906, J. J. Thomson was awarded Nobel Prize for his discovery of elementary charged particles emitting from such current sources: the electrons [5]. Its particle nature was experimentally confirmed by keeping a glass paddlewheel in its path and noticed that the paddlewheel rotated, and moved towards the anode which strongly suggested that the cathode rays had momentum, and thus the mass. About 30 years later his son G. P. Thompson shared Nobel prize with C. Davisson for his achievement that a beam of electrons could be diffracted. He interpreted this as evidence for the wave nature of the electrons [6]. From its deflection towards the anode, it was confirmed that the electrons also carry a negative charge, which is the basis of modern electronics which relies on the storage and flow of such charges within a material to encode digital information. The charge is a scalar quantity and only have magnitude; therefore, its physical presence and absence could be implemented by the binary bits 0_s and 1_s . The same approach has adopted in metal–oxide–semiconductor field-effect transistor (MOSFET) to encode the ON and OFF states, which has driven the semiconductor information technology to remarkable heights.

In the 20th century, it finally became clear that electrons do not only possess mass and charge but contain an additional degree of freedom, which is intrinsic to electrons called spin. The concept of spin was first theoretically postulated by George Uhlenbeck, and Samuel Goudsmit [7] in 1925 and interpreted as a fourth quantum number, which was proposed by Wolfgang Pauli [8]. The first experiments which needed the spin for its explanation include the observation of Einstein–de Haas effect [9] in 1915 and the famous experiment by Otto Stern and Walter Gerlach [10, 11] in 1922. The concept of spin was

very successful and could simultaneously explain these earlier experiments. Thus, the detected splitting of silver atoms by passing through an inhomogeneous magnetic field on the Stern-Gerlach experiment is considered as the first experimental observation of the electron spin, which, in contrast to classical observations, takes only certain discrete values. It is the spin that distinguishes Fermions with half-integer spin from Bosons with integer spin and determines the statistics of the elementary particles. Spin is a pseudovector (spinor) that has a fixed magnitude, but a variable direction (or polarization) and its rotation is governed by the Pauli spin matrices. The electron's spin is a quantum mechanical parameter, and could potentially be used as building blocks (qubits) for a quantum computer. For a free electron, the spin can assume two states up $|\uparrow\rangle$ and down $|\downarrow\rangle$, and therefore can be used in information processing and storage just like the binary bits of 0 and 1. One of the essential difference of qubits compared to a classical bit is that the spin states can be a superposition of these two states. In general it can therefore be written as $A|\uparrow\rangle + B|\downarrow\rangle$, where A and B are complex numbers.

Spintronics [12,13], or spin electronics is a continuously expanding area of research, which was developed mainly in the last few decades. Spintronics, an emerging interdisciplinary field of physics whose name indicates the wish to do electronics with the electron spin in addition to or in place of its charge. Using electrons spin rather than electrons charge may overcome the shortcoming of heating and low energy loss devices. Additionally, the electron's spin has inherent merits to provide electronic devices with new functionalities at the same time with non-volatility and increased data processing speed.

In the last few decades, several magnetoresistance (MR) effects were studied. MR is the variation in electrical resistance of a magnetic multilayered structure in response to an applied magnetic field. The ordinary magnetoresistance or anisotropic magnetoresistance (AMR) effect in ferromagnetic materials was discovered by William Thomson in 1857 [14]. In this effect, the dependence of electrical resistance of ferromagnetic transition metals on the angle between the orientation of the magnetization and the direction of electric current was observed. In AMR, the amplitude of an MR effect was very small around 2-4% and was utilized in the generation of magnetic random access memories AMR-MRAM. The discovery of giant magnetoresistance (GMR) effect in 1988 is considered as a birth of spintronics. In this effect, it was shown independently by the groups of Albert Fert [15] and Peter Grünberg [16] that resistance of materials comprised of alternating magnetic and non-magnetic layers changes greatly with the magnitude of external magnetic field (B_{ext}) applied in the layers plane. The observed change in the electrical resistance in (Fe/Cr) multilayers was of the order of 80% at $B_{ext} = 2$ T, which was much larger than AMR and was, therefore, called "Giant magnetoresistance". The B_{ext} changes the relative orientation of the magnetization in the successive magnetic layers. For example, with the application of B_{ext} in a certain direction, the magnetic

FIGURE 1.1: Simple multilayer stacks in current-perpendicular-to-the-plane (b) and current-in-the-plane (CIP) configuration. Developed from Ref. [17].

configuration departs from antiparallel and for high enough field all the magnetizations get aligned in the same direction. Due to spin-dependent scattering effects occurring in bulk and at the interfaces of magnetic layers, the change in orientation of the magnetization in the multilayers system strongly influences the electrical resistance of the system. The resistance ($R_{\uparrow\uparrow}$) of the structure is relatively low for parallel orientation of magnetization in the ferromagnetic layers, while the antiparallel orientation of magnetization is characterized by an electrical state of high resistance ($R_{\uparrow\downarrow}$). The GMR effect size is defined as:

$$GMR = \frac{\Delta R}{R} = \frac{R_{\uparrow\downarrow} - R_{\uparrow\uparrow}}{R_{\uparrow\uparrow}} \quad (1.1)$$

The GMR effect has been observed in two configurations: current-perpendicular-to-the-plane (CPP) and current-in-the-plane (CIP) schematically shown in Fig. 1.1. In the case of CPP configuration, see (Fig. 1.1(a)), the current flows in a direction which is perpendicular to the layers planes while in CIP the current is applied parallel to the layers planes as depicted in Fig. 1.1 (b). For a better understanding of GMR phenomenon, we started with the simple trilayer structure that consists of a nonmagnetic metal layer sandwiched between two ferromagnetic layers i.e. F/N/F. To qualitatively explain the GMR effect we use the simple *resistor model* [17] as illustrated in Fig. 1.2 for the CPP configuration. In the F/N/F configuration, each magnetic layer is represented by two resistors of different resistances; one for the spin $\uparrow$ and the second for the spin $\downarrow$. The electrons are weakly scattered when they pass through the ferromagnet with the spin orientations parallel to its magnetization while strongly scattered in the case of antiparallel configuration see Fig. 1.2 (a) and (b). Depending on the orientation of layer's magnetization, the smaller resistor, r , is associated to the spin parallel to the layer magnetization, while the larger one, R , is associated to the spin antiparallel as illustrated schematically in Fig. 1.2 (c) and (d). For the ferromagnetic layers with

FIGURE 1.2: *Resistor model of GMR: The electron scattering in the trilayer system in case of (a) parallel and (b) antiparallel magnetic configurations. Figs. (c) and (d) depicts the corresponding resistor model for (a) and (b) [17].*

parallel magnetization ($\uparrow\uparrow$), the spin $\uparrow$ electrons have a combined resistance $2r$, and the spin-down electrons see a resistance of $2R$, so the total combined resistance of the trilayer in the parallel configuration is:

$$R_{\uparrow\uparrow} = \frac{2rR}{r+R} \quad (1.2)$$

Similarly, in the antiparallel configuration of magnetizations ($\uparrow\downarrow$), both spin $\uparrow$ and spin $\downarrow$ electrons have the same resistance of $(r+R)$, so the overall resistance in the antiparallel configuration can be written as:

$$R_{\uparrow\downarrow} = \frac{1}{2}(r+R) \quad (1.3)$$

Finally, inserting Eqs. 1.2 and 1.3 into the definition of GMR which leads to:

$$GMR = \frac{\Delta R}{R} = \frac{1}{4} \frac{(r-R)^2}{rR} \quad (1.4)$$

Which is obviously a positive value confirming that $R_{\uparrow\downarrow} > R_{\uparrow\uparrow}$, which qualitatively explains the phenomenon of GMR. The discovery of the GMR effect motivated scientific community to exploit its technological potential. In a remarkably short period, only a decade, engineers successfully implemented in the development of hard disk drives in the readout head. IBM was the first company to market the GMR-based hard disk drives in 1997. Also, new types of non-volatile memory (i.e., retains memory when power

is off): Magnetoresistive Random Access Memory (MRAM) which was announced in 2003 [18] has come onto the scene that is based on the concept of using the direction of magnetization to store information and magnetoresistance as the data readout.

Both MRAM and GMR devices are based on metallic magnetic materials rather than semiconductors and are more appropriately described as magnetoelectronic [13, 19]. However, conventional electronics, on the other hand, is dominated by semiconductors for obvious reasons and, therefore, it is of great importance to consider the potential of semiconductor spintronic devices. Semiconducting materials are the backbone of modern electronics and offer a broad range of functionality, controllable through combinations of material composition and doping. Combining these advantages of semiconducting materials with the concept of spins and thereby introducing the spin degree of freedom into semiconductor devices is, therefore, a logical step and opens a new platform for spintronics.

The fundamental studies in spintronics include the generation, manipulation, and detection of spin polarization. The basic concept of spintronics can be explained with the help of spin field effect transistor (sFET) proposed by Supriyo Datta and Biswajit Das [20] in 1990, as depicted in Fig. 1.3. While this may or may not be a practical or useful device, however, it captures the above-said features of a spintronic device. The sFET consists of a semiconductor two-dimensional electron gas (2DEG) acting as a transport channel between the ferromagnetic source and drains electrodes and a Schottky gate above the channel for controlling the current. The source contact provides spin-polarized electrons which are injected into the semiconducting channel of length (L) and travel ballistically to the detection contact. Once the spin is injected, it is important to manipulate or control it. One possibility is to apply B_{ext} to rotate the spin. Also, an effective magnetic field originating from the spin-orbit coupling (SOC) due to the confinement of the channel allows one to control spin electronically which can be tailored by applying gate voltage [21]. After traversing the semiconductor channel, the spin is either blocked or let passed by the ferromagnetic drain contact, depending on the relative spin orientation and the magnetization of the drain contact. Thus, tuning the Rashba SOC parameter α by applying the gate voltage one can tune the channel length for spin precession as:

$$L = vt \tag{1.5}$$

According to the energy-time uncertainty

$$\Delta E \Delta t = \hbar \tag{1.6}$$

$$\alpha K \Delta t = \hbar \tag{1.7}$$

FIGURE 1.3: Schematic drawing of the proposed spin field effect transistor by Datta and Das. The spin polarization is injected from a ferromagnetic source contact into the semiconducting channel, where they are manipulated using a gate voltage, and then read out using a ferromagnetic drain contact. The current is controlled by the applied gate voltage through the Rashba spin-orbit interaction, being large if the electron spin at the drain is parallel to its initial direction (top), and small if antiparallel (bottom) [22].

$$\Delta t = t = \frac{\hbar}{\alpha K} \quad (1.8)$$

Inserting Eq. 1.8 in Eq. 1.5 we get

$$L = \frac{v\hbar}{\alpha K} = \frac{v\hbar^2}{\alpha K\hbar} = \frac{v\hbar^2}{\alpha p} = \frac{v\hbar^2}{\alpha m^* v} = \frac{\hbar^2}{\alpha m^*} \quad (1.9)$$

where m^* is the effective electron mass and $\hbar$, is the reduced Planck's constant. Since α can be tuned by modifying the gate voltage [23], the amount of precession, and thus the orientation of spins reaching the drain can be efficiently controlled. As a result, the current can be switched on and off through the modulation of the electron spins between the parallel and antiparallel configuration at the drain.

1.2 Focus of this thesis

The central theme of the research work presented in this thesis is to shine some light on the key issues described in the above-proposed device that is: (i) How can we generate spin polarization and spin currents in semiconductor materials, especially that in

multilayer GaAs/AlGaAs quantum QWs. (ii) How long the electron can maintain its spin coherence as it propagates through the channel i.e. how spin can carry the information before it is lost due to scattering or dephasing. (iii) How to detect these spin polarization before it lost. (iv) How to address various fundamental aspects of the spin ensemble like dephasing, decoherence, relaxation, g-factor distribution, and drift. (v) How can we efficiently measure the magnitude and direction of the spin-orbit field (B_{so})? (vi) Special focus is put on the understanding of spin dephasing times and its anisotropy in the different direction. Attaining longer lifetimes will aid in the quest for practical quantum computers. More specifically, we set out to characterize how these parameters will change when exposed to different experimental conditions like an external magnetic field, optical power, sample temperatures and pump-probe wavelength.

1.3 Structure of the thesis

This thesis is structured in eight chapters as follow: In Chapter 1 a brief motivation and a journey toward spintronics are reviewed and a short note on proposed sFET is given. In chapter 2, we provide a brief introduction to the fundamental concepts required for the understanding and analysis of the experimental data throughout the thesis. An emphasis is put on the important concepts like spin-orbit coupling and spin relaxation mechanisms. In chapter 3 we focus on the physics of time-resolved Kerr rotation, resonant spin amplification, and current-induced spin polarization. The sample details, as well as the experimental setup to perform those experiments, are discussed briefly. The experimental results are presented in five chapters, describing the observation of long-lived spin coherence (chapter 4), Large spin relaxation anisotropy (chapter 5), the robustness of spin coherence against high temperature (chapter 6), the effect of Al concentration on the spin dynamics (chapter 7), and macroscopic transverse drift of long current-induced spin coherence (chapter 8). Appendix A is devoted to the mathematical proofs of the equations used in the main text.

Chapter 2

Theoretical Background

For the better understanding of the results discussed in the following chapters, it is important to provide an overview of the basic principles of the physics of spin in semiconductors. This chapter is devoted to provide a background for the understanding of the spin-related phenomena in semiconductors especially GaAs. Therefore the overview starts with some basic of the semiconductor GaAs (Sec. 2.1). After a brief explanation of GaAs two-dimensional electron gas in Sec. 2.2, the spin precession around the applied magnetic field and the so-called Landé g-factor is discussed briefly in Sec. 2.3. Section 2.4 narrows the focus of the discussion on the spin-orbit coupling as it pertains directly to the analysis in chapters 5 and 8. In Sec. 2.5 a little focus is put on the metal-insulator transition. Finally, a brief overview of the main mechanism responsible for spin dephasing in semiconductor nanostructures is given in Sec. 2.7.

2.1 The Semiconductor GaAs

The samples investigated in this thesis are based on III-V semiconductor GaAs. Thus, it is essential to give an overview of the key details concerning the crystal structure, band structure and optical properties of this material.

2.1.1 Crystal structure of GaAs

The III-V semiconductor formed by the elements of group III and V of periodic table (GaAs in our case) crystallizes in the zinc-blende structure at normal pressure [24]. As schematically illustrated in figure 2.1, the zinc-blende crystal is characterized by two identical interlocked face-centred-cubic (f.c.c) sublattices. It has the same structure as diamond where the two sublattices are occupied by Ga (group III) and As (group V)

FIGURE 2.1: Zinc-blende crystal structure of GaAs. The crystal structure is characterized by single lattice constant a . (Taken from <http://elcrost.com/gaas-crystal-structure/gallium-arsenide-wikipedia-the-free-encyclopedia-gaas-crystal-structure-ppt-unit-cell-3d/>)

element ions, that are shifted against each other by one fourth along the cubic diagonal, that is $1/4(1,1,1)$. In such structure each gallium atom is tetrahedrally surrounded with four arsenic atoms. The crystal structure belongs to T_d point symmetry group which is invariant under the symmetry operations and is characterized by a single lattice constant $a = 5.65 \text{ \AA}$ at $T = 300 \text{ K}$.

2.1.2 Band structure of GaAs

Most of the properties quoted here are taken from Ref. [25,26]. GaAs is a direct band gap semiconductor, which means that in the energy-momentum space the minimum of the conduction band lies exactly above the maximum of the valence band. Where the energy difference between the top of the valence band and the minimum of the conduction band is the band gap energy, which is about $E_g = 1.519 \text{ eV}$ at $T = 0 \text{ K}$ and 1.424 eV at $T = 300 \text{ K}$ for GaAs. The empirical relation describing the temperature influence on the band gap is given by

$$E_g(T) = 1.519 - 5.408 \times 10^{-4} \frac{T^2}{T + 204} \text{ eV} \quad (2.1)$$

where T is in Kelvin. Fig. 2.2 shows the band structure of GaAs calculated by using pseudopotential method. The striking feature, which makes GaAs favorably suitable for optical studies, is the direct band gap at the Γ -point ($K = 0$) which allows direct excitation and recombination of electrons and holes. The conduction band (CB, Γ_6) has s-like character with orbital angular momentum ($L = 0$) and is two-fold degenerated due

FIGURE 2.2: The band structure of GaAs: The calculated band structure of GaAs using the pseudopotential method. The direct band gap and split-off hole band are highlighted by green and orange colors respectively. (Taken from [24])

to electron's spin, while the valence band (VB) is p-like with orbital angular momentum ($L = 1$). At Γ -point the dispersion relation of the band structure is parabolic as shown in Fig. 2.3, which is the magnified form of the encircle region shown in the complete band structure (Fig. 2.2). The valence band is somewhat more complicated due spin-orbit interaction (SOI) i.e., the spin, $\vec{S}$, is not a good quantum number anymore, instead the total angular momentum, $\vec{J} = \vec{L} + \vec{S}$, has to be used. At $K = 0$, the SOI splits the valence band into states with total angular momentum $\vec{J} = \vec{L} + \vec{S} = 3/2$ and $1/2$. The $\vec{J} = 3/2$ (Γ_8 -band) is four-fold degenerated, while $\vec{J} = 1/2$ (Γ_7 -band) is two-fold degenerated. The Γ_7 -band is called the split-off hole (so) band and is well separated from Γ_8 by the spin-orbit coupling energy $\Delta_{so} = 0.34$ eV and therefore, can not be accessed by optical pulses as long as the excitation energy does not exceed $E_g + \Delta_{so} = 1.860$ eV.

The Γ_8 -states are degenerate at $K = 0$ while, for $K \neq 0$ it further splits into two bands with different curvatures called heavy-hole (hh) and light-hole (lh) bands, named according to their different effective masses (See Fig. 2.3(a)). At $K = 0$ the dispersion relation for each band can be written as

$$E_n(K) = E_n(K = 0) + \frac{\hbar^2 K^2}{2m^*} \quad (2.2)$$

where the index n stands for CB, hh, lh and split-off hole bands. $E_n(K = 0)$ is the

FIGURE 2.3: The band structure of GaAs: The band structure of (a) bulk GaAs and (b) GaAs QWs near $K=0$. The conduction band (Γ_6) is separated from valence band by $E_g = 1.519$ eV. The valence band is split into heavy hole (hh, Γ_8) band, the light hole (lh, Γ_8) band, and the split-off (so, Γ_7) hole band. Due to SOI the sixfold degeneracy of the valence bands at $K=0$ is reduced to four-fold and the split-off band is shifted to lower energies by $\Delta_{so} = 0.34$ eV in respect to the hh and lh bands. (Adopted from [26])

energy of the band at Γ -point and m^* is the effective mass. In a quantum well, the degeneracy of light-hole and heavy-hole bands is further removed at $k=0$ by the QW potential (See Fig. 2.3(b)). According to Eq. 2.2 the dispersion relation for all the four bands (for bulk GaAs) at Γ -point can be written as:

$$E_c(K) = E_g + \frac{\hbar^2 K^2}{2m_e^*} \quad (2.3)$$

$$E_{hh}(K) = -\frac{\hbar^2 K^2}{2m_{hh}^*} \quad (2.4)$$

$$E_{lh}(K) = -\frac{\hbar^2 K^2}{2m_{lh}^*} \quad (2.5)$$

$$E_{so}(K) = -\Delta_{so} - \frac{\hbar^2 K^2}{2m_{so}^*} \quad (2.6)$$

Where m_e^* is the effective electron mass, m_{hh}^* is the effective heavy hole mass, m_{lh}^* is the effective light hole mass and m_{so}^* is the effective split-off hole mass. Some of the fundamental properties of GaAs with experimental conditions are summarized in table 2.1.

TABLE 2.1: Important parameter values for GaAs with experimental conditions.

Description	Symbol	Value	Unit	Ref.
Critical Si concentration for MIT ¹ (bulk)	n_c	2.0×10^{16}	cm^{-3}	[48, 105]
Critical Si concentration for MIT (QWs)	n_c	5.0×10^{10}	cm^{-2}	[51]
Band gap energy ($T = 0$ K)	E_g	1.519	eV	[25]
Effective electron mass ($T = 0$ K)	m_e^*	0.067	m_e	[25]
Effective heavy hole mass ($T \leq 100$ K)	m_{hh}^*	0.51	m_e	[25]
Effective light hole mass ($T \leq 100$ K)	m_{lh}^*	0.082	m_e	[25]
Effective split-off hole mass ($T \leq 100$ K)	m_{so}^*	0.154	m_e	[25]
Lattice constant ($T = 300$ K)	a	5.65	Å	[25]
Split-off energy ($T = 4.2$ K)	Δ_{so}	0.341	eV	[25]
Electron g -factor ($T = 4$ K)	g	-0.44		[25]

2.1.3 Optical selection rules

The physics quoted here is taken from [26–28]. In the current study, we use optical spin polarization which is an easy and widely used method to generate spin polarization in a direct band gap semiconductor, where the absorption of circularly polarized light results in the spin-polarized electron-hole pair. The possibility to produce a spin imbalance in the CB by exciting with a circularly polarized light is known as optical spin orientation. If the light is right or left circularly polarized (denoted by σ^+ and σ^-) they induce the same spin polarization except the sign. This method is based on energy and momentum conservation: especially the angular momentum of the photon, which leads to the distinct selection rules for the total angular momentum J and the magnetic quantum number (z component of J).

$$\Delta J = \pm 1, \quad (2.7)$$

and

$$\Delta m = \pm 1 \quad (2.8)$$

A schematic view of the allowed transition at $K = 0$ for the bulk GaAs and GaAs QWs fulfilling Eqs. 2.7 and 2.8 are illustrated by a simple term scheme in Fig. 2.4(a) and (b) respectively. For σ^+ the possible transitions are: (i) from the heavy hole band with $m = -3/2$ to the conduction band with $m = -1/2$, (ii) from the light hole band with $m = -1/2$ to the conduction band with $m = 1/2$ and (iii) from the split-off holes with $m = -1/2$ to the conduction band with $m = 1/2$. However, for photon energies, $E_g + \Delta_{so} > E_{ph} > E_g$, direct excitation from the split-off band to the conduction band will not be possible and only the lh and hh subbands will contribute. The encircled numbers represent the relative transition probabilities between the different states. Foreexample, for σ^+ , transition (i) is favored three times over transition (ii) resulting in three to one spin orientation in the CB. The degree of spin polarization (P) is described as the net

FIGURE 2.4: Term scheme for the optical selection rule at Γ -point: Allowed interband transitions for (a) bulk GaAs and (b) GaAs quantum wells. The transition probability between hh and lh are differed by 3 to 1 ratio. (Developed from [26])

spin population of the excited electron normalized to all excited electrons i.e. the ratio of the difference of carrier densities with spin-up and spin-down to the total carrier density [27]:

$$P = \frac{n_{\uparrow} - n_{\downarrow}}{n_{\uparrow} + n_{\downarrow}} \quad (2.9)$$

In the case of bulk GaAs, the heavy hole and light hole valence bands are degenerate in the vicinity of Γ -point and thus both can participate in the optical transition with different probabilities. By excluding the split-off hole band, Eq. 2.9 yields in a spin polarization of 50 % in the conduction band.

$$P = \frac{1 - 3}{1 + 3} = \frac{-1}{2} = -50\% \quad (2.10)$$

Interestingly, if there is no spin-orbit interaction then the p -type valence band will be six-fold degenerate, and hence all the bands (i.e. hh, lh and split-off hole) will contribute to the optical transition; as a result, no net spin polarization will be generated in the conduction band.

$$P = \frac{1 + 2 - 3}{1 + 2 + 3} = 0. \quad (2.11)$$

However, in the case of quantum wells, as discussed above, the degeneracy is removed, and both the hh and lh bands are further split. Thus, we can energetically select a single transition (e.g., from hh to CB) which allows us to generate 100 % spin polarization in the conduction band.

$$P = \frac{0 - 3}{0 + 3} = -1 = -100\% \quad (2.12)$$

The negative sign reflects the relationship between circularly polarized light (σ^+ or σ^-) and spin polarization.

2.2 GaAs two-dimensional electron gases and quantum wells

The two-dimensional electron gas (2DEG) in a semiconductor can be obtained at the interface of two different materials. The experimental results on spin dynamics presented in this thesis are based on the 2DEGs confined in GaAs/AlGaAs quantum well grown by molecular beam epitaxy (MBE)². The choice of these materials have several advantages like they exhibit defect free, super clean 2DEGs with highest measured mobility observed so far. To reduce the strain in the crystal lattice, materials with identical lattice spacing should be used. As both the GaAs and AlGaAs have approximately the same lattice constant (mismatch below 0.05 %) therefore, by growing GaAs/AlGaAs heterostructure results in an outstanding strain-free interface. Growing a heterostructure based on GaAs and AlGaAs would not yield a 2DEG due to the unavailability of free electrons. To introduce free electrons, a thin layer of silicon (Si) donors is added within the AlGaAs layer as a dopant³. Because of the minimum thickness achieved this process is referred to as modulation doping [29].

FIGURE 2.5: Conduction band of modulation-doped (a) GaAs/AlGaAs heterostructure and (b) GaAs quantum well. A 2DEG is formed in GaAs layer because of the band gap relationship, $E_g^{\text{AlGaAs}} > E_g^{\text{GaAs}}$. The 2DEG is shown in blue color and the positive charges on Si donor, after donating electrons, are shown in gray colors.

By bringing both materials in contact, the free electrons (donated by Si ions) from the wide-gapped AlGaAs will fall over the GaAs and become trapped in the GaAs layer. The electron's transfer at the interface leave a positive charge behind themselves which will create an attractive potential which will further bend the energy bands at the interface and will results in an approximately triangular well (See Fig. 2.5(a)). Due to a small lattice mismatch of these materials, the AlGaAs/GaAs interface is nearly free of disorder,

²Molecular beam epitaxy (MBE) is based on the epitaxial growth of crystalline layers under an ultra-high vacuum environment. The molecular beams are provided by thermal effusion cells containing an ultra-pure amount of the growth materials.

³Si acts as a donor for electrons and results in an n-doped semiconductor.

and hence the trapped electrons can move freely along the interface (xy-plane), while their motion in the direction perpendicular to the interface ($\hat{z}$ -direction) is restricted due to the small confinement potential. This restriction leads to a nearly ideal 2DEG. The striking feature, which makes GaAs/AlGaAs structure more advantageous among other semiconductors, is the extremely high purity 2DEGs with high mobility which can be accomplished by modulation doping technique, i.e., by introducing the Si doping at a certain distance from the interface separated by the spacer. The reason for placing donor ions at relatively large distance from the interface is to reduce the scattering effect because these donor ions can also act as very efficient scatters.

Additionally, the GaAs quantum well structure can be formed by growing a symmetrically doped AlGaAs/GaAs/AlGaAs substrate as shown in Fig. 2.5(b). The quantum well width (w) can be controlled by changing the layer thickness during the growth process. The number of quantum wells confined electron alter with the spacer thickness and has been reported to reduce strongly with growth of spacer thickness [30]. The number of 2DEG confined electrons is measured per unit area which is referred to as the electron density (see Sec. 3.4.2 for more details). The shape of the resulting wavefunction can be deduced by solving the Schrödinger and Poisson equations for the finite square well potential [31].

In the experimental work presented in the following chapters the samples with high purity 2DEG, grown by our collaborator A. K. Bakarov from the Novosibirsk State University are studied. All the samples have high mobility and are symmetrically doped. The growth structure of the investigated samples shall be discussed using the example of sample A (See table 3.1) as shown schematically in Fig. 2.6. First, a GaAs buffer layer was grown on GaAs substrate with (001) surface orientation. The aim of the buffer layer was to get the uniform deposition of the following layers. The buffer layer was followed by a GaAs/AlAs superlattice to block the entrance of the impurities from the substrate to the principal structure. Then the growth of a second buffer layer was carried out to provide a smooth, clean and defect-free surface for the growth of the layer of interest.

In order to shield the migration of electrons, donated by delta-doped Si donors, to buffer layer a graded layer of GaAs/Al_xGa_{1-x}As with $0 > x > 0.3$ was grown. The Al concentration was kept smaller to avoid the roughness of the growing surface (i.e. high concentration of Al destroy the smoothness of the surface). The passage of electrons from Si donor to graded layer was further restrained by growing a GaAs/Al_{0.3}Ga_{0.7}As layer known as the interior barrier of the QW. The density of Si delta-doping ($2.2 \times 10^{12} \text{ cm}^{-2}$) was symmetrically separated from the GaAs quantum well by seven periods of the short-period GaAs/AlAs superlattices (spacer layer) with 4 AlAs monolayers and 8 GaAs monolayers per period. The doping layer was grown to provide electrons to the

FIGURE 2.6: Growth scheme of sample A: Schematic layer structure of the wide GaAs/AlGaAs quantum well grown by MBE along $\hat{z} \parallel [001]$.

QW and was spatially separated from QW by short-period superlattices for the purpose to efficiently improve the mobility by shielding the doping ionized impurities [32].

After a 45 nm wide GaAs QW followed by a spacer layer (GaAs/AlAs superlattice) a layer of GaAs/Al_{0.3}Ga_{0.7}As was grown to prevent the transport of electrons from the second delta-doped Si layer to the surface, which separates the region of interest from the surface and allows further deposition of monolayer with Si doping. To saturate the dangling bonds on the surface, known as the states of the surface, a third Si doping was carried out. The delta-doping layer was covered with GaAs/Al_{0.3}Ga_{0.7}As. Finally, to prevent the structure from oxidation it was capped with a layer of GaAs. The electronic system of the studied sample A, results in a double quantum well configuration with an artificial soft barrier inside the well, due to the Coulomb repulsion of electrons in the wide quantum well (see Sec. 3.2 for details). However, in the case of triple quantum well samples investigated in the present work, potential barriers of GaAs/Al_xGa_{1-x}As were used to separate the QWs.

2.3 Larmor precession and electron g -factor

This section gives a short review of the Larmor precession frequency of the electron under application of an external magnetic field. To study the influence of external magnetic field on the electron spin dynamics, we apply an external magnetic field perpendicular to the direction of initial spin polarization (Voigt geometry). The magnetic field causes the spin to precess around it as shown schematically in Fig. 2.7 which gives a direct measure of electron g -factor. The magnetic field is applied along $\hat{y}$ -direction and the

FIGURE 2.7: *The spin polarized electrons generated by a right circularly polarized light precessing about the applied external magnetic field.*

initial spin polarization are produced in the direction of light propagation ($\hat{z}$ -direction). In general, the Hamiltonian for a free electron spin in a uniform magnetic field oriented along y -direction, $\vec{B} = B_y$ is [33],

$$H = \vec{\mu}_s \cdot \vec{B} \quad (2.13)$$

The spin $\vec{S}$ of the conduction band electron give rise to the spin magnetic moment μ_s which are related by the electron g -factor as

$$\vec{\mu}_s = \frac{g\mu_B}{\hbar} \vec{S} \quad (2.14)$$

where $\hbar$ is the reduced Planck's constant, $g = 2.00231930436170(76)$ is the Landé factor⁴ for free electron in the vacuum [34] and $\mu_B = e\hbar/2m_e$ is the Bohr magneton with elementary charge e and the electron rest mass m_e . Using Eq. 2.14 in 2.13 it simplifies to

$$H = \frac{g\mu_B}{\hbar} \vec{S} \cdot \vec{B} \quad (2.15)$$

⁴For the QW confined electrons this value is much smaller due to the modified band energy and spin-orbit energy [35]

$$H = \frac{g\mu_B}{\hbar} B_y S_z \quad (2.16)$$

where

$$\omega_L = \frac{g\mu_B B_y}{\hbar} \quad (2.17)$$

denotes the Larmor frequency with the Landé factor g . For bulk GaAs the SO interaction results in a negative electron g -factor of about $g = -0.44$ at $T = 4$ K. This value changes greatly with the sample temperature as well as on the doping concentration [36, 37]. Throughout the thesis (except temperature dependence) we use absolute value of g -factor.

2.4 Spin-Orbit Coupling

The spin-orbit coupling (SOC) is a relativistic effect [38], which have attracted lots of interest in semiconductor spintronics due to the following two reasons. (i) It provides a channel to manipulate the electron's spin degree of freedom by using an electric field rather than magnetic field. (ii) The SOI regulates the rate of spin relaxation in semiconductors. Therefore, it is a demanding issue to explore the origin of spin-orbit coupling. A key example of this effect is the SOI in the atomic systems. For better understanding, we will first investigate this effect for an electron bound to a hydrogen atom in the vacuum. Consider an electron moving through an electric field $\vec{E}$, with a velocity $\vec{v}$, experiences an effective magnetic field in its own frame of reference [39] as shown by cartoon picture in Fig. 2.8.

$$\vec{B}_{eff} = \frac{-1}{c} (\vec{v} \times \vec{E}) \quad (2.18)$$

FIGURE 2.8: (a) In the lab frame, the electron orbiting around the nucleus (b) In the reference frame of the electron, the nucleus orbiting around it creates an internal magnetic field. As a result, the electron spin degree of freedom couples to its momentum.

Using the definitions $\vec{p} = m\vec{v}$ and $e\vec{E} = -\vec{\nabla}V$ the above expression can be written as

$$\vec{B}_{eff} = \frac{1}{2mec} (\vec{p} \times \vec{\nabla}V) \quad (2.19)$$

$$\vec{B}_{eff} = \frac{e}{2mc} \frac{(\vec{p} \times \vec{r})}{r^3} \quad (2.20)$$

The factor 2 in the denominator is the Thomas factor due to the fact that the electron is in an accelerating reference frame [40]. The magnetic moment, $\vec{\mu}_s$, interacts with this magnetic field through the Zeeman interaction as

$$H_{so} = -\vec{\mu}_s \cdot \vec{B}_{eff} = \frac{-e}{2mcr^3} \vec{\mu}_s \cdot (\vec{p} \times \vec{r}) \quad (2.21)$$

$$H_{so} = \frac{e}{2mcr^3} (\vec{\mu}_s \cdot \vec{L}) \quad (2.22)$$

where $\vec{L} = \vec{r} \times \vec{p}$ is the orbital angular momentum and m the electron mass. Using the value of the magnetic moment of electron spin from Eq. 2.14 the spin-orbit Hamiltonian can be written as

$$H_{so} = \frac{eg\mu_B}{2mcr^3\hbar} (\vec{S} \cdot \vec{L}) \quad (2.23)$$

Using $\mu_B = e\hbar/2mc$ the above equation can be simplified as

$$H_{so} = \frac{g\mu_B^2}{r^3\hbar^2} (\vec{S} \cdot \vec{L}) \quad (2.24)$$

It is the most simplified form reported in the most of the quantum mechanics books. However the more general form can be obtained by rewriting 2.21 in terms of $(\vec{p} \times \vec{\nabla}V)$

$$H_{so} = \frac{1}{2mec} \vec{\mu}_s \cdot (\vec{p} \times \vec{\nabla}V) \quad (2.25)$$

$$H_{so} = \frac{g\mu_B}{2mec\hbar} \vec{S} \cdot (\vec{p} \times \vec{\nabla}V) \quad (2.26)$$

Using $\vec{S} = \vec{\sigma}\hbar/2$ and $\vec{p} = \hbar\vec{K}$ the above equation leads to

$$H_{so} = \frac{g\hbar^2}{8m^2c^2} \vec{\sigma} \cdot (\vec{K} \times \vec{\nabla}V) \quad (2.27)$$

or

$$H_{so} = \lambda_{so} \vec{\sigma} \cdot (\vec{K} \times \vec{\nabla}V) \quad (2.28)$$

where, $\lambda_{so} = g\hbar^2/8m^2c^2 \simeq \hbar^2/4m^2c^2$, is the spin-orbit coupling constant and $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ is the vector of Pauli spin matrices.

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (2.29)$$

2.4.1 Spin-Orbit Coupling from the Dirac Equation

The above discussion painted the physical origin of spin-orbit coupling. However, a more satisfactory derivation can be obtained by taking the nonrelativistic limit of the Dirac equation. The derivations follow the one presented in Ref. [41]. In the standard representation, Dirac equation is given by

$$(\vec{\alpha} \cdot c\vec{p} + \beta mc^2 + V) |\Psi\rangle = E |\Psi\rangle \quad (2.30)$$

where $\vec{\alpha} = \begin{bmatrix} 0 & \vec{\sigma} \\ \vec{\sigma} & 0 \end{bmatrix}$, $\beta = \begin{bmatrix} I & 0 \\ 0 & -I \end{bmatrix}$, V the Coulomb potential and $\Psi = \begin{bmatrix} \Psi_1 \\ \Psi_2 \end{bmatrix}$ is a four component spinor in terms of two-component vectors. Where the component Ψ_1 is a two-component vector describing a spin two particle (spin up and down states) and Ψ_2 is the component which vanishes in the non-relativistic limit. In matrix form the Dirac equation becomes

$$c\vec{p} \cdot \begin{bmatrix} 0 & \vec{\sigma} \\ \vec{\sigma} & 0 \end{bmatrix} \begin{bmatrix} \Psi_1 \\ \Psi_2 \end{bmatrix} + mc^2 \begin{bmatrix} I & 0 \\ 0 & -I \end{bmatrix} \begin{bmatrix} \Psi_1 \\ \Psi_2 \end{bmatrix} + V \begin{bmatrix} \Psi_1 \\ \Psi_2 \end{bmatrix} = E \begin{bmatrix} \Psi_1 \\ \Psi_2 \end{bmatrix} \quad (2.31)$$

where $E + mc^2$ is the total energy with E be the sum of kinetic and potential energy. Eq. 2.31 leads to two coupled equations in terms of Ψ_1 and Ψ_2 .

$$\vec{\sigma} \cdot c\vec{p} \Psi_2 = (E - mc^2 - V) \Psi_1, \quad (2.32)$$

$$\vec{\sigma} \cdot c\vec{p} \Psi_1 = (E + mc^2 - V) \Psi_2 \quad (2.33)$$

Using Eq. 2.33 we obtain

$$\Psi_2 = \left[\frac{1}{E + mc^2 - V} \right] \vec{\sigma} \cdot c\vec{p} \Psi_1 \quad (2.34)$$

Substituting this expression for Ψ_2 into the Eq. 2.32, We obtain

$$(E - mc^2 - V) \Psi_1 = \vec{\sigma} \cdot c\vec{p} \left[\frac{1}{E + mc^2 - V} \right] \vec{\sigma} \cdot c\vec{p} \Psi_1 \quad (2.35)$$

By substituting $\epsilon = E - mc^2$, where ϵ is the energy from Schrödinger's equation, the above equation takes form

$$(\epsilon - V) \Psi_1 = \vec{\sigma} \cdot c\vec{p} \left[\frac{1}{2mc^2 + \epsilon - V} \right] \vec{\sigma} \cdot c\vec{p} \Psi_1 \quad (2.36)$$

The term on the RHS can be simplified as

$$\left[\frac{1}{2mc^2 + \epsilon - V} \right] = [2mc^2 + \epsilon - V]^{-1} = (2mc^2)^{-1} \left[1 + \frac{\epsilon - V}{2mc^2} \right]^{-1} \quad (2.37)$$

$$\left[\frac{1}{2mc^2 + \epsilon - V} \right] \simeq \frac{1}{2mc^2} \left[1 - \frac{\epsilon - V}{2mc^2} \right] = \frac{1}{2mc^2} - \frac{\epsilon - V}{4m^2c^4} \quad (2.38)$$

We can then substitute this into Eq. 2.36 and obtain

$$(\epsilon - V)\Psi_1 = c\vec{\sigma} \cdot \vec{p} \left[\frac{1}{2mc^2} - \frac{\epsilon - V}{4m^2c^4} \right] c\vec{\sigma} \cdot \vec{p}\Psi_1 \quad (2.39)$$

$$\Rightarrow (\epsilon - V)\Psi_1 = \left[\frac{p^2}{2m} - \frac{\vec{\sigma} \cdot \vec{p}(\epsilon - V)\vec{\sigma} \cdot \vec{p}}{4m^2c^2} \right] \Psi_1 \quad (2.40)$$

$$\Rightarrow \epsilon\Psi_1 = \left[\frac{p^2}{2m} + V - \frac{\vec{\sigma} \cdot \vec{p}(\epsilon - V)\vec{\sigma} \cdot \vec{p}}{4m^2c^2} \right] \Psi_1 \quad (2.41)$$

Using the fact that V and $\vec{\sigma}$ commute we can get rid of ϵ on the right hand side

$$\vec{\sigma} \cdot [(\epsilon - V), \vec{p}] \Psi_1 = (\epsilon - V)\vec{\sigma} \cdot \vec{p}\Psi_1 - \vec{\sigma} \cdot \vec{p}(\epsilon - V)\Psi_1 \quad (2.42)$$

$$(\epsilon - V)\vec{\sigma} \cdot \vec{p}\Psi_1 = \vec{\sigma} \cdot \vec{p}(\epsilon - V)\Psi_1 + \vec{\sigma} \cdot [(\epsilon - V), \vec{p}] \Psi_1 \quad (2.43)$$

$$(\epsilon - V)\vec{\sigma} \cdot \vec{p}\Psi_1 = (\vec{\sigma} \cdot \vec{p}) \frac{p^2}{2m} \Psi_1 + (\vec{\sigma} \cdot [\vec{p}, V]) \Psi_1 \quad (2.44)$$

Substituting this equation in Eq. 2.41 we obtain

$$\epsilon\Psi_1 = \left[\frac{p^2}{2m} + V - \left(\frac{(\vec{\sigma} \cdot \vec{p})(\vec{\sigma} \cdot \vec{p})}{4m^2c^2} \frac{p^2}{2m} + \frac{(\vec{\sigma} \cdot \vec{p})(\vec{\sigma} \cdot [\vec{p}, V])}{4m^2c^2} \right) \right] \Psi_1 \quad (2.45)$$

Using the useful identity

$$(\vec{\sigma} \cdot \vec{X})(\vec{\sigma} \cdot \vec{Y}) = \vec{X} \cdot \vec{Y} + i\vec{\sigma} \cdot [\vec{X} \times \vec{Y}] \quad (2.46)$$

The above equation yields

$$\epsilon\Psi_1 = \left[\frac{p^2}{2m} + V - \frac{p^4}{8m^3c^2} - \frac{(\vec{\sigma} \cdot \vec{p})(\vec{\sigma} \cdot [\vec{p}, V])}{4m^2c^2} \right] \Psi_1 \quad (2.47)$$

The expression can be further simplified by using the identity given in Eq. 2.46.

$$\epsilon\Psi_1 = \left[\frac{p^2}{2m} + V - \frac{p^4}{8m^3c^2} - \frac{\vec{p} \cdot [\vec{p}, V]}{4m^2c^2} - \frac{i\vec{\sigma} \cdot (\vec{p} \times [\vec{p}, V])}{4m^2c^2} \right] \Psi_1 \quad (2.48)$$

The first two terms describe the Hamiltonian of non-relativistic Schrödinger equation. The third term is the mass correction, and the fourth term is the Darwin term. The last

term is, the one we are interested, the so-called spin-orbit interaction term. To analyze the concerned term (H_{so})

$$[\vec{p}, V] |\psi\rangle = (\vec{p}V - V\vec{p}) |\psi\rangle = \left(\iota\hbar \vec{\nabla} \frac{e^2}{r} - \frac{e^2}{r} \iota\hbar \vec{\nabla} \right) |\psi\rangle \quad (2.49)$$

$$[\vec{p}, V] |\psi\rangle = \iota\hbar e^2 \left[\left(\vec{\nabla} \left(\frac{1}{r} \right) \right) |\psi\rangle - \frac{1}{r} \vec{\nabla} |\psi\rangle \right] \quad (2.50)$$

$$[\vec{p}, V] |\psi\rangle = \iota\hbar e^2 \left[\left(\vec{\nabla} \frac{1}{r} \right) |\psi\rangle + \frac{1}{r} \vec{\nabla} |\psi\rangle - \frac{1}{r} \vec{\nabla} |\psi\rangle \right] \quad (2.51)$$

$$[\vec{p}, V] |\psi\rangle = \iota\hbar e^2 \frac{\hat{r}}{r^2} |\psi\rangle \quad (2.52)$$

Inserting in the spin-orbit term, we obtain

$$H_{so} = \frac{\iota\vec{\sigma} \cdot (\vec{p} \times [\vec{p}, V])}{4m^2c^2} = \frac{\iota\vec{\sigma} \cdot [\vec{p} \times (\iota\hbar e^2 \frac{\hat{r}}{r^2})]}{4m^2c^2} \quad (2.53)$$

$$H_{so} = \frac{\hbar}{4m^2c^2} \vec{\sigma} \cdot (\vec{p} \times \vec{\nabla} V) = \frac{\hbar^2}{4m^2c^2} \vec{\sigma} \cdot (\vec{K} \times \vec{\nabla} V) \quad (2.54)$$

$$H_{so} = \lambda_{so} \vec{\sigma} \cdot (\vec{K} \times \vec{\nabla} V) \quad (2.55)$$

Thus we got the same result as in Eq. 2.28 with $\lambda_{so} = \hbar^2/4m^2c^2$ the spin-orbit coupling constant. Two types of spin-orbit effects, the Rashba [42] and Dresselhaus [43] spin-orbit interaction are important in 2DEG systems. These interactions arise from a Bulk Inversion Asymmetries (BIA) and Structural Inversion Asymmetries (SIA).

Bulk Inversion Asymmetry

In the crystal structures with the lack of inversion center, the electron sees different atomic nuclei in the opposite directions. Fig. 2.9 shows the comparison of the atoms along a particular crystal direction in the diamond and a zinc blende crystal structure.

FIGURE 2.9: The arrangement of atoms in diamond (black) and zinc blende structure (blue and red). The inversion asymmetry arises from different atoms in zinc blende structure.

It is clearly evidenced that the zinc blende crystal structure is not symmetric under inversion. This center inversion asymmetry results in a coupling between the electron spins and its orbital motion, which further leads to the splitting of the spin states.

Structure Inversion Asymmetry

The lattice geometry and the occupancy of lattice site by different atoms result in bulk inversion asymmetry, while the structure inversion asymmetry stems from the growth process of 2D structures or by external parameters, as e.g. an electric field or strain. The SIA can be introduced from nonequivalent interfaces, asymmetric doping or an unusual shape of the quantum well as illustrated in Fig. 2.10. Additionally, the SIA can originate in the QWs based on materials with inversion symmetry by applying external electric field along the growth direction which results in the tilting of band structure and the SIA.

FIGURE 2.10: *The structures resulting in SIA (a) single side Si doping and (b) tilted structure.*

2.4.2 The Rashba spin-orbit interaction

In the semiconductor QWs, the electrons are confined along the growth direction. The asymmetry of this confining potential results in built-in electric fields along the growth direction which further results in the so-called Rashba spin-orbit coupling (SOC). In the symmetric QW structures, the Rashba spin-orbit coupling is tunable by applying the gate voltage which controls the symmetry of the well [23,44] and makes it obviously easier to manipulate the spins electrically. To understand the Rashba spin-orbit Hamiltonian consider electrons confined at the interface in a triangular well as shown in Fig. 2.11. In the rest frame of electron the electric field, arose from the potential gradient at the interface, transform into an effective magnetic field as a consequence the spin of electron

FIGURE 2.11: (a) Electrons confined at the interface in a triangular well. (b) The orientation of the Rashba spin-orbit field, due to structure inversion asymmetry, plotted in the momentum space. Fig. 2.11(b) is taken from [45].

couples to its momentum given by the Hamiltonian

$$H_{so} = \frac{\hbar^2}{4m^2c^2} \vec{\sigma} \cdot [\vec{K} \times \vec{\nabla} V] \quad (2.56)$$

$$H_R = \frac{\hbar^2}{4m^2c^2} \vec{\nabla} V_z \cdot [\vec{\sigma} \times \vec{K}] \quad (2.57)$$

$$\vec{\sigma} \times \vec{K} = \begin{vmatrix} \hat{i} & \hat{j} & \hat{k} \\ \sigma_x & \sigma_y & \sigma_z \\ k_x & k_y & k_z \end{vmatrix} = \hat{k} (\sigma_x k_y - k_x \sigma_y)$$

Inserting in Eq. 2.57 we obtain

$$H_R = \frac{\hbar^2}{4m^2c^2} \nabla V_z (\sigma_x k_y - k_x \sigma_y) \quad (2.58)$$

The above equation can also be written as

$$H_R = \alpha (\sigma_x k_y - k_x \sigma_y) \quad (2.59)$$

where $\alpha = \hbar^2/4m^2c^2 \nabla V_z$, is the Rashba spin-orbit coupling coefficient, which describes the magnitude of the interaction caused by an electric field (or asymmetric interfaces) in the confinement direction. If the electrons are moving along the $\hat{x}$ -direction, the Rashba term will cause the spin precession in the $\hat{y}$ -direction and vice versa, with rotation axis perpendicular to the velocity. The Rashba field is isotropic as shown in Fig. 2.11(b) i.e. the Rashba field is perpendicular to the direction of momentum.

2.4.3 The Dresselhaus spin-orbit interaction

In zinc blende crystal structure like GaAs, there exists a bulk inversion asymmetry. This asymmetry arises from the breaking of inversion symmetry at the midway of the bond between two nearest neighbours (i.e., Ga and As nucleus). An electron moving along the bond sees a Ga nucleus if it is travelling on one side of that point and an As nucleus if it is travelling on the opposite side. This lacking of inversion symmetry induce the SOC known as Dresselhaus SOC and is given by the Hamiltonian [43]

$$H_D = \gamma [k_x (k_y^2 - k_z^2) \sigma_x + k_y (k_z^2 - k_x^2) \sigma_y + k_z (k_x^2 - k_y^2) \sigma_z] \quad (2.60)$$

Where γ is the coupling strength, a material-specific parameter. $H_D = 0$ if the motion is along one of the axes $\hat{x}$, $\hat{y}$ and $\hat{z}$ as evidenced from $\vec{\nabla}V \times \vec{K}$ term in the Hamiltonian of the SOI. In other words, if the momentum is along or against the direction of the potential gradient there will be no spin splitting. In two-dimensional system i.e., in a QW with confinement along the z direction one can approximate $\langle k_z \rangle = 0$ and $\langle k_z^2 \rangle = (\pi/w)^2$. Where w is QW width. Then the above equation takes form

$$H_D = \gamma [k_x (k_y^2 - k_z^2) \sigma_x + k_y (k_z^2 - k_x^2) \sigma_y] \quad (2.61)$$

$$= \gamma [k_x k_y^2 \sigma_x - k_x k_z^2 \sigma_x + k_y k_z^2 \sigma_y - k_y k_x^2 \sigma_y] \quad (2.62)$$

$$= -\gamma k_z^2 [k_x \sigma_x - k_y \sigma_y] + \gamma [k_x k_y^2 \sigma_x - k_y k_x^2 \sigma_y] \quad (2.63)$$

$$= -\gamma (\pi/w)^2 (k_x \sigma_x - k_y \sigma_y) + \gamma [k_x k_y^2 \sigma_x - k_y k_x^2 \sigma_y] \quad (2.64)$$

$$= -\beta_1 (k_x \sigma_x - k_y \sigma_y) + \gamma [k_x k_y^2 \sigma_x - k_y k_x^2 \sigma_y] \quad (2.65)$$

$$= H_D^{(1)} + H_D^{(3)} \quad (2.66)$$

where, $H_D^{(1)}$ and $H_D^{(3)}$ are the Dresselhaus terms linear and cubic in k .

2.5 Metal-insulator transition

At $T = 0$ K, the intrinsic GaAs is an electrical insulator where the filled valence band is departed from the vacant conduction band by a band gap energy E_g . However, at larger dopings, the GaAs is metallic even at $T = 0$ K. The landmark that separates the insulating regime from the metallic is referred to as metal-insulator transition (MIT). Using the time-resolved Faraday rotation (TRFR) Awschalom and Kikkawa studied a series of n-GaAs samples with the different Si donors concentration. They not only found the longest spin lifetimes, exceeding 100 ns [46], but they also reported that the

FIGURE 2.12: *Spin lifetime dependence on the donor concentration in n-GaAs. The vertical dashed line marks the critical Si concentration of the metal-insulator transition. The Symbols are experimental data, and the solid lines are theoretical estimates. (Adopted from [48])*

spin lifetime changes considerably with the donor concentration yielding the maximum close to the metal-insulator transition [47].

Dzhioev and collaborator studied the spin lifetime in n-GaAs as a function of donor concentration [48] as shown in Fig. 2.12. The spin lifetime shows a complex dependence on the doping concentration. The climax was found at MIT where the spin lifetime decrease remarkably towards both sides of MIT. It has been observed that not only n-GaAs but other systems like n-GaN [49], ZnSe [50] and two-dimensional electron gases [51] also exhibit the same behaviour.

2.6 Spin lifetimes and carrier recombinations

In the previous section, metal-insulator transition has been discussed! In this section, we focus on the persistence of spin coherence exceeding the carrier recombination time ($\tau_c \approx 1$ ns [52]⁵) in the samples doped beyond metal-insulator transition. The effect that leads to long spin coherence is schematically displayed in Fig. 2.13. In the case of an

⁵In the referred experiment; the spin was electrically injected from ferromagnetic MnAs metal layers into GaAs

FIGURE 2.13: *Schematic representation of the mechanism for optical generation of electron spins in an n-doped semiconductor including hole-spin dephasing, charge carrier recombination, and electron spin relaxation. (Adopted from [53])*

n-doped semiconductor, a large number of electrons are always present in the conduction band. If a circularly polarized light is incident, the generated electrons, therefore, add up to the already existing electrons, the former being spin-polarized and the latter being unpolarized. The system can recover its equilibrium back mainly by two processes i.e. the carriers recombination and spin dephasing. Due to a strong spin-orbit coupling of the valance band in GaAs, the hole spins lose their spin polarization and relax rapidly. Soon after hole spin relaxation, the electron-hole recombination from the bottom of conduction band will start. However, as recombination takes place on a longer time scale, therefore, this process is almost negligible during hole spin relaxation. Additionally, as the holes are unpolarized, they can recombine with the unpolarized electron from 2DEG without affecting the number of spin-polarized electron in the conduction band. As a result, the spin polarization can be observed on a time scale longer than recombination time. If also, an external magnetic field is applied, this imbalance of the electron beam states will additionally perform a precession around the applied magnetic field. However, this process is also independent of the other unpolarized electrons. In this case, the pure evolution of spin polarization can also be observed. After the spin polarization completely decays, the system reaches the thermodynamics equilibrium. To start the process again, a new optical pulse will require, however, the case if the system does not attain equilibrium and the next pulse arrives, will be discussed in Sec. 3.7.

2.7 Spin relaxation mechanisms

In the previous sections, we discussed the optical injection of nonequilibrium ensemble of electron spins into the conduction band of semiconductors. The details can be seen in Sec. 2.1.3 in which the spins can be oriented by circularly polarized photons where

they transfer their angular momentum to the electrons. Now we are in a position to address the spin relaxation of these electrons due to its coupling with the orbital motion and the scattering effect. In a semiconductor nanostructure, there exist a number of mechanisms that lead to the relaxation of artificially generated spin polarization: the Elliot-Yafet, Dyakonov-Perel, Bir-Arnov-Pikus, hyperfine interaction and g -factor inhomogeneity. Which of these spin relaxation mechanism is dominant, depends on the material characteristic as well as experimental parameters, like sample temperature, magnetic field, doping concentration and crystal symmetry. For example, the Elliot-Yafet mechanism is dominant in the small band-gap semiconductors with large spin-orbit coupling while the Dyakonov-Perel mechanism is more relevant in wide band-gap semiconductors and at high temperature. Moreover, the Bir-Arnov-Pikus mechanism is efficient in highly p -doped semiconductors. The hole spin relaxation rate is typically several magnitudes higher than that of the electrons due to the stronger influence of spin-orbit coupling in the valance band compared to the conduction band [54]. More than one relaxation mechanism can be present in a material, where their interplay will lead to the thermodynamics equilibrium. In this part of the chapter, we give an overview of the main spin relaxation mechanisms that are most relevant in our structures.

2.7.1 The Elliot-Yafet mechanism

The Elliott-Yafet mechanism suggests the spin relaxation due to the spin flips at the scattering events. It was theoretically proposed by R. J. Elliot [35] in 1954, and Y. Yafet described its temperature influence [55] in 1963. It is based on the fact that in the real crystal, due to the SOI as discussed in Sec. 2.4, the electron wavefunctions are an admixture of spin up and spin down states. Because of the admixture of these opposite spin states, the wavefunctions $\Psi_\uparrow$ and $\Psi_\downarrow$ are no longer the spin eigenstates i.e. $\Psi_\downarrow$ contains some small component representing the opposite spin ($\uparrow$). In other words, it is impossible to figure out the electron wavefunction into its orbital and spin components.

If an ordinary scattering event, i.e. spin-independent interaction with impurities, boundaries, interfaces, phonons, and even with other electrons occur the amplitude of the spin up ($\uparrow$) and spin down ($\downarrow$) parts of the wavefunction can vary (due to spin flip) which leads to the relaxation of the electron spin. Thus, the EY mechanism is characterized by the spin dephasing time τ_s^{EY} , proportional to the momentum scattering time τ_p . The more the electron scattering events, the faster will be the spin relaxation. In an III-V bulk crystal, the rate has been estimated as [26, 56]

$$\frac{1}{\tau_s^{EY}} = A \left(\frac{\Delta_{so}}{E_g + \Delta_{so}} \right)^2 \frac{1}{\tau_p} \quad (2.67)$$

FIGURE 2.14: *Schematic representation of Elliot-Yafet spin relaxation mechanism: The direction of electron wave vector (wine arrows) changes with the occurrence of scattering events. At each scattering event, there exist a non-vanishing probability of spin-flip, leading to spin relaxation. (Adopted from <http://www.iue.tuwien.ac.at/phd/osintsev/dissersch4.html>)*

Here, Δ_{so} is the separation energy of the split-off band, and A is a constant number in the range 2 to 6, depending on the dominant momentum relaxation process. Due to the dependence on the band gap energy E_g , the EY mechanism is more important in metals and small band-gap semiconductors, whereas it is considered less significant in the GaAs-based semiconductor due to its wide band gap. However, in GaAs, the SO coupling is quite large for hh and lh states, compared to the conduction band states. Therefore the EY mechanism is especially more significant for the spin dephasing of the hole states.

2.7.2 The Dyakonov-Perel mechanism

Another potent spin relaxation mechanism for electrons in the GaAs-based system is the Dyakonov-Perel (or precessional) mechanism [57]. As already addressed in Sec. 2.1.2, that in systems with lacking inversion symmetry, like zinc blende structure, where the inversion asymmetry arise from the presence of two different atoms in the lattice, the spin degeneracy is entirely lifted at $\vec{K} \neq 0$. The presence of confining potential in the heterostructure or the application of an electric field can also lead to broken inversion symmetry. The spin-up and spin-down states possess different energies for the same $\vec{K}$ state, resulting in a spin splitting $\Delta E_{so} = E_{\uparrow}(\vec{K}) - E_{\downarrow}(\vec{K})$. Such energy level splitting can be described by an effective magnetic field $\vec{B}_{eff}(\vec{K})$. In quantum wells based on GaAs, this effective magnetic field is large enough and leads to an efficient dephasing because of the electron spin precession about it between the collisions. The presence of this effective magnetic field implies that, because of their different momentum, the spins of individual electrons will precess at different rate; as a result, the ensemble electron spins will dephase inhomogeneously in the momentum space.

FIGURE 2.15: Scheme of Dyakonov-Perel spin relaxation mechanism: The electron spin (shown by red arrows) precess around an effective magnetic field $\vec{B}_{eff}(\vec{K})$ (depicted by blue arrows) while traveling through a crystal with a wave vector $\vec{K}_n$ (shown by wine arrows), where n represents the number of scattering events. After each scattering event, the electron experiences a new spin-orbit effective magnetic field, which results in a change in the spin precession direction and hence the spin dephasing. (Adopted from [58])

By acknowledging the fact that if the spin precession time is greater than the momentum scattering time τ_p . The spins of individual electrons will precess about an unstable magnetic field, whose magnitude and direction varies randomly. i.e. each scattering event greatly alters the direction of electron momentum, which in turn changes the magnitude and direction of the spin-orbit effective field experienced by the electron as shown schematically in Fig. 2.15. The more momentum scattering events occur, the more random the electron spin will precess; as a result, the electron spin dephasing will be slower. The spin relaxation time τ_s^{DP} is therefore inversely related to the momentum scattering time τ_p [59, 60]

$$\frac{1}{\tau_s^{DP}} \propto \tau_p T \quad (2.68)$$

where T is the sample temperature. The increase of spin dephasing time with momentum scattering can be explained by considering the fact that with faster momentum scattering events the effective field will appear quickly and irregularly oscillating to the external field which will average to zero over time.

Few important features differentiate EY from the DP mechanism. The main difference is their opposite dependence on momentum scattering time τ_p . In EY mechanism the relaxation takes place during scattering event while in DP mechanism the relaxation takes place between the scattering events. Furthermore, as the mobility is proportional to the momentum scattering time thus for high mobility samples, we assume the DP mechanism to be more efficient while for low mobility samples the EY mechanism will be more dominant.

FIGURE 2.16: *Schematic of Bir-Aronov-Pikus spin relaxation mechanism: Collision between electrons (red) and holes (blue) leads to a spin-flip via e-h spin exchange interaction.*

2.7.3 The Bir-Aronov-Pikus mechanism

The Bir-Aronov-Pikus (BAP) mechanism, as depicted from its name, was proposed by Bir, Aronov, and Pikus in 1975 [61]. It is an efficient mechanism in the p -doped semiconductors at low temperatures, which arises from the exchange interaction between electron and hole spins while the total spin is preserved. In the presence of optically generated holes, the exchange interaction leads to a spin-flip where the spin of an electron in conduction band exchange with the spin of the hole resulting in the dephasing of electron spin (Fig. 2.16). It can be explained in a similar fashion as the Dyakonov-Perel relaxation mechanism, with electron spins precessing about an effective magnetic field generated by the hole spin [62]. The spin relaxation rate $1/\tau_s^{BAP}$ of the BAP mechanism is proportional to the hole concentration p [63]

$$\frac{1}{\tau_s^{BAP}} \propto p \quad (2.69)$$

In n -doped semiconductors, the BAP mechanism is typically less efficient due to the lack of holes. In the present work, all the measurements are conducted on n -doped samples so that the BAP mechanism can affect electron spin dephasing only due to optically generated holes.

2.7.4 The g -factor inhomogeneity

The presence of an inhomogeneously broadened g -factor for an ensemble of electron spins in an external magnetic field applied perpendicular to the initial spin orientation (Voigt geometry) leads to a distribution of the spin precession frequencies $\Delta\omega$. Which further results in an inhomogeneous dephasing of the electron spin as

$$\frac{1}{\tau_s} = \frac{\Delta g \mu_B B_{ext}}{2\hbar} \quad (2.70)$$

This dephasing rate $1/\tau_s$ is linear in B_{ext} .

Chapter 3

Samples and measuring techniques

The thesis at hand investigates carrier spin dynamics in various GaAs/AlGaAs multilayer systems. Before discussing our experimental results, it is fruitful to introduce major experimental techniques which had been employed in our experiments for the determination of carrier spin dynamics. Therefore, in this chapter, we discuss how to manipulate and detect the spin polarization. The samples were immersed in a split-coil superconductor magnet cryostat in the Kerr configuration which provides high magnetic fields of about up to 8 T. The samples could be exposed either to a bath of liquid helium or helium gas at $T = 1.2$ K. The sections are organized as follows: The investigated samples (Sec. 3.2), Sheet density and mobility calculation (Sec. 3.4), Magneto-optical Kerr effect (Sec. 3.5), the physics of the time-resolved Kerr rotation (Sec. 3.6), Resonant spin amplification (Sec. 3.7) and current-induced spin polarization (Sec. 3.8). The chapter concludes with an overview of the optical set-up to perform these measurements.

3.1 Sample Growth and Fabrication

The samples under investigation are all GaAs/AlGaAs heterostructures. Heterostructures are semiconductor structures that contains layers of different materials grown on the top of a thicker substrate. There are a number of different technologies to grow semiconductor heterostructures. Some of the possible techniques are liquid phase epitaxy (LPE), vapor phase epitaxy (VPE), metal organic chemical vapor deposition (MOCVD) or metal organic vapor phase epitaxy (MOVPE). However, using these techniques, it is not possible to control the thickness of material depositions with atomic monolayer precision. To control the concentration of a dopant the molecular beam epitaxy (MBE)

FIGURE 3.1: Schematic of an MBE system with heatable sample holder, effusion cells, and shutters in a vacuum chamber. (Adopted from <http://www.explainthatstuff.com/molecular-beam-epitaxy-introduction.html>)

is used, which is the most sophisticated method. MBE, as its name indicates, uses vapor beams of atoms or molecules in an ultra-high vacuum (basic pressure 10^{-13} bar) environment to provide a source of the constituents to the growing surface of a substrate crystal. This sophisticated growth technique invented in the late 1960s at the Bell Telephone Laboratory by A. Y. Cho and J. R. Arthur [64, 65], allows growing heterostructures of high crystalline quality with very sharp boundaries between different materials grown on top of each other. Further advantage of this technique is the absence of carrier gases as well as the ultra-high vacuum environment which leads to a high purity of the grown films.

A schematic of MBE system is displayed in Fig. 3.1 which consists of a sample holder mounted in the vacuum chamber that can be heated. The epitaxial growth of the heterostructure takes place when a molecular beam hits the heated substrate. The molecular beams are provided by a number of thermal effusion cells containing ultra-pure ($>99.99999\%$) amounts of the growth materials. They can be heated to a temperature where the material starts to evaporate and delivered to the substrate with a growth rate between 0.001 monolayer/second and two monolayer/second. Typically, each element is delivered in a separately controlled beam, so that the stoichiometry can be set for any given layer, thereby defining the precise composition and electrical and optical characteristics of that layer. A computer control shutters in front of each effusion cell are used to immediately turn on and off the molecular beam to precisely control the composition of the structure being deposited. Additionally, MBE offers tremendous control over the layer thickness of the epitaxially grown layers by rotating the sample

holder. The growth rate of this system is around one monolayer per second because the atoms at the surface need some time to arrange themselves in monolayers.

3.2 Samples under investigation

This section gives an overview of the QW structures investigated in this thesis by pump-probe Kerr rotation spectroscopy. All the samples measured are intentionally doped barely on the metallic side of the metal-insulator transition (See sec. 2.5). This doping level was chosen as a compromise between the need for a conductive channel and the desire for achieving long spin lifetime, which is largest for the donor concentration close to the metal-insulator transition [48, 51]. The experimental results presented in Chapters. 4, 5, 6, 7 and 8 require a precise control of sample parameters, such as structural symmetry, quantum well width, electron density and the aluminium content in the barriers. To meet this demand, the investigated samples were grown by our collaborator Prof. A. K. Bakarov from the Novosibirsk state University, Russia using molecular beam epitaxy (MBE). In the present work, the study of spin dynamics was carried out in seven different quantum wells samples grown along $z \parallel [001]$, containing a dense two-dimensional electron gas. For all the samples the barriers are short-period AlAs/GaAs superlattices (SPSL) to enhance the mobility by shielding the doping ionized impurities [32]. The density of Si delta-doping was $2.2 \times 10^{12} \text{ cm}^{-2}$ symmetrically separated from the QW by seven periods of the SPSL with 4 AlAs monolayers and 8 GaAs monolayers per period.

Sample A (RC447)

Sample A is a 45-nm-wide GaAs quantum well containing a 2DEG. Due to the Coulomb repulsion of the electrons in the wide quantum well that is doped in both barriers, the electronic system results in a double quantum well (DQW) configuration with a soft barrier inside the well. Electrical transport measurements determine a total electron density $n_s = 9.2 \times 10^{11} \text{ cm}^{-2}$, and mobility $\mu = 1.9 \times 10^6 \text{ cm}^2/Vs$ at low temperature.

Sample B (#480)

Sample B is GaAs triple quantum well (TQW) with 2-nm-thick $Al_{0.3}Ga_{0.7}As$ barriers, $n_s = 9 \times 10^{11} \text{ cm}^{-2}$, and $\mu = 5 \times 10^5 \text{ cm}^2/Vs$ measured at low temperature. The central well width is 22 nm, and both side wells have an equal width of 10 nm. The central well has a larger width to be populated because the electron density tends to concentrate

FIGURE 3.2: Scheme of a Hall-bar for magnetotransport measurements demonstrating the Longitudinal and transverse contacts.

mostly in the side wells as a result of electron repulsion and confinement. The estimated density in the central well is 35% less than that in the side wells.

Sample C (#299)

Sample C is a symmetrically δ -doped GaAs/AlGaAs TQW with total density $n_s = 9.6 \times 10^{11} \text{ cm}^{-2}$ and low temperature mobility $\mu = 5.5 \times 10^5 \text{ cm}^2/\text{Vs}$. It contains a 26-nm-thick GaAs central well with electron density of about $1.4 \times 10^{11} \text{ cm}^{-2}$ and two 12-nm-thick lateral wells with approximately equal electron density of $4.1 \times 10^{11} \text{ cm}^{-2}$, each separated by 1.4-nm-thick $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barriers. For the electrical generation of spin polarization the sample was patterned into a six-contact Hall bar geometry. The main channel have length $L = 500 \mu\text{m}$ between the side probes (in the y-axis), width $w = 200 \mu\text{m}$ and four transverse channels of equal width $15 \mu\text{m}$ that extend out from the main channel (in the z-axis) as shown schematically in Fig. 3.2. The studied samples are summarized in table 3.1.

TABLE 3.1: Studied samples where n_s is the electron density, and μ is the mobility.

Name	Structure	QW width (nm)	$\delta - \text{Si}(\text{cm}^{-2})$	$n_s(\text{cm}^{-2})$	$\mu(\text{cm}^2/\text{Vs})$
Sample A	DQW	45	2.35×10^{12}	9.2×10^{11}	1.9×10^6
Sample B	TQW	10-22-10	2.2×10^{12}	9.0×10^{11}	5.0×10^5
Sample C	TQW	12-26-12	2.2×10^{12}	9.6×10^{11}	5.5×10^5

3.3 Samples with Al content in each well

Additionally, we studied the spin dynamics in four different samples one single and three double quantum wells. All the samples are remotely delta-doped beyond the metal-insulator transition. The single quantum well (SQW) sample is named sample D,

and the other three DQW samples are named samples E, F, and G respectively. All the samples have an equal well width with different Al concentration in each well.

Sample D (#3252)

Sample D is a 14-nm-wide AlGaAs single quantum well with total electron density $n_s = 4.79 \times 10^{11} \text{ cm}^{-2}$ and mobility $\mu = 35.8 \times 10^3 \text{ cm}^2/Vs$. The quantum well is grown with $x = 10 \%$ Al content inside the well.

Sample E (#3255)

Sample E is a DQW with 1.4-nm-thick AlAs barrier. Both quantum wells have an equal width of 14-nm with $x = 10 \%$ Al contents in each well. Sample E have total electron density $n_s = 4.76 \times 10^{11} \text{ cm}^{-2}$ and mobility $\mu = 12 \times 10^3 \text{ cm}^2/Vs$.

Sample F (#3242)

Sample F is a DQW with total electron density $n_s = 6.63 \times 10^{11} \text{ cm}^{-2}$ and mobility $\mu = 37.9 \times 10^3 \text{ cm}^2/Vs$. Sample F has the same structure of sample E with a barrier thickness (5-nm) of about four times thicker than sample E. The well close to the substrate have $x = 8.2 \%$ Al concentration and the second well have $x = 14.2 \%$ Al concentration.

Sample G (#3231)

Finally, the sample G has grown with Al concentration of $x = 11 \%$ and 16% in QW1 and QW2 respectively. The both quantum wells are separated by a 5-nm-thick AlAs central barrier. Sample G have mobility $\mu = 39 \times 10^3 \text{ cm}^2/Vs$ and total electron density $n_s = 5 \times 10^{11} \text{ cm}^{-2}$. The characteristics of the samples with Al concentration in each individual well are summarized in table 3.2.

TABLE 3.2: Studied single and double quantum wells with Al concentration in each well where, n_s is the electron density, and μ is the mobility. All the wells have an equal width of 14 nm. The density of Si delta doping was $2.2 \times 10^{12} \text{ cm}^{-2}$ for all the samples.

Name	Structure	$x\%$ QW1	$x\%$ QW2	$n_s(\text{cm}^{-2})$	$\mu(\text{cm}^2/Vs)$
Sample D	SQW	10	-	4.79×10^{11}	35.8×10^3
Sample E	DQW	10	10	4.76×10^{11}	12×10^3
Sample F	DQW	8.2	14.2	7.39×10^{11}	37.9×10^3
Sample G	DQW	11	16	5.0×10^{11}	39×10^5

3.4 Sheet Density and Mobility

The determination of carrier density and mobility of the sample can be accomplished by electrical transport measurements which require the sample to have Hall bar geometry (See Fig. 3.2). Electrical transport is the process which describes the motion of carrier's in the semiconductor when bias voltages are applied to it. The drift and diffusion of the carriers are the two most important transport processes. The drift is the carrier's motion under the influence of applied electric field while diffusion occurs even though there's not an electric field applied to the semiconductors. Diffusion is mainly the flow of carriers due to the density variation. To define the limits of classical and quantum mechanical transport regimes, it is important to scale the size of the structure with de Broglie wavelength λ as given by:

$$\lambda = \frac{h}{p} = \frac{h}{\sqrt{2m^*E}} = \lambda_0 \sqrt{\frac{m}{m^*}} \quad (3.1)$$

where h is the Planck's constant, $\vec{p}$ is the carrier momentum, E is the kinetic energy, m^* is the effective mass of the carrier in semiconductor and λ_0 is the de Broglie wavelength of a free electron. The de Broglie wavelength of an electron in GaAs at room temperature is 295 Å [66]. At cryogenic temperatures, such as 4.2 K, the de Broglie wavelength increases up to a fraction of a micron which is comparable with the structure size in the nanoscale limit. Therefore, the quantum mechanical aspects of transport properties become more valuable in microscopic structures. However, due to scattering effects caused by the impurities (including dopants) and imperfections of the host crystal the electrons in semiconductor lose their wavelike behavior and can be treated classically.

3.4.1 Classical Transport

In the Classical approach, we treat the electrons as a classical particle and apply the Drude model to describe the carrier transport. In equilibrium, the rate at which the electrons receive momentum from an applied electric field $\vec{E}$ is exactly equal to the rate at which they lose momentum due to scattering forces [67]

$$\left[\frac{d\vec{p}}{dt} \right]_{field} = \left[\frac{d\vec{p}}{dt} \right]_{scattering} \quad (3.2)$$

In Drude model, it is assumed that the momentum of an electron changes due to strong scattering events, and hence the direction of an electron after scattering events does not match with its initial direction. Introducing the momentum relaxation time τ_m i.e. the

average duration between two collisions the above equation reads as:

$$e\vec{E} = \frac{m^*\vec{v}_d}{\tau_m} \quad (3.3)$$

$$\vec{v}_d = \mu\vec{E} \quad (3.4)$$

where $\mu = e\tau_m/m^*$ is the mobility. In the Drude picture, the drifting electrons due applied electric field results in current, the current density $\vec{j}$ is related to the electron density n_s by the following relation

$$\vec{j} = -en_s\vec{v}_d = -en_s\mu\vec{E} = \vec{\sigma}\vec{E} \quad (3.5)$$

where $\vec{\sigma} = en_s\mu$ is the conductivity tensor. An additional quantity describing the electronic system should be introduced, the mean free path ℓ . Between two scattering events, the electron moves with the Fermi velocity $\vec{v}_F$. Thus, the mean free path between two scattering events can be easily deduced from the Fermi velocity as:

$$\ell = \tau_m v_F = \sqrt{2\pi n_s} \frac{\hbar\mu}{e} \quad (3.6)$$

The Drude model is only valid for low magnetic fields and overlapping Landau levels (LLs) ($\omega_c\tau_m \ll 1$), where Landau quantization is neglected. At higher magnetic fields the classical model fails and we have to use quantum mechanics to describe our 2DEG properly. That is at high enough magnetic field an electron can turn many cyclotron orbits before it is scattered and thus there will be no transport due to drifting charge carriers. If there is a current flowing through the central channel (1-2) of the sample (see Fig. 3.2), a small magnetic field $\vec{B} = (0, 0, B)$ perpendicular to the structure will deflect the electrons due to the Lorentz force. As a consequence, an electric field in the x-direction will be set up, compensating the deflection. This situation could be described by Newton's equation of motion [68] as:

$$m^* \frac{d\vec{v}_d}{dt} = -e \left(\vec{E} + \vec{v}_d \times \vec{B} \right) - m^* \frac{\vec{v}_d}{\tau_m} \quad (3.7)$$

where $m^*\vec{v}_d/\tau_m$ is a damping force on the electron due to scattering. Using $\vec{v}_d \times \vec{B} = v_y B \hat{i} - v_x B \hat{j}$, the above equation reads as

$$m^* \frac{dv_x}{dt} = -eE_x - ev_y B - m^* \frac{v_x}{\tau_m} \quad (3.8)$$

$$m^* \frac{dv_y}{dt} = -eE_y + ev_x B - m^* \frac{v_y}{\tau_m} \quad (3.9)$$

where v_x and v_y are the x and y components of the drift velocity. In steady state, $d\vec{p}/dt = 0$ i.e. $d\vec{v}_d/dt = 0$, so that the set of equations for the velocity of electron is

given as:

$$v_x = \frac{-\mu}{(1 + \omega_c^2 \tau_m^2)} [E_x - \omega_c \tau_m E_y] \quad (3.10)$$

$$v_y = \frac{-\mu}{(1 + \omega_c^2 \tau_m^2)} [E_y + \omega_c \tau_m E_x] \quad (3.11)$$

where $\omega_c = eB/m^*$ is cyclotron frequency and $\mu = e\tau_m/m^*$ is the mobility. Using eq. 3.5 we can rewrite the above expressions as:

$$j_x = \frac{en_s \mu}{(1 + \omega_c^2 \tau_m^2)} [E_x - \omega_c \tau_m E_y] = \frac{\sigma_o}{(1 + \omega_c^2 \tau_m^2)} [E_x - \omega_c \tau_m E_y] \quad (3.12)$$

$$j_y = \frac{en_s \mu}{(1 + \omega_c^2 \tau_m^2)} [E_y + \omega_c \tau_m E_x] = \frac{\sigma_o}{(1 + \omega_c^2 \tau_m^2)} [E_y + \omega_c \tau_m E_x] \quad (3.13)$$

where σ_o is the Drude conductivity in the absence of an applied magnetic field. Writing these expressions in matrix form, we obtain the expression for conductivity tensor.

$$\begin{pmatrix} j_x \\ j_y \end{pmatrix} = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{yx} & \sigma_{yy} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \end{pmatrix} \quad (3.14)$$

where E_x and E_y are the x and y components of applied electric field. Rewriting eqs. 3.12 and 3.13 in matrix form as:

$$\begin{pmatrix} j_x \\ j_y \end{pmatrix} = \frac{\sigma_o}{1 + \omega_c^2 \tau_m^2} \begin{pmatrix} 1 & -\omega_c \tau_m \\ \omega_c \tau_m & 1 \end{pmatrix} \begin{pmatrix} E_x \\ E_y \end{pmatrix} \quad (3.15)$$

where

$$\sigma = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{yx} & \sigma_{yy} \end{pmatrix} = \frac{\sigma_o}{1 + \omega_c^2 \tau_m^2} \begin{pmatrix} 1 & -\omega_c \tau_m \\ \omega_c \tau_m & 1 \end{pmatrix} \quad (3.16)$$

In isotropic systems, the components of the conductivity tensor are symmetric: $\sigma_{xx} = \sigma_{yy}$ and $\sigma_{xy} = -\sigma_{yx}$. The resistivity tensor ρ is obtained by a simple tensor inversion of conductivity σ .

$$\begin{pmatrix} \rho_{xx} & \rho_{xy} \\ -\rho_{xy} & \rho_{xx} \end{pmatrix} = \frac{1}{\sigma_{xx}^2 + \sigma_{xy}^2} \begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ -\sigma_{xy} & \sigma_{xx} \end{pmatrix} \quad (3.17)$$

The resistivity components ρ_{xx} and ρ_{xy} can be written as

$$\rho_{xx} = \frac{\sigma_{xx}}{\sigma_{xx}^2 + \sigma_{xy}^2} = \frac{1}{en_s \mu} \quad (3.18)$$

$$\rho_{xy} = \frac{\sigma_{xy}}{\sigma_{xx}^2 + \sigma_{xy}^2} = \frac{B}{en_s} \quad (3.19)$$

Hence for the Hall bar geometry of length L and width w the resistivities ρ_{xx} and ρ_{xy} are related to the longitudinal and transverse resistances by:

$$\rho_{xx} = R_{xx} \frac{w}{L} \quad (3.20)$$

$$\rho_{xy} = R_{xy} \quad (3.21)$$

where $\frac{w}{L} = 200/500$ for the Hall bar geometry in this work. The Drude model predicts that the longitudinal resistivity (resistance) is independent of the magnetic field while the Hall resistance increases linearly with the magnetic field. The carrier density n_s and the mobility μ can be extracted from the measured low-field resistivities using Eqs. 3.18 and 3.19:

$$\mu = \frac{1}{en_s \rho_{xx}} = \frac{L}{en_s R_{xx} w} \quad (3.22)$$

$$n_s = \frac{B}{e R_{xy}} \quad (3.23)$$

Therefore, one can first find the density using Eq. 3.23 by measuring the Hall resistance, and then restore the value of the mobility from Eq. 3.22. In the limit of a clean system, $\rho_{xx} = 0$ and $R_{xx} = 0$ and thus, $\mu \rightarrow \infty$.

3.4.2 Quantum Mechanical Transport

In the limit of small magnetic fields ($\omega_c \ll \tau_m^{-1}$), the resistivity tensor designate that the longitudinal resistance R_{xx} remains constant while the Hall resistance R_{xy} increases linearly with magnetic field (see Eqs. 3.18 and 3.19). In 1930, L. Schubnikov and W. J. de Haas reported [69] that above some characteristic magnetic field the longitudinal resistance no longer remains constant but oscillate as a function magnetic field. These oscillations are called Shubnikov-de-Haas (SdH) oscillations as shown in Fig. 3.3 for the studied sample C observed at 1.2 K. Additionally, the Hall resistance also deviates from linear behavior and shows a series of plateaus whenever R_{xx} goes through a minimum. Such oscillations were first observed in the bulk metals in 1930; however, the effect is much larger in 2D semiconductors. These features are usually absent at high temperatures but more pronounced at cryogenic temperatures of 4 K and below. For better understanding of these features, we need to go beyond the Drude model and discuss the formation of LLs and Landau quantization (See Appendix A.1 for details) which are a quantum mechanical effects.

The SdH oscillations, originate from the oscillations of the density of states with the filling factor $\nu = h/eBn_s$. For a given n_s , the change in magnetic field results in the variation of filling factor which is equivalent to the motion of Fermi level across LLs.

FIGURE 3.3: Measured longitudinal resistance R_{xx} and transverse resistance R_{xy} for TQW (sample C) as a function of applied magnetic field B at $T = 1.2$ K. SdH oscillations in R_{xx} and Hall plateaus in R_{xy} observed at fields greater 0.4 T.

The longitudinal resistance has a maximum whenever a LL crosses the Fermi surface, and it becomes zero if the Fermi energy is exactly in between two LLs. SdH oscillations can be used to extract some important parameters like a quantum lifetime, effective mass, g -factor and carrier density, etc. In the present section, we will focus on the calculation of the carrier density from the period of oscillations. The SdH oscillations can be described by Dingle formula [70]

$$\rho_{xx}(B) = \rho_{xx}(0) + \left[\cos \left(\frac{2\pi E_F}{\hbar\omega_c} \right) E(m^*, \tau_q) D(m^*, T) \right] \quad (3.24)$$

where τ_q is the quantum lifetime, and the term $D(m^*, T)$ describes the temperature and effective mass dependence of the oscillation

$$D(m^*, T) = \frac{\xi}{\sinh \xi}, \quad \xi = \frac{2\pi^2 K_B T}{\hbar\omega_c} \quad (3.25)$$

An increase in the oscillation amplitude with the magnetic field is predicted by the exponential term (known as Dingle-term)

$$E(m^*, \tau_q) = \exp \left(\frac{-\pi}{\omega_c \tau_q} \right) \quad (3.26)$$

Using the fact that the interval between two consecutive oscillation is 2π , we can extract the carrier's density from the argument of cosine term ($2\pi E_F / \hbar\omega_c = [2\pi E_F m^* / \hbar e] \frac{1}{B}$)

in the Eq. 3.24.

$$\frac{2\pi E_F m^*}{\hbar e} \left(\frac{1}{B_1} - \frac{1}{B_2} \right) = 2\pi \quad (3.27)$$

Using $n_s = m^* E_F / \pi \hbar^2$ in the above equation we get

$$n_s = \frac{2e}{h \Delta(\frac{1}{B})} \quad (3.28)$$

where $\Delta(\frac{1}{B}) = \left[\frac{1}{B_1} - \frac{1}{B_2} \right]$ is the period of oscillation and 1 and 2 are the numbers of two subsequent SdH minima. The Eq. 3.28 can also be written as

$$f = \frac{\hbar n_s}{2e} \quad (3.29)$$

where f is the frequency of oscillation. The usual procedure to extract the carrier density from the magnetotransport data shown in Fig. 3.3 is to determine the frequency of oscillation in R_{xx} in the inverse magnetic field. For that purpose, one has to plot the longitudinal resistance as a function of $1/B$. After plotting the data against $1/B$, a fast Fourier transform (FFT) is carried out. The FFT results in a distinct peak at a certain frequency from which the carrier's density could be determined by equation 3.29. See sec. 8.10 for more details.

3.5 The Faraday/Kerr effect

Faraday and Kerr's effects are basically the same physical phenomena: the rotation of the linear polarization of a light beam after transmission through (Faraday) or reflection from (Kerr) the material. In 1845, Michael Faraday discovered that, if a linearly polarized light is passed through a piece of glass or other transparent material it's polarization rotates by an angle Θ_F [71]. This rotation of polarization upon transmission is proportional to the external field applied along the propagation direction of the light:

$$\Theta_F = \nu_v B L \quad (3.30)$$

In this expression, ν_v is the Verdet constant (which depends on wavelength and temperature) that represents the strength of the Faraday effect for a particular material, L is the length of the optical path where the light and magnetic field interact, and B is an external magnetic field. Faraday effect was one of the first experimental evidence demonstrating that light and electromagnetism are related. The original experiments by Michael Faraday and John Kerr were conducted in the 1840s and 1870s respectively but got lots of attention with the development of ultrafast Ti: Sapphire lasers, which made it possible to study the spin polarization dynamics on very short timescales. A similar

effect can occur due to magnetization which is present in a material itself, causing the rotation of the linear polarization of a beam of light upon reflection (Kerr effect) or upon transmission (Faraday effect) [72].

In the scientific community, the Kerr and Faraday rotations are the two widely used effects. The underlying mechanism behind these effects is the same, and depending on the sample structure one of them will be preferable. The experimental work presented in the following chapters used the time-resolved Kerr effect to study the electron spin dynamics in GaAs-based materials. This effect can also be explained in the context of a spin-polarized conduction band in GaAs. Because of the optical selection rules (as described in Sec. 2.1.3), the states with spin up polarization are filled with a higher energy than the states with spin down polarization. This unequal population of up and down spins in the conduction band results in a shift in the absorption curves for the right (σ^+) and left (σ^-) circularly polarized light (see Fig. 3.4(a) known as circular birefringence. According to Kramers-Kronig relations [73], this difference in absorption translates to a difference in refractive indices of σ^+ and σ^- circular polarizations, as

FIGURE 3.4: Model for Kerr/Faraday effect. (a) shift in the absorption edge for σ^+ and σ^- light. (b) Associated indices of refraction as a function of photon energy. The black curve represents the index of refraction of linearly polarized light which is proportional to the Kerr rotation angle. (Adopted from [53])

depicted in Fig. 3.4(b). A difference in the index of σ^+ and σ^- will cause a phase shift between σ^+ and σ^- light passing through a material. The linearly polarized light is composed of σ^+ and σ^- light, whose phase shift results in the rotation of its polarization axis by an angle Θ_K upon reflection on an interface of the sample. Therefore, in the case of magnetized materials, the Θ_K is proportional to the component of the magnetization along the direction of propagation.

3.6 Time-resolved Kerr rotation

The main investigation technique used in this work, for the determination of transverse spin relaxation times and Landé g -factor, is the so-called time-resolved Kerr rotation (TRKR). It is a widely used technique that allows spin dynamics to be investigated on the timescales from few femtoseconds up to several nanoseconds [74]. The circularly-polarized pump pulses provided by mode-locked Ti: sapphire laser are used to generate electron spin ensemble along a certain direction in the sample. Based on the effect described in Sec. 3.5, the evolution of these optically generated spin ensemble can be monitored via rotation of polarization plane of a linearly polarized probe pulse reflected by the sample. This is accomplished with the help of a mechanical delay line that varies the optical path length of one beam relative to the other. An external magnetic field B is applied perpendicular to both the pump and the probe laser beam direction (Voigt geometry) as shown in Fig. 3.5. The external magnetic field force the spin precess around it. The amount of polarization rotation (Θ_K) of the probe beam upon the reflection on the sample is a direct measure of the amount of spin orientation at that moment. This small rotation of the direction of linear polarization can be detected using sensitive electronics as described in section 3.10. A typical oscillatory response of such a TRKR experiment in the presence of an applied external magnetic field is shown in Fig. 3.6. Due to the homogeneous and inhomogeneous spin dephasing mechanism described in chapter 1, the spin polarization decay with an exponential envelope. The frequency of oscillation is a direct measure of electron g -factor, $g = \hbar\omega_L/\mu_B B$. The combination of both, spin dephasing and spin precession, leads to an exponentially decaying cosine function of the Kerr rotation described by:

$$\Theta_K = A \exp\left(\frac{-\Delta t}{T_2^*}\right) \cos\left(\frac{|g|\mu_B B}{\hbar} \Delta t + \varphi\right) \quad (3.31)$$

Where A is the initial amplitude, μ_B is the Bohr magneton, B is the external magnetic field, $\hbar$ is the reduced Planck constant, $|g|$ is the electron g factor, φ is the initial phase and T_2^* is the ensemble dephasing time. The cosine factor reflects spin precession about the external magnetic field. This technique is suitable for the observation of spin

FIGURE 3.5: Schematic for the Time-resolved pump-probe setup: Circularly polarized pump and linearly polarized probe are focused approximately normal to the sample. Pump pulse generates coherent spin ensemble. The transverse magnetic field is applied to the sample (Voigt geometry) which cause the precession of the spin ensemble. The probe pulses, delayed by the time Δt , detect the net magnetization by means of the Kerr rotation angle Θ_K .

dynamics on the time scale within the laser's pulse repetition interval ~ 13.2 ns. The ensemble spin dephasing times on longer timescales, exceeding pulse repetition period can be obtained by using a slightly modified form of the detection scheme (see Sec. 3.7), with similar experimental and data acquisition setup.

For the analysis of the Kerr effect, it is important to distinguish different geometries, which depend on the relative orientation of the incident light and the magnetization

FIGURE 3.6: Schematic curve of time-resolved Kerr rotation with Kerr rotation angle Θ_K (representing electron spin orientation) as a function of pump-probe delay Δt . The envelope is caused by spin relaxation, whereas the oscillatory character is due to the spin precession about the applied magnetic field.

FIGURE 3.7: The three fundamental Kerr geometries that are considered for describing the magneto-optical Kerr effect. (a) Polar (b) Longitudinal and (c) Transverse. The general Kerr effect for an arbitrary direction of magnetization is the superposition of these three basic effects. The superscripts i and r indicate incident and reflected rays. (adapted from [79]).

$\vec{M}$ in the sample. Because of a large number of parameters, a complete mathematical description will be omitted here as it exceeds the scope of this work by far; however, the three fundamentally differentiated geometries are presented (See Fig. 3.7). The interested readers are referred to [75–78] for the complete theoretical description and calculation. In the case of polar Kerr effect (see Fig 3.7(a)) the magnetization lies perpendicular to the surface of the sample. Fig 3.7(b) shows the case in which the magnetization vector lies both in the sample plane and in the plane of incidence of the light known as longitudinal Kerr effect. Where the transverse Kerr effect concerns to the case where the magnetization lies in the sample plane but is perpendicular to the plane of incidence of the light (see Fig 3.7(c)).

FIGURE 3.8: The polar, longitudinal and transverse Kerr effects dependence on the angle of incidence in permalloy. [80].

All the three geometries are involved in the optical orientation of spin's due normal incidence of circularly polarized light. Upon injection, the main effect is of polar nature and the magnetization produced are normal to the sample surface. Due to the spin precession caused by an applied external magnetic field, the effect translates from polar to transverse nature. In the presence of longitudinal spin relaxation the spin's get align in the direction of magnetic field resulting in the longitudinal Kerr effect.

For the detection angle close to the normal incidence the above-said effects can be explained regarding $\vec{M} \cdot \vec{K}_i$. The magnitude of reflection coefficients of a p-wave going into an s-wave (r_{ps} for polar and longitudinal) and the change of reflection coefficient of the p-wave (Δr for transverse) are depicted in Figure 3.8 showing the domination of polar effect at the angle of detection close to the normal incidence. Where the subscripts p and s indicate polarization parallel and perpendicular to the plane of incidence respectively.

3.7 Resonant spin amplification

In the standard TRKR technique, as described above, a circularly polarized light emitted from the pulsed laser system create the spin-polarized electrons where a weak linearly polarized beam from the same laser is used to detect the spin dynamics of those electrons. Using this technique, the spin dephasing can be observed in a time interval shorter than the pulse repetition which is 13.2 ns in our case. A problem arises when the spin dephasing time becomes equal or greater than the repetition period (t_{rep}) of the pulsed laser. In such cases, a new pump pulse excites spins before the previous ensemble has completely dephased. This is the case schematically shown in Fig. 3.9, as evidenced by the Kerr rotation signal at negative delays¹.

In this regime, the direct determination of spin dephasing time by TRKR method becomes inapplicable. Thus, for measuring long-lived spin dynamics the resonant spin amplification (RSA) technique [46], based on the interference of spin polarization's created by subsequent pulses, can be used. In this technique, we must take into account the contribution to the Kerr rotation from all previous pulses. The resulting sum over all precessions of the spin polarization created by previous pulses becomes:

$$\Theta_K(B, \Delta t) = \sum_{n=1}^{\infty} A \exp\left(-\frac{(\Delta t + nt_{rep})}{T_2^*}\right) \cos\left(\frac{g\mu_B(\Delta t + nt_{rep})B_{ext}}{\hbar}\right) \quad (3.32)$$

¹Here negative delay indicates that the probe pulse arrives before the pump pulse

FIGURE 3.9: Schematic of the TRKR demonstrating the spin dephasing time shorter/longer than the laser repetition period. (Taken from the <http://www.ioffe.ru/spin2012/present/We-4s.pdf>)

The above formula can be written in a more compact form without the infinite sum

$$\Theta_K(B, \Delta t) = \frac{A}{2} \exp\left(-\frac{(\Delta t + t_{rep})}{T_2^*}\right) \left[\frac{\cos(\omega_L \Delta t) - \exp(t_{rep}/T_2^*) \cos[\omega_L (\Delta t + t_{rep})]}{\cos(\omega_L t_{rep}) - \cosh(t_{rep}/T_2^*)} \right] \quad (3.33)$$

with $\Delta t \in [0; t_{rep})$. For derivation of above formula see Appendix A.2.

Usually, the repetition rate is fixed, while the Larmor precession frequency varies with

FIGURE 3.10: Schematic mechanism of formation of the RSA signal: Depending on the spin precession frequency, the interference is constructive or destructive, leading to particular multi-peak shape. (Adopted from [58])

magnetic field. Therefore in the RSA measurement, the magnetic field is scanned while the delay between pump and probe is kept constant, usually at a small negative delay. A sharp resonance peak is detected when the applied magnetic field is fixed to a particular value where the optically oriented spin polarization precesses around it by an integer (n) multiple of the laser repetition rate so that constructive interference occurs:

$$\omega_L = \frac{g\mu_B B}{\hbar} = n f_{rep} \quad (3.34)$$

In contrast, Larmor frequencies ω_L , which don't meet the condition 3.34, results in the destructive interference of the spin ensembles generated by subsequent pulses and in general to a low overall polarization of the entire spin ensemble. A scheme of the working principle of this technique is depicted in Fig. 3.10. The experimental setup for RSA technique is the same that of the TRKR measurement, as shown in Fig. 3.5. The only difference is that in RSA the pump-probe delay time Δt is held constant, typically at a small negative value, and the Kerr rotation is monitored as the external magnetic field is swept. When the resonance condition is satisfied, a series of sharp resonance peaks appears, as depicted in Fig. 3.11.

The line width of these sharp resonance peaks allows to evaluate the spin dephasing time, and their spacing yields the carrier g -factor. The systems with a long spin dephasing time are characterized by narrower peaks while wider peaks can be observed in systems with a short spin dephasing time. If the spin dephasing times are only about half of the repetition period, in such situation the RSA peaks degrade to sinusoidal modulation of magnetization, and accurate spin dephasing time can not be deduced. In the case of

FIGURE 3.11: Typical shape of RSA trace for Sample B. The data has taken by sweeping external magnetic field while keeping the pump-probe delay fixed at $\Delta t = -0.24$ ns.

isotropic spin relaxation, all the peaks have the same height, and the spin components of the carriers oriented along the growth axis and normal to it (in-plane oriented spins) relax at the same rate. In such case the overall spin dephasing time and electron g -factor can be extracted by fitting the data to Eq. 3.33 [46]. The presence of spin relaxation anisotropy influences the relative amplitudes of the RSA peaks as evidenced by a shorter zero-field peak compared to the side peaks in Fig. 3.11. Chapter 5 is devoted to understanding the nature of anisotropic spin relaxation in our studied structure.

3.8 Current-induced spin polarization

Current-induced spin polarization (CISP) is a phenomenon in which the charge carriers of a semiconductor become spin-polarized when an electric current is driven through the device. This technique of creating spin polarization under the action of electrical bias differs radically from those described so far. In this technique, the circularly polarized optical pulses are replaced by electrical voltage pulses, which directly generate the spin imbalance in the semiconductor. It has potential to be employed in the technological platforms and is useful for device applications such as spin-based information processing [81], the electrical control of magnetization [82], Resonant optical control of electrically induced spin polarization [83], and as a technique of electrically inducing spin polarization [13]. The idea to orient the spin of charge carriers by all electrical

FIGURE 3.12: Schematic for the CISP measurements: Kerr rotation in reflection is used to measure the out-of-plane component of spin polarization.

FIGURE 3.13: *Magnetic field dependence of FR for $\vec{E} = 0$ (top), $\vec{E} \perp \vec{B}_{ext}$ (middle), and $\vec{E} \parallel \vec{B}_{ext}$ (bottom). where the insets show the geometries [95].*

means was first proposed by Ivchenko and Pikus in gyrotropic crystals about two decades ago [84], where an inverse effect was observed i.e. photogalvanic current was generated by illuminating tellurium crystal with circularly polarized light. While using the same material the rotation of linearly polarized light upon application of an electric current was observed by Vorob'ev and collaborator [85]. They found that the angle of rotation was proportional to the magnitude of the current, and reverses its sign as a result of change in the direction of current, this was supposed to be an electrical analog of the Faraday effect [86].

In the 1970s, it was theoretically suggested by Dyakonov and Perel [87] that in semiconductors with strong spin-orbit coupling, an electric field acts on a moving charge carrier as an effective magnetic field and may be used to orient the spin. In low dimensional systems, the theory of spatially homogeneous spin polarization resulting from an electrical current was developed in [88–90]. Eventually, the first experimental evidence of this effect was received in semiconductor quantum wells [91, 92] as well as in strained bulk InGaAs [93]. Further in two-dimensional electron gases confined in AlGaAs quantum wells it was spatially imaged along with the spin Hall effect [94].

In this work, the measurements of the electrically generated spin polarization were done using Kerr rotation without the use of an optical pump as depicted in Fig. 3.12. The

conduction band electron spins were oriented electrically by using AC voltages modulated at 1.1402 KHz for lock-in detection. An internal magnetic field was produced by applying an electric field of different magnitudes across different contacts of macroscopic Hall bar in a liquid helium flow cryostat. By measuring spatially-resolved Kerr rotation, we can quantify the direction and magnitude of the spin-orbit field. The authors of Ref. [95] reported that, if the component of the spin-orbit field is perpendicular to the applied magnetic field it will cause the suppression the zero-field peak compared with the nearby peaks. In the opposite case, if the component of the spin-orbit field is parallel to the applied magnetic field it will cause the curve to shift horizontally (see Fig. 3.13).

3.9 Equipments for measuring low temperature spin dynamics

3.9.1 The magneto-optical cryostat

In order to be able to see various quantum phenomena in semiconductor nanostructures, it is advantageous to use very low temperatures. The excitations, far above the band gap, which are necessary to access a large density of states, leads to the deposition of excess energy in the sample. The heating effect caused by this excess energy can result in sample damage and degradation with time. To avoid the samples from such light-induced damage one should lower the temperature. The standard technique for lowering

FIGURE 3.14: (a) Schematics of the ^4He cryostat and (b) internal structure of the cryostat with a variable temperature insert for measuring down to 1.2 K.

temperatures involves the use of cryogenic liquids N_2 , 4He and 3He . To lower the sample temperatures and apply magnetic fields a 4He cryostat was used as shown in Fig. 3.14.

The 4He cryostat, at LNMS, is outfitted with a split coil superconducting magnet (that can reach magnetic fields up to ± 8 T), and a variable temperature insert (VTI) for measurement in the range from 1.2 K to 300 K. The cryostat is built like an onion (See Fig. 3.14 (b)). The core is VTI, where the sample is inserted and cooled by He vapors, is completely isolated from the surrounding by vacuum chamber and liquid He main bath. The He vapor can be injected into the bottom of the VTI by opening the needle valve in a tube connecting it with the main bath. The main bath is well isolated from the VTI insert by vacuum. The He main bath (4.2 K) is isolated from surrounding by vacuum, and liquid nitrogen which helps to decrease the temperature up to 77 K. The cryostat has four windows, allowing access to the sample parallel and perpendicular to the direction of the magnetic field. The pump and probe beams are incidents normal to the sample surface and perpendicular to the external magnetic field (for details see Sec. 3.10). The beams are focused by a motorized lens on the sample. In the case of CISP, the lens can be automatically positioned in the plane normal to the beam direction and thus, the laser spot can scan the sample.

3.9.2 Temperature Control

For temperature-dependent spin dynamics measurements, a variable temperature insert can be used. To lower the temperature down to 4.2 K the sample space should be filled with liquid He by opening the needle valve. The temperature can be further decreased by pumping the vapors above the liquid to reduce the pressure to its lowest possible level. In this way, it is achievable to reduce the temperatures down to 1.2 K; however, this procedure results in significant helium consumption. A resistor mounted close to the sample can be used as a heater to go to the temperature above 4.2 K.

3.9.3 Magnetic field control

As described above, a split coil superconducting magnet located in the bottom part of the cryostat is used to generate a magnetic field. At cryogenic temperature the current flows through the coils without energy dissipation and produces a magnetic field in the range from -8 T to +8 T. Depending on the choice of the sample both in- and out-of-plane magnetic field can be applied to the sample. We measure the spin dynamics in the Voigt geometry where the magnetic field is applied parallel to sample plane and laser beams are incidents normal to sample plane.

3.9.4 Data acquisition setup

In the data acquisition setup, the personal computer plays the central role which controls different devices via National Instruments's LabVIEW program. A VTI controls the sample heating while temperature controller regulates the sample temperature through a heating coil on the sample element. The mechanical delay line is regulated by a program in LabVIEW. The magnetic field can be set via current-to-field calibration by a power supply (magnetic field controller). The balanced bridge and lock-in detection scheme are used for recording the polarization rotation (See Sec. 3.10 for more details).

3.10 Optical setup

The schematic representation of the optical setup is shown in figure 3.15. In this section, we are going to describe the beam path and the optical components used as well as alignment procedure.

3.10.1 Laser Light Sources

We start with the laser as an excitation light source. The laser pulses are generated by a mode-locked Ti: sapphire laser (Coherent Mira), pumped by a Coherent Verdi laser system. This is an ultrafast laser that provides optical pulses in the near-infrared wavelength range, with a repetition rate 76 MHz (corresponding to the repetition period of 13.2 ns) and pulse duration of 100 fs.

3.10.2 Collimation of beam

After the laser beam (black) gets out of the laser, it is then collimated with an iris, and a lens pair labeled C in Fig. 3.15. As we deal with non-ideal Gaussian beams, it is not possible to get a perfectly collimated beam. For the pump beam, it is important because it goes through the delay line and for different positions of the delay line it passes different distances to the surface of the sample. During delay scan, it causes the drift of the diameter of the beam; as a result, the photon density changes. To minimize this unwanted effect beam is collimated in such a way that the neck of the non-ideal Gaussian beam is kept in the middle of the delay line.

FIGURE 3.15: Block diagram of the Optical setup: Pump (red) and probe (green) beam are separated via beam splitter cube (BS). The detection system consists of an optical bridge with a balanced detector and double lock-in amplifiers. For detailed explanation of the elements see main text.

3.10.3 Pump, probe, and delay

After collimation, a beam splitter cube (BS) divides the principal beam into the pump and probe beam. Thus, the intensity ratio of pump and probe is adjustable, and the beams are polarized perpendicular to each other. Usually, the pump and probe intensity ratio is chosen to about 9:1. The probe intensity can be additionally tuned by a neutral density filter (F). The pump beam is directed to the delay line because it is preferable to have a very stable beam path for the probe beam onto the detectors. This computer-controlled mechanical delay line with a retro-reflector (used to minimize beam misalignment due to stage movement) mounted on it induces the time delay between pump and probe beams, by changing the path length of pulse beam. The total length of the mechanical delay line used in our setup is 0.5 m, with $c = 3 \times 10^8$ m/s it allows for the time delay up to $\Delta t = 3.25$ ns for a single pass. To access longer Δt , one should align the pump such that it pass twice the delay line, and will induce a total delay length up to 6.5 ns (i.e. double of the single pass).

To create non-equilibrium spin polarization, circularly polarized light is needed. For that reason, the pump pulse after being delayed is passed through a photo-elastic modulator (PEM). PEM is a device which modulates the initially linearly polarized light so that the transmitted beam oscillates from left to right circular polarization at 50 kHz. Compared to the pump the probe beam is directed via a fixed and relatively simple path. To roughly equalize the optical path lengths of the pump and the probe beam, the probe beam is reflected off by a set of mirrors. The length of this path determines the range of the pump-probe delay time. In the present work, the optical path is adjusted so that it allows for the time delay between -0.75 and 2.5 ns. The probe beam is then modulated at a few kHz by a mechanical chopper wheel.

3.10.4 Focusing and Imaging

As the pump and probe pulses are delayed with respect to each other, modulated and filtered, they are ready to be focused on the sample surface. With a manual steering mirror, the pump and probe are aligned parallel to each other and directed to the entrance window of the cryostat. Both the pump and probe beams are then focused by a lens (L) onto the sample at the same point. The angle within the pump and probe beams is about 2-3 degrees, and it can be considered as the normal incident condition. For TRKR analysis the spatial overlap is crucial. For this purpose, the pump spot size is adjusted so that it exceeds the probe spot size at the location of pump-probe overlap in the sample plane. Such configuration guarantees that the signal will not change by small variations of (i) the probe position within the sample plane and/or (ii) the probe

spot size due to beam divergence while scanning the delay. Preliminary alignment of the spot overlapping can be done by directing and focusing the beam reflected from the sample onto a video camera, which shows the position of spots on the sample.

3.10.5 The Balanced Photo-diode Bridge

Upon reflection from the surface of the sample, the pump is blocked, and the probe is guided through an iris to reduce as much of the stray light stemming from the pump as possible. A $\lambda/2$ -retarder is used to rotate polarization followed by a polarizing beam splitter, which splits the reflected probe beam into its horizontally and vertically linear polarization components. The two orthogonal beams are then directed onto photodiodes of a balanced diode bridge, which is highly sensitive to intensity differences (A-B) of both beams. Before running the experiment polarization bridge is being balanced. That is accomplished by adjusting the polarization of the probe beam using $\lambda/2$ -plate which allows balancing the detector contributions while the optical paths of the beams remain unchanged. For the automatic balance, the $\lambda/2$ retarder is rotated until the (A-B) signal from the diode bridge is minimized. That can be done while pump beam is blocked. By unblocking pump spin polarization imbalance is introduced into the system, that results in the change of the probe polarization and thus destroys the balance of the diode bridge. In the absence of spin polarization in the sample, the difference between the signals from both detectors is now zero. The intensities of light in the detectors become different when there is a spin polarization in the sample. The simplified working principle of the diode bridge is illustrated in Fig. 3.16. After focusing the two orthogonal beams onto photodiodes of the balanced bridge, a respective photocurrent I_A or I_B and

FIGURE 3.16: Optical setup of diode bridge: The two perpendicularly polarized components (black) are directed through lenses onto the photodiodes. Electrical currents I are represented by green arrows. Where the triangles ($\triangleright$) with $\pm$ symbols are op-amps arranged for linear amplification and conversion of the probe-induced photocurrent to a measuring voltage by resistance. (Adopted from [96])

also a difference of these currents $I_A - I_B$ at their junction are generated. The diode bridge contains electronics, that convert all these currents into voltages V_A , V_B and V_{diff} depending on the values of the resistors $R_{A/B}$ and R_{diff} . Measuring these voltages allows one to measure precisely a very tiny angle of rotation of the probe polarization.

3.10.6 Lock-in detection

The fundamental concept behind the lock-in techniques is to improve the signal to noise ratio in these measurements. A lock-in amplifier efficiently rejects the noise due to pump scatter and room lights by isolating the component of the input signal which is modulated at a known reference frequency. To retrieve Kerr signal, we use two lock-in amplifiers which isolate the component of the measured signal which is modulated at both the PEM frequency and the chopper frequency. The signal from the diode bridge is sent to the lock-in 1 (See Fig. 3.15) which is synchronized with PEM. The output signal of the lock-in 1 is further amplified by lock-in 2 at the trigger frequency of the chopper. Finally, the Kerr rotation signal is measured at the output of lock-in 2.

In the case of multilayer sample where each layer could contribute to the Kerr rotation, or in case if the region of interest in the sample is not homogeneously spin polarized, the measured signal is a superposition of these various contributions. In such situations, the resulting Kerr signal is complicated and requires a very careful and precise analysis.

3.11 Summary

We presented the realization of a setup for time-resolved Kerr rotation, Resonant spin amplification, and current-induced spin polarization measurements which can be used in the temperature range from 2 to 300 Kelvin while applying a homogeneous magnetic field with a value in the range from 0 to ± 8 T.

Chapter 4

Long-lived nanosecond spin coherence in high-mobility 2DEGs confined in double and triple quantum wells

The present chapter is assigned to the electron spin dynamics of a high mobility dense two-dimensional electron gas in multilayer GaAs quantum wells. The dynamics of the spin polarization was optically studied using pump-probe techniques: time-resolved Kerr rotation and resonant spin amplification. For samples doped beyond the metal-to-insulator transition, the tailoring of the spin-orbit interaction by the sample parameters of structural symmetry (Rashba constant), width and electron density (Dresselhaus linear and cubic constants) allows us to attain long dephasing time in the few nanoseconds range in double and triple quantum wells. The determination of the scales: transport scattering time, single-electron scattering time, electron-electron scattering time, and the spin polarization decay time with and without magnetic field further supports the possibility of using n-doped multilayer systems for developing spintronic devices.

4.1 Motivation

A milestone for practical applications of spintronic devices is a long-lasting spin coherence time (T_2^*) for ensembles [97]. The tunability of T_2^* have been widely studied in semiconductor quantum wells (QWs) with a variety of optical techniques developed for

The results presented in this chapter are based on Ref. [1] on page 147

the study of the spin polarization dynamics and the spin relaxation mechanisms [98–101]. For example, in n-type samples, it was observed that the doping level has a major role in attaining or limiting long coherence time with T_2^* changing from tens of picoseconds up to nanoseconds [46, 102–104]. The turning point, where T_2^* decreases with an increase of the electron concentration, was found at the metal-to-insulator transition for bulk ($2 \times 10^{16} \text{ cm}^{-3}$) [48, 105] and GaAs QWs ($5 \times 10^{10} \text{ cm}^{-2}$) [51]. Beyond this point, the Dyakonov-Perel (DP) spin relaxation mechanism is dominant and controlled by electron-electron collisions [106]. The DP mechanism defines a decay time, for the spin polarization along the QW out-of-plane direction, limited by the spin-orbit mechanisms giving the path to long-lived spin coherence. It can be calculated according to:

$$t_z^{-1} = 8D_s m^2 \hbar^{-4} [\alpha^2 + (\beta_1 - \beta_3)^2 + \beta_3^2] \quad (4.1)$$

Where, D_s is the spin diffusion constant, α is the Rashba coefficient due to structural inversion asymmetry, and β_1 and β_3 are the linear and cubic spin-orbit constants due to the Dresselhaus bulk inversion asymmetry [107, 108].

Recently, our group demonstrated that multilayer QWs are exceptional platforms for the investigation of current-induced spin polarization effects [83, 172]. While such complex systems also offer new possibilities for applications, for example in the production of spin blockers [110] and filters [44]. The study of long-living spin coherence in double (DQW) and triple quantum wells (TQW) is still required. Here, we report on the coherent spin dynamics in multilayer quantum wells using time-resolved Kerr rotation (TRKR) and resonant spin amplification (RSA). The sample structure allowed to control the DP mechanism by tailoring the spin-orbit constants through the well width, symmetry and subband concentration parameters. Remarkably, it results in coherence times in the nanoseconds range even for DQW and TQW samples with individual subband density beyond the metal-to-insulator transition.

4.2 Experimental realization

The TRKR and RSA techniques were used to probe the coherent spin dynamics of the electron gas here. For optical excitation, we used a mode-locked Ti-sapphire laser with pulse duration of 100 fs and repetition rate of $f_{rep} = 76 \text{ MHz}$ corresponding to a repetition period (t_{rep}) of 13.2 ns. The time delay Δt between pump and probe pulses was varied by a mechanical delay line. The pump beam was circularly polarized by means of a photoelastic modulator operated at a frequency of 50 kHz. The probe beam was not modulated, and the rotation of its polarization upon reflection was recorded as a function of Δt and detected with a balanced bridge using coupled photodiodes. The

FIGURE 4.1: *The DQW band structure and the charge density for the first and second subbands.*

laser wavelength was tuned to obtain the TRKR energy dependence in each sample. The samples were immersed in the variable temperature insert of a split-coil superconductor magnet in the Voigt geometry.

The spin dynamics are performed here with experimental results on three different samples grown in the [001] direction, one double (Sample A) and two triple quantum well (Sample B and C), containing a dense two-dimensional electron gas (2DEG) with equal total density (See Sec. 3.2 for more details). The DQW sample is a wide doped GaAs well where the electronic system forms a DQW configuration with symmetric and antisymmetric wave functions for the two lowest subbands with subband separation $\Delta_{12} = 1.4$ meV and approximately equal subband density n_s [111]. Fig. 4.1 shows the calculated DQW band structure and the charge density for both subbands.

4.3 Spin dynamics in Sample A

4.3.1 TRKR signal as a function of applied magnetic field

The time evolution of the spin dynamics for the DQW with no magnetic field and in the transverse magnetic field up to 2 T is demonstrated in Fig. 4.2 with pump/probe power of 1 mW/300 μ W. The signal results into a slow decay in the absence of applied magnetic field B and into weakly damping oscillations in the presence of B . The TRKR oscillations are associated with the precession of coherently excited electron spins about the in-plane magnetic field. To obtain the spin coherence time, the evolution of the Kerr

FIGURE 4.2: *KR as function of the pump-probe delay for different magnetic fields. The symbols correspond to experimental data while the solid lines correspond to a single exponential decay ($B = 0$ T) and exponentially damped cosine functions ($B \neq 0$ T).*

rotation angle can be described by an exponentially damped harmonic:

$$\Theta_K(\Delta t) = A \exp(-\Delta t/T_2^*) \cos(\omega_L \Delta t + \varphi) \quad (4.2)$$

where A is the initial spin polarization build-up by the pump, φ is the oscillation phase, and $\omega_L = g\mu_B B/\hbar$ is the Larmor frequency with magnetic field B , electron g -factor (absolute value) g , Bohr magneton μ_B , and reduced Planck's constant $\hbar$. The solid lines are a single exponential decay or exponentially decaying cosine functions to extract spin dephasing times and the effective Landé g -factor. The resulting magnetic field dependence of ω_L and T_2^* are shown in Fig 4.3. One can clearly see that the spin precession frequency increases with B following the linear dependence of the Larmor frequency on the applied field. The value of the fitted g -factor is 0.4532 ± 0.0002 which is close to absolute value for bulk GaAs and similar to the value measured for a quasi-two-dimensional system in a single barrier heterostructure with two-subbands occupied [112].

According to the Dyakonov-Perel mechanism, the observed exponential decay at $B = 0$ corresponds to the strong scattering regime. In the opposite case, where the spin precesses more than a revolution before being scattered, an oscillatory behavior would be expected [106, 113]. The measured value of the decay time of the spin polarization along the z -direction (out-of-plane) at zero external fields is 1.1 ns. For our symmetric, wide, and dense quantum well, we estimate $\alpha \simeq 0$, $\beta_1 = -\gamma(\pi/w)^2 = 0.49 \times 10^{-13}$ eVm, and

FIGURE 4.3: (a) Larmor frequency ω_L and (b) T_2^* fitted as function of B .

$\beta_3 = -\frac{1}{2}\gamma\pi n_s = 0.70 \times 10^{-13}$ eVm for the first subband using $\gamma = -10$ eVÅ³ [114]. The charge diffusion constant can be estimated from the Fermi velocity $v_F = \hbar\sqrt{2\pi n_s}/m^*$ and the transport scattering time $\tau_m = \mu m^*/e = 70$ ps, using the effective mass m^* and the electron's charge e , by $D_c = v_F^2\tau_m/2 = 3$ m²/s. The spin diffusion constant is approximately two orders of magnitude smaller than the same constant for charge [107]. Scaling $D_s = 100$ to 300 cm²/s, we obtain $t_z \sim [8D_s m^{*2} \hbar^{-4} \beta_3^2]^{-1} = 1.1$ to 3.3 ns. Thus, the data at $B = 0$ agrees with a DP mechanism where the spin dynamics are dominated by the cubic Dresselhaus term. The cancellation of $\alpha \simeq 0$ and $\beta_1 - \beta_3 \simeq 0$ through the sample parameters shows a practical path for long-lived spin coherence in highly doped QWs.

Increasing the magnetic field up to 0.5 T, the data yields a longer T_2^* . In this situation, the cyclotron motion acts as momentum scattering and leads to a less efficient spin relaxation in agreement with the DP model [115]. It is important to note that an in-plane magnetic field was applied using the Voigt configuration, it implies that the cyclotron motion is realized perpendicular to the QW plane and allowed due to the large QW width. The increase follows a quadratic dependence [116] with $T_2^*(B)/T_2^*(0) = 1 + (\omega_c\tau_p^*)^2$ where ω_c is the cyclotron frequency and τ_p^* is the single-electron momentum scattering time. We found $\tau_p^* = 0.92$ ps in agreement with the magnitude of the quantum lifetime measured by transport from the Dingle factor of the magneto-intersubband oscillations on the same sample [117]. The value for τ_p^* is also in agreement with the determination of approximately 0.5 ps for QWs of shorter width [114]. One of the reasons for the significant difference between τ_m and τ_p^* is the insensibility of the first to electron-electron scattering. The ratio of $\tau_m/\tau_p^* \simeq 100$ implies that the dominant scattering from impurities is due to remote instead of background impurities [118]. If we consider that

$1/\tau_p^* = 1/\tau_m + 1/\tau_{ee}$, we get a time scale of $\tau_{ee} \approx \tau_p^*$ which demonstrates that electron-electron collisions dominate the microscopic scattering mechanisms as expected [106]. Additionally, a further increase of the magnetic field leads to a faster oscillations decay due to the spread of the g-factor within the measured ensemble [119, 120]. The size of the inhomogeneity Δg can be inferred by fitting the data according to $1/T_2^*(B) = 1/T_2^*(0) + \Delta g \mu_B B / \sqrt{2} \hbar$ as shown in Fig. 1(d). From the $1/B$ dependence [46, 103], we obtain $\Delta g = 0.002$ or 0.44% and $T_2^*(0) = 2$ ns.

4.3.2 Effect of optical power on spin dephasing of electron

The optical power influence on the time evolution of the spin dynamics at $B = 1$ T for the DQW sample are shown in Fig 4.4(a). Only at low pump power, we observed negative delay oscillations of considerably large amplitude. The observation of electron spin polarization before the pump pulse arrival indicates that the spin polarization persists from the previous pump pulse, which took place $t_{rep} = 13.2$ ns before. The excitation power dependence of T_2^* is plotted in 4.4(b) yielding an exponential decay. For single QW structures, the decreasing of the coherence time at high pump density was associated with the electrons delocalization caused by their heating due to the interaction with the photogenerated carriers [119]. A similar decrease was also attributed to an increased efficiency of the Bir-Aronov-Pikus mechanism induced by the larger hole photogenerated density in GaAs QWs [121]. However, it is unlikely to be relevant in our dense 2DEG where the photogenerated hole loses its spin and energy quickly and recombines with an

FIGURE 4.4: (a) TRKR of the DQW as function of pump power and (b) the corresponding T_2^* .

electron from the 2DEG. Nevertheless, as a key parameter for spin devices, we note that T_2^* remains near the nanoseconds range when the power is raised by almost one order of magnitude. At high excitation power, an additional short-living component in the signal becomes more significant. Upon the association of the 2DEG electrons dynamics with the long-lasting oscillations, rather than with excitons or photo-excited electrons [119], the use of a single exponential fitting mixes and underestimates T_2^* .

4.3.3 The influence of the pump-probe delay on the spin dephasing time

In systems where T_2^* is comparable or longer than the laser repetition period, one can use the RSA technique to extract the spin dephasing time by scanning the magnetic field at a fixed pump-probe delay [46]. Fig 4.5 displays the RSA signals measured at different Δt with pump/probe power of 1 mW/300 μ W. We observed a series of sharp resonance peaks as a function of B corresponding to the electron spin precession frequencies which are commensurable with the pump pulse repetition period obeying the periodic condition: $\Delta B = (\hbar f_{rep})/(g\mu_B)$ [46]. As function of the magnetic field, the RSA peaks amplitude decreases as result of the g -factor variation within the measured ensemble as commented. The RSA resonances are modulated by a slow oscillation that depends on $f_d = 1/\Delta t$

FIGURE 4.5: RSA scan of the DQW system obtained for different time delays. $\lambda = 817$ nm, $T = 5$ K.

FIGURE 4.6: The comparison of zero-field resonance peaks at different pump-probe delay. Open symbols are experimental data and solid lines are Lorentzian fit to the data.

according to the same periodic condition described above. Increasing the pump-probe delay causes the broadening of RSA peaks corresponding to a shortened spin dephasing time (See Fig. 4.6).

FIGURE 4.7: (a) T_2^* and (b) Amplitude dependence on Δt extracted from Fig 4.6

T_2^* can be directly evaluated from the width of the zero-field resonance using a Hanle (Lorentzian) model [46, 83]:

$$\Theta_K = A/[(\omega_L T_2^*)^2 + 1] \quad (4.3)$$

with half-width $B_{1/2} = \hbar/(g\mu_B T_2^*)$. The fitting result is displayed in 4.6 for negative and positive delays. The extracted values of T_2^* and amplitude are shown in 4.7 as function of Δt . For positive delays, both quantities display an exponential decay (solid line). However, the system coherence is recovered just before the pump arrival for the long-lived spin component in the system dynamics [122]. The RSA signal measured at negative delay gives $T_2^* = 4.4$ ns. Concerning the subband dependence, the spin relaxation time was calculated to be identical in an electron system with two occupied subbands, although the higher subband may have a much larger inhomogeneous broadening, due to strong intersubband Coulomb scattering [112, 123]. In our samples, we studied the pump/probe wavelength dependence as reported in TRKR [112] and photoluminescence [124, 125] studies of similar multilayer systems.

4.3.4 The pump-probe wavelength dependence of spin dephasing time

Figure 4.8 displays the RSA of the DQW sample for different pump-probe wavelengths at a fixed delay while scanning the external magnetic field. The zero-field resonance

FIGURE 4.8: RSA scans of the DQW sample measured for different pump-probe wavelengths.

FIGURE 4.9: (a) The Spin coherence time T_2^* and (b) the amplitude extracted from Fig 4.8.

peaks were fitted to to Eq. 4.3 as described above. T_2^* and the amplitude obtained from the fit increase with the pump-probe wavelength as shown in Figures 4.9(a) and (b). Increasing the pump-probe energy about 3 meV ($\simeq 2\Delta_{12}$), from 817 nm to 815 nm, T_2^* decreases less than 10% in Figure 4.9(a). This small change could be associated with the relative similitude between the charge density distribution for both subbands. On the other side, fast intersubband scattering may hide a difference in the spin-orbit interaction for the second subband as shown in recent calculations [126].

FIGURE 4.10: Band diagram and charge density for the TQW (Sample B).

4.4 Spin dynamics in Sample B

In the present section, we turn our attention to the TQW (sample B). Fig. 4.10 shows the calculated band diagram and charge density for three occupied subbands. The coupling strength between the quantum wells is characterized by the separation energies Δ_{ij} of the three occupied subbands ($i, j = 1, 2, 3$) given by $\Delta_{12} = 1.0$ meV, $\Delta_{23} = 2.4$ meV, $\Delta_{13} = 3.4$ meV [127].

4.4.1 Excitation wavelength and temperature dependence of spin dynamics

For the choice of the right excitation energy, the laser wavelength was tuned looking for the maximum Kerr rotation signal with long-lasting spin coherence time. In the current section, we give an overview of what will be different upon a change in the excitation wavelength. In order to characterize the Kerr rotation amplitude, T_2^* , and Landé g -factor dependence on the excitation wavelength the TRKR measurements were performed with and without external magnetic field. Characteristic TRKR data as function of Δt for different excitation wavelengths at $T = 4.2$ K are shown in Fig. 4.11 without external magnetic field.

FIGURE 4.11: Time resolved Kerr rotation of sample B without external magnetic field at $T = 4.2$ K and $p_{\text{pump}} = 1$ mW.

FIGURE 4.12: (a) Amplitude at time delay $\Delta t = 0$ and (b) decay time extracted from Fig. 4.11 plotted as a function of wavelengths.

The respective signs of amplitudes are different for the low and high wavelength scan. The experimental data are fitted well by an exponential decay. The fit, again represented by a red solid line, gives an amplitude of the initial spin polarization and the decay time, which are displayed in Fig. 4.12(a) and (b). One can clearly see that as we pass through

FIGURE 4.13: Kerr rotation signal measured for sample B at different excitation wavelengths. $B = 1 \text{ T}$, $T = 10 \text{ K}$ and $p_{\text{pump}} = 1 \text{ mW}$.

the band gap energy, the Kerr rotation extrema occur in the opposite direction. A similar observation of Kerr rotation extrema occurring on either side of the band gap has been reported in TRKR study of similar multilayer system [31]. The signal at $\lambda = 819$ nm included two exponential decay and was fitted to a bi-exponential decay function. The fast decay may include the hole spin dynamics yields a decay time of 14 ps and has been omitted from the data in Fig. 4.12(b). In the wavelength range from 813 nm to 821 nm, the spin lifetime increases and yield a maximum of 1 ns at 821 nm.

Fig. 4.13 displays the wavelength dependence of TRKR as a function of the time delay at a fixed magnetic field of 1 T. For higher wavelengths the signal presents a long dephasing time with negative delay oscillations of amplitude comparable with the positive delay ones. Figure 4.14(b) shows T_2^* extracted from 4.13. T_2^* shows a decrease from 3.25 to 2.91 ns when increasing the laser energy about 3 meV ($\simeq \Delta_{13}$) from 823 nm to 821 nm. The decrease is stronger on the RSA measurements at positive and negative delays presented in Sec. 4.4.3. The linear dependence of Larmor precession frequency as a function of the magnetic field for several wavelengths is presented in Figure 4.14(a). The curves are vertically offset clarity. The electron's g-factor was obtained from the slope of 4.14(a) and plotted as a function of wavelength in Figure 4.14(b). We found that g-factor varies from 0.455 ± 0.001 to 0.446 ± 0.002 when the laser wavelength is scanned from 815 to 823 nm.

To give an impression of the temperature influence on the spin dynamics of electrons, Figure 4.15 shows KR signal measured on sample B for three different temperatures.

FIGURE 4.14: (a) Larmor frequency as a function of applied magnetic field for different excitation wavelengths. (b) Spin dephasing time T_2^* (open circles) and electron g-factor (solid circles) extracted from Fig. 4.13 and panel (a) respectively. The curves in 4.14(a) are vertically offset for clarity.

FIGURE 4.15: *TRKR traces of sample B, obtained at three different sample temperatures of 1.2 K, 5 K, and 10 K using excitation power of 1 mW at external magnetic field of 1 T.*

The curves are normalized to zero points on the time scale which corresponds to moment when spin polarization was induced in the sample by a circularly polarized pump pulse. A common feature of the signal is the appearance of a long-lived spin beating as can be seen at negative time delays. One can clearly see that at a lower temperature (1.2 K) the Kerr signal displays some unusual time-resolved curve near the time origin. Additionally, at low temperatures, the signal displays oscillatory behavior in the background which produces difficulty for the analysis. However, the signal have larger amplitude at $T = 10$ K. In chapter 6, the influence of sample temperature on the spin dynamics will be discussed in more details.

In a nutshell, we found a strong dependence on a wavelength with the maximum signal at 817 and 821 nm and long coherence time at a higher wavelength. Therefore, the experimental conditions for maximum KR signal with long-lasting spin coherence, namely $\lambda = 817, 821$ nm and $T = 10$ K, were used in the following sections.

4.4.2 Magnetic field dependence of TRKR signal

In this section, the influence of magnetic field on the electron spin dynamics is addressed. Figure 4.16 (a) shows the dependence of TRKR signal on the magnetic field for sample B

FIGURE 4.16: (a) TRKR data of sample B as function of Δt recorded for different magnetic fields at $\lambda = 817$ nm. For clarity of presentation, the data has scaled to the same initial amplitude. The spin dephasing times extracted by fitting raw data to decaying cosine function are plotted as function of applied field in (b).

at a pump-probe wavelength of 817 nm. We do not observe long electron spin coherence and the spin polarization completely decay before the next pulse arrival.

FIGURE 4.17: (a) Kerr rotation signal measured for sample B as function of delay between pump and probe at different applied magnetic fields. (b) The Larmor frequency (open circles) evaluated from (a) as a function of applied magnetic field. The solid red line is linear fit to the data. $\lambda = 821$ nm, $T = 10$ K.

The TRKR signals are well modeled by exponentially decaying cosine functions mapping the Larmor precession of the optically induced electron spin polarization. The fits

extract spin relaxation times which are represented in Fig. 4.16(b). The spin dephasing time decreases from 1.1 ns to 635 ps with growing magnetic field from 0.5 T up to 4 T. The TRKR scans for the electron spin coherence is shown in Fig. 4.17 without external magnetic field and for different magnitudes of applied field. The TRKR scans are measured on sample B after excitation at pump-probe wavelength $\lambda = 821$ nm and optical pump power 1 mW while the sample is cooled to $T = 10$ K. We observed here long-lived electron spin coherence as reflected by the KR oscillations at negative time delays. Due to the long spin coherence comparable with the laser repetition period, there is almost no decay over the measured time window (2.5 ns). The extracted values of Larmor frequency, by fitting the data to Eq. 4.2, are displayed in Fig. 4.17(b) as a function of applied magnetic field. Linear fit to the data yields a g -factor of about $|g| = 0.453 \pm 0.001$.

4.4.3 Pump-probe delay dependence of Resonant spin amplification

Due to the long-lived spin coherence, as commented above, the signal does not completely decay from the previous pulse and overlaps with the signal induced by the subsequent pulses. As a result, the spin polarization accumulates in the sample and the dephasing time can not be extracted by using the TRKR technique. Therefore, in analogy to the DQW sample, we use the constructive interference of the coherence oscillations from successive pulses for extracting the spin coherence time by the RSA technique. Fig. 4.18(a) and (b) present the magnetic field scans of the KR amplitude performed at different pump/probe separation for 821 and 823 nm, respectively. From the Lorentzian

FIGURE 4.18: RSA scans of the TQW sample measured for different pump-probe delays with the corresponding extracted spin dephasing time at (a) 821 nm and (b) 823 nm.

fit of the zero-field peak, as for the DQW, the spin dephasing for the TQW sample was obtained revealing the longest $T_2^* = 10.42$ ns at a negative delay. In this case, the same energy increase (~ 3 meV $\simeq \Delta_{13}$), leads to strong T_2^* decrease of almost 50%/30% at negative/positive delay. We note that, contrary to the DQW case, the third subband for the TQW have opposite charge distribution if compared with the lower subbands. While the third subband has the charge density more localized in the central well, the electrons in the second and first subbands are distributed in the side wells.

4.5 Spin dynamics in Sample C

Finally, we focus on the spin dynamics in TQW sample C. The detailed description of the sample is given in section 3.2. The TQW band structure and subband charge density are illustrated in Fig. 4.19.

FIGURE 4.19: *TQW band structure and subband charge density. The black lines shows the potential profile, and the colored lines show the occupied eigenstates of the first (green), second (red) and third (blue) subbands.*

4.5.1 Pump-probe wavelength dependence of spin dynamics

TRKR signals measured by the varying excitation wavelength of the laser pulses over the range from 811 nm to 821 nm with the same pump power under the external magnetic field of 1 T are shown in Fig. 4.20. We can clearly see that the decay of the Kerr rotation signals is changing with excitation wavelengths. In order to get information over spin dephasing time and electron g -factor the precessional signal displayed in Fig. 4.20(a) was fitted with mono-exponential decaying cosine function as shown by the red curves plotted on the top of experimental data. The obtained T_2^* and g -factor are shown

FIGURE 4.20: *TRKR traces taken at a sample temperature of 10 K for different pump-probe wavelengths at fixed pump power of 1 mW. The spin beats live longer at lower wavelengths. (b) The relative spin dephasing time and g -factor extracted from (a).*

in Fig. 4.20(b). The spin dephasing time strongly decreases with the increase of laser wavelength. Additionally the electron g -factor shows a variation of 0.028 in the measured range of wavelength having the maximum at 821 nm.

FIGURE 4.21: *(a) The dependence of the TRKR signals on external magnetic field at $P_{\text{pump}} = 1$ mW. (b) The corresponding Larmor precession frequency, as well as spin dephasing time.*

4.5.2 Dependence of spin dynamics on applied magnetic field

In order to investigate the dependence of spin dynamics on the applied external magnetic field, a series of TRKR measurements for the wavelength with maximum KR signal were performed at $T = 10$ K. Fig. 4.21(a) shows TRKR scans measured with and without external magnetic fields. From TRKR signals the dependence of spin dephasing time and Larmor frequency on the applied magnetic field is plotted in Fig. 4.21(b). The spin dephasing time increases with magnetic field up to 1.5 T which is consistent with DP relaxation taking into account magnetic field induced orbital motion of the conduction band electrons [22]. Also, the linear dependence of Larmor frequency on applied magnetic field yields an effective Landé factor of 0.389 ± 0.003 .

4.6 Summary

In conclusion, we have studied the spin dynamics of a two-dimensional electron gas in multilayer QWs by TRKR and RSA. The Dependence of spin dephasing time on the experimental parameters as the magnetic field, pump power, and the pump-probe delay was demonstrated. In the DQW sample, T_2^* extends to 4.4 ns. Additionally, for the TQW sample, T_2^* exceeding 10 ns was observed. The results found are among the longest T_2^* reported for samples of similar doping level [51, 116] and comparable with nominally undoped narrow GaAs QWs [128] and low-density 2DEGs in CdTe QWs [119]. The measured long spin dephasing time was tailored by the control of the QW width, symmetry and electron density. The spin dynamics is dominated by the DP mechanism through the cubic Dresselhaus interaction. All the relevant time scales were determined indicating the importance of each scattering mechanisms in the spin dynamics. We demonstrate that the wavefunction engineering in multilayer QWs may provide practical paths to control the dynamics in spintronic devices.

Chapter 5

Large spin relaxation anisotropy in two-dimensional electron gases hosted in a GaAs/AlGaAs triple quantum wells

In this chapter, we have studied the spin dynamics of a high-mobility dense two-dimensional electron gas confined in a GaAs/AlGaAs triple quantum well by time-resolved Kerr rotation and resonant spin amplification. Strong anisotropic spin relaxation up to a factor of 10 was found between the electron spins oriented in-plane and out-of-plane. We attribute this anisotropy to the presence of an internal magnetic field and the inhomogeneity of the electron g -factor. The data analysis allows us to determine the direction and magnitude of this internal field in the range of a few mT for our studied structure. This magnetic field has been found to decrease with the sample temperature which is further supported by the optical power dependence. The dependence of the anisotropic spin relaxation was directly measured as a function of experimental parameters: pump-probe wavelength, temperature, the time delay between pump and probe and excitation power.

5.1 Motivation

The study of spin dephasing and spin relaxation processes of carriers in two-dimensional electron gas (2DEG) confined in semiconductor quantum wells (QWs) is one of the key requirements for building practical spintronics devices [98, 119]. The spin-orbit

The results presented in this chapter are based on Ref. [4] on page 147

interaction (SOI) have a great impact on the optical or electrical manipulation of carrier spins [20] due to the energy splitting induced by structure inversion asymmetry (Rashba) [42] or bulk inversion asymmetry (Dresselhaus) [43]. Besides an external magnetic field (B_{ext}), the carrier spins will randomly precess around a momentum dependent effective magnetic field opening a channel for spin relaxation via so-called Dyakonov-Perel (DP) mechanism [129].

Investigations of the spin relaxation anisotropy in semiconductor nanostructures have been done in bulk samples [130] as well as in QWs [131–138] grown along different orientations. In zinc-blende heterostructures with equal strength of the Rashba and Dresselhaus spin-orbit fields, an in-plane spin relaxation anisotropy was theoretically predicted [131] and later experimentally confirmed [132]. Using Hanle effect, three different spin relaxation times for the spin oriented along $[110]$, $[1\bar{1}0]$ and $[001]$ directions were extracted. A number of experimental groups have extensively studied the in-plane spin relaxation anisotropy by applying a magnetic field along different crystallographic directions using time-resolved Kerr rotation (TRKR). However, only a few have focused on the anisotropy of the relaxation time for electron spins oriented parallel (τ_z) and perpendicular (τ_y) to the quantum well growth direction. For QWs grown along the $z||[001]$ axis, the spin components of the resident carriers oriented along or normal to the growth axis relax at different rates where, their simultaneous action, leads to strong anisotropy [139]. Later, a similar behavior was experimentally reported for the (110) oriented GaAs QWs using the resonant spin amplification (RSA) technique [101].

It has been observed that the appearance of anisotropic spin relaxation influences the relative amplitudes of the RSA peaks [101, 122, 139]. In the case of isotropic spin relaxation, all peaks have the same height and the spin components of the carriers oriented along z , and y directions relax with the same rate. While during anisotropic spin relaxation the amplitude of the peak corresponding to $B_{ext} = 0$ differs from the peaks at finite field [122, 139].

In this chapter, we report on the experimental observation of large spin relaxation anisotropy in triple quantum wells (Sample B). Similar multiple quantum well systems have allowed the discovery of interesting phenomena, such as intrinsic spin Hall effect [172], collapse of the integer quantum Hall effect [140, 141], long-lived spin coherence [142], spontaneous interlayer phase coherence [143–145], control and drift current induced spin polarization [31, 83], and excitonic Bose condensation [146–148]. Here, we found long relaxation times for the spins oriented along growth direction and fast relaxation times for the spins oriented in-plane. We analyzed how the experimental parameters, sample temperature, and optical pump power, influences the effective in-plane magnetic field and hence the spin relaxation anisotropy in our structure.

5.2 Experimental realization

We used time-resolved pump-probe Kerr rotation (KR) and resonant spin amplification to study the coherent spin dynamics of a two-dimensional electron gas confined in a GaAs/AlGaAs TQW. The light source was a mode-locked Ti-sapphire laser with repetition rate of $f_{rep} = 76$ MHz ($t_{rep} = 13.2$ ns) delivering 100 fs pulses. Spin oriented electrons were optically created by exciting the sample with degenerate pump and time-delayed probe pulses. The time delay Δt between pump and probe pulses was varied by a mechanical delay line. For lock-in detection, the circular polarization of the pump beam was modulated at 50 kHz by a photo-elastic modulator. The probe beam was linearly polarized, and the rotation of its polarization upon reflection was detected with a balanced bridge using coupled photodiodes. The laser energy was tuned to obtain the largest KR signal in the sample. The sample was mounted in a He flow cryostat during measurements, applying transverse magnetic field (Voigt geometry), which forced the spins to precess around the field and allows us to monitor its relaxation time.

5.3 Wavelength dependence of spin dynamics

The TRKR traces measured for different pump-probe wavelengths at pump power of 1 mW, $B = 1$ T and $T = 8$ K are imaged in Fig. 5.1. The vertical lines correspond

FIGURE 5.1: *Pump-probe delay scans of the KR signal measured for different pump-probe wavelengths at $B = 1$ T and $T = 8$ K where the solid line highlights the time evolution of spin dynamics at $\lambda = 821$ nm which is shown on the top of contour plot for clarity.*

to the precession of coherently excited electron spins about the in-plane magnetic field. As reported [119, 142, 149, 150], that long-lived spin coherence can be identified by the carrier precession at negative Δt just before the pump pulse arrival. One can clearly see these negative delay oscillation in the wavelength range from 817 nm to 823 nm, which are more pronounced at higher wavelengths as shown on the top of contour plot for clarity. After the pump pulse positioned at $\Delta t = 0$, the generated spin polarization shows a very weak decay over time window of 2.5 ns.

Another feature directly accessible from the contour plot is the change of precession frequency with wavelength. Increasing the wavelength from 814 nm up to 818 nm leads to an increase in the precession frequency. Further increase in wavelength results in a slow down of the precession, where a sudden change of frequency is clearly visible at 819 nm. The TRKR profile shows two distinct regions, where the precessional frequencies are more pronounced, corresponding to $\lambda = 817$ nm and 820 nm. Which are in agreement with the two distinct lines observed in the magneto-photoluminescence study performed on the same sample [125]. The line corresponding to high wavelength region was attributed to the exciton bound to a neutral donor (DX center), while the one at low wavelength was related to the direct recombination between the states confined in the conduction and valence band.

FIGURE 5.2: *B* scan in the range from -200 mT to +200 mT as a function of different pump-probe wavelengths at $T = 8$ K, and fixed pump-probe delay $\Delta t = -0.24$ ns. The distance between the peaks corresponds approximately to one Larmor precession during the laser repetition interval.

Fig. 5.2 displays the RSA signals measured in the wavelength range, where strong negative delay oscillations were observed. The time delay between pump and probe was adjusted such that the probe pulse arrives 0.24 ns before the subsequent pump pulse, and the Kerr signal was recorded by sweeping the magnetic field over the range of -200 mT to 200 mT. The RSA spectrum consist a series of sharp peaks with a spacing of $\Delta B = (\hbar/(g\mu_B t_{rep}))$ [46] where $\hbar$ is the Planck's constant and t_{rep} is the laser repetition period. Those resonance peaks correspond to the carrier spin precession frequencies which are commensurable with the pump pulse repetition period. Increasing the pump-probe wavelength causes narrowing of the RSA peaks, which reflects prolonging of the spin relaxation time. To get information on spin relaxation times, the resonance peaks were fitted to the Lorentzian model [46, 83]:

$$\Theta_K(B) = A/[(\omega_L \tau_s)^2 + 1] \quad (5.1)$$

with full width at half maximum $B_{1/2} = \hbar/(g\mu_B \tau_s)$, where $\tau_s = [2\tau_y \tau_z / (\tau_y + \tau_z)]$ [122] and $\omega_L = g\mu_B B/\hbar$ is the Larmor precession frequency. τ_z is the carrier spin relaxation in the absence of magnetic field while the side peaks at $B \neq 0$ reveal the spin relaxation time τ_s . The inset Fig. 5.3(a) shows the comparison of four normalized zero-field peaks with fitted Lorentzian curves for several wavelengths. The extracted values of τ_z and amplitude are plotted in Fig. 5.3(a) and (b) as a function of wavelength. Increasing the pump-probe energy about 5.6 meV ($\simeq 2\Delta_{23}$) by decreasing the wavelength from 819 nm up to 816 nm there is almost no effect on the spin relaxation time, and it remains constant around 3 ns. Further increase of wavelength causes an abrupt increase in the dephasing time and yield 18.8 ns for $\lambda = 823$ nm. In contrast, the amplitude of zero-field peak

FIGURE 5.3: (a) The spin dephasing time and (b) Amplitude as a function of pump-probe wavelengths extracted from 5.2. Where the inset of figure (a) shows the Lorentzian fit of the zero-field resonance peak.

reduces sharply with higher wavelengths. We clearly see that for higher wavelengths, the RSA peaks centered around zero magnetic fields are suppressed compared with the neighboring peaks. As this effect is stronger for $\lambda = 823$ nm, so we limited our study to the higher wavelength.

5.4 Effect of inhomogeneous broadening

Fig. 5.4 (a) shows the magnetic field scan of the KR at pump power of 2.5 mW and $\Delta t = -0.24$ ns. For the comprehensive analysis of the influence of magnetic field on the spin relaxation time, every peak was fitted to the Lorentzian model as shown by the solid red lines on the top of experimental curves in panel (a). The extracted values of τ_s are shown in Fig. 5.4 (b) showing a strong reduction with increasing field. The broadening of RSA peaks and the reduction of their amplitude with the higher magnetic field is caused by the inhomogeneity of the g -factor [139].

FIGURE 5.4: (a) B_{ext} scan of KR signal at $\Delta t = -0.24$ ns and $p_{\text{pump}} = 2.5$ mW. The red line is Lorentzian fit from where τ_s is extracted and plotted in (b).

5.5 Temperature influence on spin relaxation anisotropy

We turn now to the effect of sample temperature on the evolution of the carriers spin dynamics. Figure 5.5 shows the RSA traces recorded at different temperature while keeping the time delay between pump and probe fixed at $\Delta t = -0.24$ ns. The magnetic field was scanned in the range from -150 mT to 150 mT while slowly heating up the

FIGURE 5.5: Temperature dependence of RSA signal at $\Delta t = -0.24$ ns. The solid red line is fit to Eq. 5.2.

cryostat, starting from $T = 5$ K up to higher temperatures at which the RSA signal vanishes. The observed RSA pattern is well described by [151]

$$\Theta_K(B) = A \cos \left(\frac{g\mu_B \Delta t}{\hbar} \sqrt{(B_{ext} + B_{\parallel})^2 + B_{\perp}^2} \right) \quad (5.2)$$

where A is KR amplitude, μ_B is the Bohr magneton, g is the electron g-factor, $\hbar$ is the reduced Planck's constant, $B_{\perp}$ and $B_{\parallel}$ are the components of spin-orbit field perpendicular and parallel to the external magnetic field. For clarity of presentation a selected range of RSA pattern, from -50 mT to +50 mT, was fitted to Eq. 5.2. The extracted values of $B_{\parallel}$ was negligible in the studied structure and is omitted here however, the magnitude of $B_{\perp}$ obtained is plotted in Fig. 5.6(a) which decrease with the rise of temperature. In the temperature range $5 \text{ K} < T < 36 \text{ K}$ a good linear dependence on temperature was observed, with linear dependency of $B_{\perp}(T) = -8.695 \times 10^{-5}T + 0.00324$. In order to determine the spin relaxation time and amplitude, the RSA peaks were fitted to Eq. 5.1 as described above. The corresponding values of τ_z and τ_y are depicted in fig 5.6(b). The relaxation times of both spin orientations decrease monotonically with the temperature and are qualitatively similar above 15 K. Anisotropy of relaxation time is stronger at low temperatures which decreases with temperature and reverts to a constant value above 20 K. A possible reason for the decrease of τ with high temperature is the heating of 2DEG.

FIGURE 5.6: (a) Component of B_{SO} perpendicular to applied magnetic field, (b) τ_z (open triangles) and τ_y (solid triangles) extracted from Fig. 5.5 as a function of sample temperature.

The fitted amplitude of zero-field peaks remains constant upto 24 K and then turn to an exponential decay with further increase of temperature (See Fig. 5.7(a)). The temperature dependence of relaxation times show the characteristic Dyakonov-Perel (DP) spin relaxation mechanism [87, 129]. To get information about spin life time, this mechanism

FIGURE 5.7: (a) Amplitude of zero-field peak extracted from Lorentzian fit and (b) corresponding relaxation rate of the y component of spin relaxation time (shown in 5.6(b)) yielding a linear increase with a slope of $0.012 \text{ ns}^{-1} \text{ K}^{-1}$.

was analyzed by Dyakonov and Kachorovskii in the context of QWs [59, 60]:

$$\frac{1}{\tau} = \frac{\alpha^2 E_1^2 K_B T}{\hbar^2 E_g} \tau_p(T) \quad (5.3)$$

Where α is a parameter related to the spin splitting of conduction band, T is the temperature, E_g is the band gap energy, $\tau_p(T)$ is electron momentum relaxation time and E_1 is the first confined state energy of the electron. Due to the occupation of higher momentum states the DP relaxation mechanism become more efficient at elevated temperature as a result the relaxation times drop to ~ 3 ns at 40 K.

5.6 Delay dependence of spin relaxation anisotropy

In this section, we investigate the effect of KR signal at the different pump-probe delay. Fig. 5.8(a) displays the RSA signals measured at various time delay with pump/probe power of 1mW/300 μ W. At different pump-probe delay, the shape of RSA signal differs from each other due to different phases of the spin precession [122]. At certain time delays, the resonances change the pointing direction because the spin resonance

FIGURE 5.8: Pump-probe delay dependence of resonant spin amplification (a) B_{ext} scan of KR signal at $T = 8$ K, for different time delays with pump/probe power of 1 mW/300 μ W. (b) Spin relaxation anisotropy and amplitude dependence on Δt . Inset Fig (b) shows the time evolution of KR measured at 823 nm highlighting the negative time delays (with the same color of the curves) for which RSA traces were recorded.

has precessed before the measurement during the delay time [46]. The spin relaxation anisotropy τ_z/τ_y and amplitude of the peaks corresponding to zero magnetic fields obtained from the fit with Eq. 5.1 are given in Fig. 5.8(b) where the fitted curves (red lines) are plotted on the top of experimental data. The increase of time delay between pump and probe causes the broadening of RSA peaks [142]; as a result, the relaxation time and hence the anisotropy decreases. Inset Fig. 5.8(b) shows the time evolution of KR signal measured at $B = 1$ T while keeping the pump-probe wavelength, power and sample temperature the same. The signal is normalized to its value at zero time delay in order to highlight the negative delay between the pulses. The lines at negative delay correspond to the delay points at which RSA traces were measured where the color of lines matches with the raw data presented in (a). τ_z/τ_y decreases from 6.6 at -0.71 ns down to 1.38 at -0.04 ns following an exponential decay (solid line). However, in contrast, the amplitude of the zero field resonance peak increases exponentially with larger delay.

FIGURE 5.9: KR signal while sweeping B_{ext} at $T = 8$ K for different optical power. The pump-probe delay was fixed at $\Delta t = -0.24$ ns. The data have been shifted for clarity of presentation. The red curves on the top of experimental data are fit to Eq. 5.2.

5.7 Optical power dependence of spin relaxation anisotropy

Next, we report on the optical power influence on the spin relaxation anisotropy. Fig. 5.9 presents the RSA signals measured in a range of pump power from 1 to 7 mW fitted to Eq. 5.2 in the same range of magnetic field commented in section 5.5. The fitting result is displayed in Fig. 5.10. As a function of pump power the $B_{\perp}$ follows a similar decrease like temperature dependence, with linear dependency of $B_{\perp}(T) = -3.2952 \times 10^{-4}P + 0.00347$ (See Fig. 5.10(a)). The RSA peaks are getting broader and shorter in amplitude with increasing magnetic field due to the inhomogeneity of g -factor in the measured ensemble as commented. From the Lorentzian fit to the data, τ_z/τ_y and amplitude were obtained and plotted as function of pump power in Fig. 5.10 (b). Increasing the optical power causes the reduction of spin relaxation anisotropy. This

FIGURE 5.10: (a) Component of B_{SO} perpendicular to the applied magnetic field as a function of pump power. (b) τ_z/τ_y and KR amplitude dependence on the optical power, where, the solid lines are exponential fit to the data.

reduction of τ_z/τ_y with high pump power can be correlated with the effect of heating of 2DEG and QW electron's delocalization due to the interaction with photogenerated carriers [152]. For 2DEG confined in (001) GaAs/AlGaAs heterostructure, a similar decrease of an in-plane spin relaxation anisotropy with high pump power was attributed to the domination of one of the spin-orbit (SO) coupling (Rashba and Dresselhaus) over the other [135]. The observed decrease of spin relaxation time with high pump power is consistent with the previously reported data on the (001) oriented GaAs/AlGaAs double quantum well [142]. The amplitude of the peaks centered at zero-field increase exponentially with high pump power.

5.8 Summary

In conclusion, we performed a detail experimental study of the spin relaxation in a high mobility dense two-dimensional electron gas by TRKR and RSA. A strong anisotropy of the spin relaxation for the in- and out-of-plane spin orientations was observed. The relaxation time for the spin oriented along the growth direction [001] is larger than the relaxation time along the direction normal to it. We relate the origin of this anisotropy to the presence of in-plane SO field and spread of g -factor within the measured ensemble. The degree of anisotropy shows a strong dependence on the temperature and pump-probe delay. The DP spin relaxation mechanism becomes efficient at the higher temperature which causes the reduction of anisotropy.

Chapter 6

Robustness of spin coherence against high temperature in multilayer system

In the present chapter, we address spin dynamics of (001) oriented two-dimensional electron gas confined in GaAs/AlGaAs triple quantum well by using pump-probe Kerr rotation under the transverse magnetic field. In the presence of an applied in-plane magnetic field the TRKR measurements show robustness of the carrier's spin precession against increasing temperature which can be easily traced in an extended range up to 250 K. Long-lasting spin coherence with dephasing time $T_2^ > 14$ ns has recorded at $T = 5$ K for $\lambda = 823$ nm. The spin dephasing time has been found to reduce sharply with temperature and become much shorter of the order 0.27 ns at $T = 250$ K. The observed electron g -factor increases linearly with temperature from -0.452 at 5 K to -0.341 at 250 K for $\lambda = 823$ nm and -0.46 to -0.36 in the same temperature range for $\lambda = 821$ nm. Additionally, the spin dynamics is studied through the dependencies on the applied magnetic field and optical pump power.*

6.1 Motivation

Recently, the spin dynamics of carrier's and related physics in semiconductor quantum wells (QWs) have been attracted considerable attention from both viewpoints of physics, and it's promising applications in spintronics devices [12, 22, 153, 154]. As an essential factor, for successful implementation into the future semiconductor devices and quantum

The results presented in this chapter are based on Ref. [5] on page 147

information processing, the spin dephasing time must be sufficiently long enough to allow the processing of the stored information. A lot of efforts has been put forth to enhance the lifetime of spin-polarized electrons. One of those efforts, made in the bulk [46] and II-VI QW [155] samples with doping level close to metal-insulator transition (MIT), was the observation of an extraordinary long coherence time by Awschalom group. Those findings, on the one hand, revealed that the long-lived spin coherence is restrained to a doping level in the vicinities of MIT [48, 51, 105]. On the other hand, it animated the expectation that the electron spin can be finally realized as a basis for quantum computation. For the realization of device concepts, it is highly desirable that the generation and detection of such spin polarization could be carried out at high temperature and low magnetic field.

There have been a lot of works reporting on the spin dynamics of carriers, in different dimensionally semiconductor structures, like QWs [100, 142], quantum dots (QDs) [156] and layered structures [157] of various material systems based on III-V (e.g., GaAs, GaN, (In,Ga)As) and II-VI (e.g., CdTe, ZnSe, (Zn,Cd)Se) semiconductors. To extend the spin lifetime, one has to control the spin decoherence, i.e., the loss of spin memory in semiconductor nanostructures such as QWs. Therefore, it is of crucial importance to get a comprehensive understanding of the mechanisms of spin relaxation¹ in semiconductor structures. Three major spin relaxation mechanisms in semiconductors have been established to describe the spin dephasing and relaxation dynamics [26]: the Bir-Aronov-Pikus (BAP) mechanism [61], the Elliott-Yafet (EY) mechanism [35, 55] and Dyakonov-Perel (DP) mechanism [57]. However, the spin lifetime undergoing these mechanisms greatly depends on the sample temperature and design as well as the carrier type and density.

The BAP spin relaxation mechanism, caused by spin-flip via the electron-hole exchange interaction, is only significant at lower temperatures and in highly doped p-type or insulating materials. For highly *n*-doped samples, the BAP mechanism can be ruled out from the mechanisms responsible for the spin relaxation because the photogenerated holes quickly recombine with electrons due to the availability of a large number of electrons. The EY mechanism, being based on the fact that the SOC mixes the wave functions of spin-up and spin-down states resulting in a non-zero spin flip due to impurity or phonon scattering, dominates in narrow band gap semiconductors. The DP mechanism originates from the spin-orbit splitting of the conduction band states which acts as an effective magnetic field. Previous literature shows that for GaAs, the DP mechanism is more efficient than EY at elevated temperatures due to large band gap and low scattering rate [158–160].

¹In this thesis the terms spin relaxation, spin dephasing and spin decoherence are used as synonyms

The temperature dependency of the spin dynamics can be carried out to distinguish DP mechanism from the EY mechanism in semiconductor QWs, for example Dyakonov-Perel ($1/\tau \propto \tau_p T$) and Elliott-Yafet ($1/\tau \propto \tau_p^{-1} T$). Despite a considerable number of experimental studies on the temperature influence of spin dynamics in semiconductor QWs, the measurements performed on multilayer structure are still lacking so far [161, 162].

6.2 Experimental realization

We report here the spin dynamics in a GaAs/AlGaAs TQW (sample B) with Si doping concentration beyond metal-insulator transition. The TQW sample studied here consist of a 22-nm-thick GaAs central well and two 10-nm-thick lateral wells sandwiched between AlGaAs layers. The side wells are well separated from the central well by 2-nm-thick $Al_{0.3}Ga_{0.7}As$ barriers. All experiments were performed in an optical cryostat equipped with a superconducting magnet for supplying magnetic field in the range from 0 to ± 8 T. The main measurement techniques were the well known optical pump-probe techniques: i.e. Time-resolved Kerr rotation and resonant spin amplification. The laser beam delivered by Ti: sapphire laser with ~ 100 fs pulses at repetition rate $f_{rep} = 76$ MHz were split by a beam splitter into the pump and probe parts. The polarization of pump pulse was modulated at 50 kHz with a photo-elastic modulator (PEM) for lock-in detection. The circularly polarized pump pulse, incident normal to the sample surface, created spin-polarized electrons with spin vector along the QW growth direction. Varying the time delay Δt between pump and probe pulses the rotation of linearly polarized probe upon reflection was recorded with a balanced bridge using coupled photodiodes. The laser wavelength was tuned for maximum Kerr rotation signal (See sec. 5.3 in chapter 5).

6.3 Experimental results

6.3.1 Magnetic field dependence of coherence time

Kerr rotation of optically generated spin as a function of Δt with no magnetic field and in the transverse magnetic field up to 2 T is imaged in Fig. 6.1. TRKR traces were measured at $T = 8$ K with an excitation wavelength of 821 nm and pump/probe power of 1 mW/300 μ W. The TRKR traces show periodic oscillation in the external transverse magnetic field, denoting the existence of spin signals. These oscillation results from the spin precession in the magnetic field with a beating frequency, i.e., the Larmor frequency,

FIGURE 6.1: *Magnetic field dependence of TRKR traces imaged for $\lambda = 821$ nm at $T = 8$ K. The solid lines highlight the TRKR scan at $B = 0.5$ T and 1.5 T which are shown on the top of the contour plot.*

$\omega_L = g\mu_B B/\hbar$. Where, g is the effective Landé g -factor, μ_B is the Bohr magneton and $\hbar$ is the reduced Planck's constant. The electron g -factor in semiconductors ($g_0 = -0.44$ for bulk GaAs) differs from that of the free electron, 2.0, due to the SOI and varies with the energy in the band. Additionally, the g -factor varies due to band filling and quantum confinement [163] in GaAs layer. This behavior is approximated by $g = g_0 + \gamma E$ where E stands for the excess energy of the electrons (with respect to the bottom of the conduction band) [164]. Increasing the magnitude of applied magnetic field speed up the precessional frequency as evidenced from the TRKR traces at $B = 0.5$ T and 1.5 T shown on the top of the contour plot. Another important feature evidenced from TRKR data is that over the measured range of magnetic field the electron spin beating at positive delays is accompanied by spin beating even at negative delays due to the long-lived spin coherence persisting between successive pulses.

For further analysis, the TRKR traces imaged in Fig. 6.1 were fitted by Eq. 4.2. The Larmor precession frequency and spin dephasing time retrieved from fitting are displayed in Fig. 6.2(a) and (b). The linear dependence of ω_L on magnetic field yield a g -factor (absolute value) of $g = 0.454 \pm 0.001$. Comparing with the g -factor of the bulk it was shown that the observed spin signals belong to electron carriers. The magnetic field dependence of electron spin dephasing time is shown in Figure 6.2(b). The spin dephasing time varies with increasing magnetic field. T_2^* first increases to a maximum

FIGURE 6.2: (a) Larmor precession frequencies (black circles) with a linear fit (solid red line) and (b) the spin dephasing times (red squares) as a function of external magnetic field at $T = 8$ K.

value of ~ 12.7 ns at $B = 0.4$ T and then decreases with further increase of magnetic field due to the spread in ensemble g -factor (see Sec. 5.4 for more details), resulting in an inhomogeneous dephasing of carriers spin under magnetic field. The reduction of T_2^* with magnetic field don't follow $1/B$ dependence, suggesting that the spread of ensemble g -factor, Δg , is not the main mechanism of spin relaxation. The size of inhomogeneity can be inferred from the linear dependency of spin relaxation rate on the magnetic field, $1/T_2^*(B) = 1/T_2^*(0) + \Delta g \mu_B B / 2\hbar$ [46, 103]. The spread of g -factor retrieved from the fitting is $\Delta g = 0.007$, which is only 0.11 % of the observed g -factor.

6.3.2 Spin dynamics dependence on the sample temperature with $\lambda = 821$ nm

The electron spin coherence in our GaAs/AlGaAs TQW sample is robust against increasing temperature and is clearly observed in a wide range of temperature up to 250 K. A representative selection of typical TRKR traces measured for various temperature in the range from 5 K up to 250 K are shown in Fig. 6.3 for $\lambda = 821$ nm with pump/probe power of 1 mW/300 μ W and $B = 1$ T. Background has been added to each scan and the curves at $T = 140$ K and 250 K are upscaled by multiplying with indicated number for clarity of presentation. One can clearly see significant changes in the precession of the photo-excited electron spins about the applied field with temperature. The precession frequency is slowing down with increasing temperature, and the decay of spin beats is thermally stimulated. To get the information over the spin dephasing time and electron g -factor the oscillatory part (after $\Delta t = 0$ ns) of TRKR data were

FIGURE 6.3: Dependence of KR signal on sample temperature at magnetic field $B = 1$ T. The range of temperature varies from 5 K to 250 K. The TRKR traces are vertically shifted and the top two curves are multiplied by indicated factors for clarity of presentation. $\lambda = 821$ nm, $P_{\text{pump}} = 1$ mW and $P_{\text{probe}} = 300 \mu\text{W}$.

FIGURE 6.4: Temperature dependence of resonant spin amplification at excitation wavelength 821 nm, and fixed pump-probe delay $\Delta t = -0.24$ ns. Background is added to each scan for clarity.

fitted by an exponentially damped harmonic Eq. 4.2. One can clearly see in Fig. 6.3 that at low temperatures in the range from 5 K to 35 K the signal decay is very slow and the electron spin dephasing is not completed during the repetition period of the pump pulses ($t_{rep} = 13.2$ ns) as a result we see strong negative delay oscillations. In such cases, the well-known technique of the resonant spin amplification [46] can be used to extract the spin dephasing time.

Fig. 6.4 display multiple RSA peaks at a fixed negative time delay of $\Delta t = -0.24$ ns in the magnetic field sweeping over a range of -200 mT to 200 mT. The excitation energy was kept the same of the TRKR traces in Fig. 6.3. The RSA spectrum strongly differs from each other with increasing temperature. First in the temperature range from 5 K to 20 K the RSA peak centered at $B = 0$ T is shorter in amplitude than the peaks at the finite magnetic fields. The depression of the zero field resonant peak in respect to the neighboring peaks is due to the spin relaxation anisotropy [122, 139] caused by internal magnetic field as discussed in chapter 5. The direction and magnitude of this internal field can be obtained by fitting the data to Eq. 5.2 as shown in Fig. 6.4, by red curves on the top of experimental data in a selective range from -100 to 100 mT, for $T = 15$ K. The fitting yields $B_{\perp} = 0.0017$ mT which causes spin relaxation in our structure through Dyakonov-Perel mechanism.

Second, the data support a transition from anisotropic to isotropic spin relaxation with growing temperature i.e. with the increase of sample temperature the amplitude of the peak centered at $B = 0$ T increases and become equal in amplitude to that of finite

FIGURE 6.5: (a) Electron g -factor and (b) the spin dephasing time as a function of temperature. The solid line in panel (a) is a linear fit for data. In (b) T_2^* extracted from the RSA signal (the solid circles) and the data marked by open circles are evaluated from the fit of Kerr rotation signal presented in Fig. 6.3. The error bars represent the standard error.

FIGURE 6.6: Dependence of the spin dephasing rate on the sample temperature at $B = 1$ T. Solid line is a linear fit with a slope of $0.011 \text{ ns}^{-1} \text{ K}^{-1}$ to the data.

field peaks at a higher temperature. Third, the amplitude of the finite field RSA peaks decreases with increasing magnetic field due to the ensemble spread of electron g -factor. Fourth, at low temperature (i.e. $T = 5$ K) the resonant peaks have larger amplitude at higher magnetic field indicating long hole spin coherence time involved in the generation of spin coherence time [165].

T_2^* obtained from Lorentzian fit to the zero-field peak are depicted in Fig. 6.5(b) by closed circles together with the data points extracted from TRKR (open circles). The TRKR signal recorded at $T = 250$ K showed a biphasic spin dynamics and was fitted to Eq. 4.2 plus a non-oscillatory exponential decay to account the fast decay over first few picoseconds. The fitting yields decay times with $T_{21}^* = 8.5$ ps and a relatively long $T_{22}^* = 0.357$ ns (T_{21}^* is related to the hole spin dynamics and T_{22}^* corresponds to electron spin dynamics). Fig. 6.5(a) shows the electron g -factor as a function of temperature retrieved from TRKR data. In the studied temperature range, $5 \text{ K} < T < 250 \text{ K}$, the g -factor increases from -0.46 to -0.36. For the temperature up to 160 K, a good linear dependence on temperature was observed, with linear dependency of $g(T) = -0.452 + 5.37 \times 10^{-4}T$. Our finding is in good agreement with a similar investigation of the temperature dependence of electron g -factor reported on bulk GaAs [37, 166]. In Ref. [37] the experimentally observed g -factor was approximated by $g(T) = -0.44 + 5.0 \times 10^{-4}T$ for a temperature ranged from liquid helium temperature up to room temperature.

We observed an obvious decrease in the spin dephasing time with rising temperature. T_2^* decreases from 11 ns to 0.36 ns with the increase of temperature from 5 K to 250 K. With temperature rise the 2DEG localized electrons get delocalized which results

in the decrease of electrons spin dephasing time. To identify the possible mechanism responsible for spin relaxation in our structure the spin dephasing rate ($1/T_2^*$), extracted from TRKR and RSA, was plotted, in Fig. 6.6, as a function of sample temperature. The spin relaxation rate as a function of sample temperature follows a linear dependency with a slope of $0.011 \text{ ns}^{-1} \text{ K}^{-1}$. As both EY and DP mechanism follows the observed dependence $1/T_2^* \propto T$. We don't have data on τ_p in the studied structure, but according to the previous literature [59,107,108,131,167–169] it is well reported that in QWs based on GaAs materials the spin relaxation is dominated by the DP mechanism. Therefore, we believe that the DP mechanism is the dominant mechanism responsible for the spin relaxation in our structure.

6.3.3 Temperature dependence of spin dynamics for $\lambda = 823 \text{ nm}$

The wavelength dependence of spin dynamics measured on the same sample demonstrated in Fig. 5.1 chapter 5 showed two distinct regions where the electron spin precession were more prominent. Additionally, in the region corresponding to higher wavelengths, $817 < \lambda < 823 \text{ nm}$, a long-lived spin component was traced at longer delays of about $\sim 13.2 \text{ ns}$ which was more stronger at higher wavelength. Because of long lasting spin dephasing time observed at $\lambda = 823 \text{ nm}$ the temperature influence on spin

FIGURE 6.7: Transient Kerr rotation signals under transverse magnetic field of $B = 1 \text{ T}$ for various temperatures in the range from 5 K to 250 K . The data have been shifted for clarity of presentation. $\lambda = 823 \text{ nm}$, $P_{\text{pump}} = 1 \text{ mW}$ and $P_{\text{probe}} = 300 \mu\text{W}$.

FIGURE 6.8: *RSA signals measured by degenerate pump-probe Kerr rotation by scanning magnetic field in the range from -200 mT up to 200 mT for various temperatures at $\Delta t = -0.24$ ns. The vertical lines correspond to the resonance peaks at certain magnetic fields. $\lambda = 823$ nm.*

dynamics was performed keeping this wavelength for the following discussions. Figure 6.7 shows typical Kerr rotation signals as a function of time delay between pump and probe at different temperatures up to 250 K. The laser wavelength was tuned to 823 nm for maximum KR signal at $B = 1$ T and pump/probe power of 1 mW/300 μ W.

The transverse magnetic field causes the spin precession which decays with the growth of temperature. The change in precession frequency is clearly visible, i.e., the precession frequency is getting smaller with growing temperature. The change in precession frequency directly affect the electron g -factor according to $\omega_L = g\mu_B B/\hbar$. Where μ_B is Bohr magneton and $\hbar$ is reduced planks constant. As exhibited in Fig. 6.7, all the data except the one at elevated temperatures show a single exponential decay. However, for the present study, only the spin relaxation of electron gas is concerned, while the short time spin relaxation (such as hole spin polarization) is disregarded. One can clearly see that, at low temperatures, the signal lasting more than the period between two subsequent laser pulses (13.2 ns). Which results in substantial signal oscillation at negative time delay (just before pump pulse arrival) of the amplitude comparable to the one at the positive delay, see, for example, the traces at $T = 5$ K and 12 K. The oscillation at positive time delay were fitted according to Eq. 4.2.

The pronounced oscillation at negative time delay for the temperature range, $5 \text{ K} < T < 36 \text{ K}$, suggesting that $T_2^* \geq t_{rep}$. In such conditions the direct determination of spin

dephasing time by fitting the envelope of TRKR signal becomes inapplicable; Thus the well known RSA technique [46], which takes into account the constructive interference of the coherent spin oscillations from successive pulses can be used. In Fig. 6.8 such an RSA measurement is shown. The delay between pump and probe was set to $\Delta t = -240$ ps while the magnetic field was scanned from -200 mT to 200 mT. The superposition of spins that were created by the pulse train 13.2 ns before the arrival of the next pulse causes a series of sharp resonance peaks. The rise of temperature speed up the decay of spin polarization due to the heating effect and the RSA peaks disappear into noise (white shades) at higher temperature see for example the peaks at temperature $T > 40$ K. The color map shows that at low temperature the zero-field peak is weaker compared to the finite field peaks, indicating a change of the out-of-plane spin dephasing time. Also in Fig. 6.8, the change of electron g -factor with temperature is clearly visible from the variation of the spacing, ΔB , between RSA peaks. That is with growing temperature the outer peaks are shifting toward higher magnetic fields. The spin dephasing time was calculated by a comparison of the experimental data to the model in Eq. 4.3.

The electron g -factor extracted from the TRKR oscillation is shown in Fig. 6.9(a). In the observed temperature range the electron g -factor increases from -0.452 to -0.341. Again, the present findings are in agreement with the literature results [37, 166] i.e., at temperature below 140 K, the data follows linear dependence according to the equation $g(T) = -0.446 + 4.87 \times 10^{-4}T$. Not only the absolute value of g -factor, but also the spin dephasing time decreases with growing temperature. The temperature dependence of T_2^* extracted from TRKR (open circles) and the RSA peaks at $B = 0$ T (solid circles) are shown in Fig. 6.9(b). In the low-temperature region, the spin dephasing time shows

FIGURE 6.9: (a) Electron g -factor versus temperature with linear fit to the data (solid line) and (b) the spin dephasing time obtained from RSA peak at $B = 0$ T (solid circles) and the dephasing time marked by open circles evaluated from the oscillatory part of the TRKR data shown in Fig. 6.7 as a function of temperature.

a sharp reduction in spin dephasing time. A further increase of the temperature leads to a slow decrease of T_2^* . The spin dephasing time, which is around 14.8 ns at $T = 5$ K, reduces to 0.27 ns at $T = 250$ K due to the efficient DP spin relaxation mechanism and ensemble spread of g -factor. The spin dephasing rate follows a linear dependence, $1/T_2^* \propto T$, with a slope of $0.008 \text{ ns}^{-1} \text{ K}^{-1}$.

6.3.4 Optical power influence on spin dynamics

In this section, the impact of excitation power on the spin dynamics of electrons has been investigated in sample B using time-resolved Kerr rotation. Fig. 6.10 shows the pump-probe delay scans of the KR signal measured with a magnetic field applied normal to the QW growth direction by varying excitation power from 0.5 to 7 mW at $T = 8$ K. The striking feature of the KR traces is the appearance of long-lived spin beating as can be seen at negative time delay. The vertical lines represent the spin precession of coherently excited electrons about the applied magnetic field. One can clearly observe that the increase of optical pump power have no effect on the frequency of spin beats.

FIGURE 6.10: *TRKR traces recorded on sample B with different optical pump-power at $B = 1$ T and $T = 8$ K. The TRKR scan shown on the top of contour plot highlights the KR signal at pump power of 5 mW as shown by a line with same color coding.*

FIGURE 6.11: (a) The electron g -factor and (b) the spin dephasing time extracted from Fig. 6.10 as a function of optical pump power. The error bars correspond to the fitting standard errors. $B = 1$ T and $T = 8$ K.

The electron g -factor and spin dephasing time evaluated from the fit of experimental data are shown in Fig. 6.11(a) and (b). The dependence of electron g -factor on pump-power reveal that there is no influence of the optical pump-power on the g -factor. In the low power range up to 3.5 mW we don't observe any change in T_2^* with increasing power and remains constant at ~ 7.78 ns. Further increase of pump-power causes the reduction in T_2^* possibly due to heating effect by optical excitation [119] but still remain in the nanosecond range near ~ 4.54 ns.

6.4 Summary

In this chapter, we presented the electron spin dynamics in GaAs/AlGaAs triple quantum well using pump-probe Kerr rotation technique. The dependence of spin relaxation time on the applied magnetic field, sample temperature, and optical power was studied.

It has been found that the spin precession in our sample is robust against temperature rise and can be easily traced up to $T = 250$ K. Where this precession allows to determine the electron g -factor. The electron g -factor was found to increase linearly with rising of temperature.

To get the longest spin dephasing time in our sample, we have to use the lowest possible magnetic field. For that, we used the modified form of pump-probe Kerr rotation technique known as Resonant spin amplification. The longest spin dephasing time of 14.8 ns was observed at $T = 5$ K for $\lambda = 823$ nm.

Additionally, in our dense, high mobility 2DEG sample an interesting behavior in the RSA was observed at low temperature. The signal is quite different from the typical RSA shape [46] where the amplitude of the peaks increases with the magnetic field indicating a healthy interdependence of electron and hole spin dynamics.

Chapter 7

Effect of Al concentration on the spin dynamics of $Al_xGa_{1-x}As/AlAs$ single and double quantum wells

In this chapter, we present a detailed experimental study to investigate the spin dynamics of the 2DEG confined in $Al_xGa_{1-x}As/AlAs$ single and double quantum wells. Pump-probe reflection technique was used to characterize that how the spin dephasing time and electron g -factor changes with the Al content in each well under different experimental conditions like the applied magnetic field, pump power and sample temperature. In sample D and F we observed the spin signal with two contributions from different electron populations. The observed contributions to the signal oscillating with different frequencies were identified by extracting the corresponding g -factors. From the analysis of the data, we conclude that the Al composition in each well allows tuning the spin dephasing time and electron g -factor.

7.1 Experimental realization

We used the pump-probe Kerr rotation technique, a well-established tool [170], to study the electron spin dynamics. The electron spin polarization was generated by circularly polarized pump pulses emitted by a mode-locked Ti: Sapphire laser with pulse duration of 100 fs, operating at a repetition frequency of 76 MHz (repetition period $t_{rep} = 13.2$ ns). The induced spin coherence was measured by a relatively weak linearly polarized probe pulses of the same photon energy as the pump pulses (degenerate pump-probe system). A computer controlled mechanical delay line was used to vary the time delay between the probe and pump pulses. The laser wavelength was tuned in each sample

for maximum Kerr signal with long coherence time. The pump beam was modulated by a photo-elastic modulator (PEM) operated at 50 kHz for lock-in detection. The Kerr rotation (KR) angle of the initially linearly polarized probe pulses upon the reflection from the sample surface was measured by a balanced bridge.

The spin dynamics was investigated here with experimental results on four different samples one single and three double quantum wells named sample D, E, F, and G respectively. All the samples are remotely delta-doped $Al_xGa_{1-x}As$ quantum wells, containing a dense two-dimensional electron gas (2DEG). The samples were grown by molecular beam epitaxy with different Al content ($8.2\% < x < 16\%$) in each individual well. For more details on the sample structure, quantum well width, barrier width, mobility and electron density the readers are referred to subsection 3.3.

7.2 Spin dynamics in sample D

In the current section, we concentrate on the spin dynamics of the single quantum well structure (sample D) with $x = 10\%$ Al content in the well. In this sample, the laser energy has direct access to the electrons belonging to two different populations. Under applied magnetic field, the oscillatory signal allows us to extract the g -factor which in turn allow a tool to identify the type of carriers and differentiate the signals belonging

FIGURE 7.1: KR vs time delay between pump and probe for different excitation wavelength measured on sample D. The powers were set to 5 and 1.3 mW for the pump and probe respectively. $B = 1$ T and $T = 5$ K.

to different electron populations in the sample. The same approach has been used in Ref. [171] to distinguish the photo-excited electrons and hole.

7.2.1 Dependence of spin dynamics on excitation energy

For the selection of the right excitation energy, the TRKR dependence on the laser wavelength was measured. A series of TRKR traces were measured for different excitation energy ranging from $\lambda = 726$ nm to 806 nm, with an excitation power of 5 mW under the applied magnetic field of $B = 1$ T and sample temperature $T = 5$ K (see Fig. 7.1). There is almost no effect of the excitation wavelength on the spin beats frequency. We found the TRKR trace with the maximum signal at $\lambda = 731$ nm. Additionally, the signal shows an unusual behavior with two contributions resulting from different electron population, each oscillating with its frequency and decay time.

The short living component contributes to the signal at very short pump-probe delays and oscillates in the initial few of tens picoseconds while the persistence signal oscillates with the same frequency of the signals recorded at other wavelengths. To have an idea of the spin dephasing time and the oscillation frequency of each component a TRKR scan with a pump power of 20 mW while keeping the magnetic field, wavelength and temperature the same as of Fig. 7.1 was recorded as shown in Fig. 7.2. The total spin

FIGURE 7.2: Decomposition of TRKR signal recorded at $B = 1$ T, $\lambda = 731$ nm and $T = 5$ K. The top curve is the measured signal and the bottom traces are the components obtained from decomposition. The red curve plotted on the top of experimental trace corresponds to exponentially damped cosine function displayed in Eq. 7.1.

signal is well described by a superposition of two exponentially decaying cosine functions

$$\Theta_K = \sum_{i=1}^2 A_i \exp\left(\frac{-\Delta t}{T_{2,i}^*}\right) \cos\left(\frac{|g_i|\mu_B B}{\hbar} \Delta t + \varphi_i\right) + y_0 \quad (7.1)$$

where A_i is the initial amplitude, μ_B is the Bohr magneton, B is the external magnetic field, $\hbar$ is the reduced Planck's constant, $|g_i|$ is the electron factor, φ_i is the initial phase, y_0 is the offset and $T_{2,i}^*$ is the ensemble dephasing time. The overall spin polarization, plotted by the cyan color, represents the measured transient of Kerr rotation signal where the fitted curve shown by red color is plotted on the top of the experimental trace. The extracted components from the decomposition of experimental data are shown by orange and wine colors. The fit extract the spin relaxation times of 0.513 ± 0.01 ns and 0.116 ± 0.001 ns for components 1 and 2 respectively. Additionally, the data yields two frequencies $38.35 \pm .042$ GHz and 13.68 ± 0.17 GHz corresponding to effective Landé factors (absolute values) of 0.434 ± 0.0005 and 0.155 ± 0.01 for components 1 and 2 respectively.

FIGURE 7.3: KR transients recorded for sample D at different magnetic fields. The traces are shifted vertically for clarity of presentation. The red lines plotted on the top of experimental data are fits to Eq. 7.1. $T = 5$ K, $P_{pump} = 5$ mW and $P_{probe} = 1.3$ mW.

7.2.2 Dependence of spin dynamics on external magnetic field

To give a deeper insight into the signals obtained in the TRKR trace shown in Fig. 7.2 and identify the nature of oscillation the time evolution of KR signal was recorded as a function of external magnetic field. Figure 7.3 shows a set of TRKR traces measured for different magnetic fields in the range from 1 to 5 T at a pump power of 5 mW while keeping the experimental conditions ($T = 5$ K and $\lambda = 731$ nm) the same as were in the previous section. For all the applied magnetic field two oscillatory signals were observed. One can clearly see that the spin beat frequency speed up with the growing magnetic field. Furthermore, at $B = 1$ T the short living spin component oscillate with a very small frequency of 2 GHz. To extract the ensemble dephasing time and electron g -factor the experimental traces were fitted to Eq. 7.1.

The resulted magnetic field dependence of Larmor precession frequency and spin dephasing time for both components are displayed in Fig. 7.4. The electron $|g|$ factors evaluated from the linear dependence of Larmor precession frequencies on the magnetic field are 0.439 ± 0.003 GHz and 0.151 ± 0.003 GHz. The observed g -factors help us to understand the physics underlying the Kerr signal from two different populations. The g -factors of component 1 is in good agreement with the g -factor of the bulk ($|g| = 0.44$) which indicates that the signal of component 1 corresponds to the electron population in the bulk while the g -factor of component 2 is attributed to the signal from electron population in the quantum well. The spin dephasing time of the signal from the bulk shows an obvious decrease with increasing magnetic field while for the signal from QW we do not observe any change in T_2^* with a magnetic field.

FIGURE 7.4: Magnetic field dependence of (a) Larmor precession frequency and (b) the ensemble dephasing time. The solid red lines are linear fit to the data. The size of error bars show the uncertainty in the measured values.

7.2.3 Dependence of spin dynamics on sample temperature

In section 7.2.2 two contributions to the spin signal were observed in a series of TRKR data taken under various magnetic. To explore whether these contributions are robust against temperature, a set of TRKR scan were recorded as a function of sample temperature over the range of 5 to 250 K as shown in Fig. 7.5. The TRKR traces are vertically shifted and normalized to the time origin ($\Delta t = 0$ ns) for clarity. From Fig. 7.5 the following significant features can be directly extracted. First, in the temperature range, $5 \text{ K} < T < 190 \text{ K}$, both signals (bulk and QWs) are contributing to the total spin polarization. Second, the spin precession frequency changes with the rise of temperature as shown by the dashed line. Third, compared to the bulk, the signal from the quantum well is robust against temperature and is traced up to 250 K while the bulk signal is getting weaker with the rising temperature and disappears above 190 K.

Fit to the data shown in Fig. 7.5 yields the electron g -factors and spin dephasing times, which are displayed in Fig. 7.6 as a function of temperature. One can clearly see from this dependence, that there is no influence of the sample temperature on the g -factor of QW. However, the bulk g -factor increases with temperature and follows a linear dependence with, $g(T) = -0.438 + 4.76 \times 10^{-4}T$, which is in complete agreement

FIGURE 7.5: TRKR signals recorded for sample D as a function of lattice temperature in the range from 5 K up to 250 K. The symbols are experimental data and the solid lines are fit to Eq. 7.1. $B = 1 \text{ T}$, $P_{\text{pump}} = 20.2 \text{ mW}$ and $P_{\text{probe}} = 1.3 \text{ mW}$.

FIGURE 7.6: (a) The electron g -factor and (b) the ensemble dephasing time extracted from Fig. 7.5 as a function of sample temperature. The plotted error bars are shorter than the symbol size.

with previously published data on bulk GaAs [37]. The evaluated spin dephasing times as depicted in Fig. 7.6(b) shows pronounced temperature dependence. T_2^* for both the QW and bulk signals first increases with the temperature reaching to its maximum value and then start to decrease with a further rise of temperature, however, the reduction is much strong in the bulk and completely disappears after 190 K.

7.2.4 Dependence of spin dynamics on optical pump power

In section 7.2.3 was shown the influence of sample temperature on Kerr signal, where, the signal from the quantum well was robust against temperature and was traced up to 250 K. However, the signal from bulk didn't last longer and was observed only up to 190 K. After a certain limit of temperature, both the signals showed reduction with temperature possibly due to heating effect. In order to confirm the reduction of spin dephasing time due to heating effect, the TRKR traces were measured as a function of excitation power in a wide range from 1 mW up to 73 mW as shown in Fig. 7.7(b). With increasing pump power the bulk signal is getting weaker and disappears at 73 mW as evidenced from TRKR traces shown in Fig. 7.7(a) for low and high pump power. The spin dephasing time extracted from (b) for both the bulk and quantum well signal are shown in 7.7(c). T_2^* received from the QW signal increases linearly from 36 ps at 1 mW up to 129 ps at 73 mW. However, T_2^* retrieved from the bulk signal decreases exponentially as shown by the solid red curve. For single QW structure, a similar decrease was assigned to the heating effect induced by optical excitation [119]. The observed behavior of T_2^* with increasing pump power is in good agreement with the

FIGURE 7.7: Pump power influence on spin dynamics of sample D: (b) TRKR signals as a function of excitation power in the range from 1 mW up to 73 mW. The horizontal lines on the top of contour plot highlight the TRKR signals at low and high pump power as shown in the top panel (a) with the same color coding. (c) The evaluated (d) spin dephasing times and (d) electron g -factors as a function of excitation power. $T = 5$ K, $B = 1$ T and $\lambda = 731$ nm.

previously reported data on a wide GaAs quantum well [142]. The resulting pump power dependence of g -factor are shown in Fig (d). From this dependence, it is clear that there is no influence of excitation pump-power on the g -factor (absolute value) evaluated from the QW signal. However, the g -factor obtained from bulk signal decreases up to 30 mW and then saturate with the further rise of pump-power.

7.3 Spin dynamics in sample E

Now we turn to the DQW structure, where both QW are separated by a narrow barrier of 1.4 nm and both have an equal Al contents ($x = 10$ %). The spin dynamics in this structure was studied by tuning to the energy, corresponding to a maximum (both in negative and positive direction), in the Kerr rotation amplitude dependent on excitation wavelength at $B = 0$ T [172]. These amplitude maxima were observed at 726 and 731 nm respectively. Due to the narrower barrier, the structure behaves like coupled quantum wells and the photoluminescence (PL) results in a single peak at 730.6 nm (not shown here).

7.3.1 Magnetic field dependence of spin dynamics

The TRKR data for electron spin dynamics is shown in figure 7.8 without external magnetic field and with applied magnetic field magnitudes in the range $B = (1-6)$ T. The data discussed in this section, are recorded in sample E using laser excitation wavelength 726 nm while the sample is cooled down to liquid He temperature. Here we do not observe the signal from the electron populations in bulk. The experimental curves are fitted by a mono-exponential decay for $B = 0$ and by exponentially decaying cosine functions for $B \neq 0$. The fitted curves are presented by solid lines. The evaluated Larmor frequencies (in the case of $B \neq 0$) and decay times are shown in Fig. 7.8 (b) and (c). A Linear fit to Larmor frequency reveals a g-factor of 0.181 ± 0.003 . The extracted spin dephasing time increases with the magnetic field and follows the same behavior as was for the SQW shown in 7.4.

FIGURE 7.8: Time-resolved Kerr rotation on sample E: (a) KR as a function of pump-probe delay, Δt , under the various magnetic field. Experimental data is shown by colors (symbols and line) and fit to the data is shown by a solid red line. The measurement parameters are listed inside panel (a). Magnetic field dependence of (b) Larmor frequency and (c) spin dephasing time.

Fig. 7.9 shows the dependence of TRKR signal on the applied magnetic field recorded in sample E. The laser energy was tuned to the wavelength corresponding to the positive maximum observed in the KR amplitude dependence on laser detuning at $B = 0$. The same wavelength corresponds to peak observed in PL data on the same sample. The TRKR response with and without external field is shown in panel (a). At this energy, we also didn't observe the signal from the bulk. The dependence of spin beat frequency on the applied field is depicted in Fig. 7.9(b) which yield exactly the same electron

FIGURE 7.9: Fits to the Kerr rotation of optically induced spin polarization for various magnetic fields. The data was recorded with experimental parameters given inside the panel. The evaluated electron (b) spin beats frequency and (c) the spin dephasing times as a function of applied field.

g -factor as reported for $\lambda = 726$ nm in Fig. 7.8(b). The spin dephasing time, on the other hand, follow the same dependence on the magnetic field like in the case of the previous section.

7.4 Spin dynamics in sample F

Now we turn to the double quantum well structure, where the QW next to the substrate (QW₁) have $x = 8.2$ % Al content and the nearest to the surface (QW₂) have $x = 14.2$ % Al content. The spin dynamics in this structure was studied by tuning to each quantum well separately.

7.4.1 Magnetic field dependence of spin dynamics in QW₁

In this section, we performed the TRKR measurements when the laser energy was tuned to QW₁ of the DQW sample (sample F). The spin precession was measured under an external magnetic field applied in the Voigt geometry. Fig. 7.10 shows the typical TRKR traces for various magnetic field together with a fit to Eq. 7.1. The salient feature of the data is that again we have two contributions from different electron populations,

FIGURE 7.10: Typical oscillations of the KR signal under external magnetic field together with a fit to the data (solid red curves). The magnetic field was varied in the range from 0 up to 6 T. $T = 4$ K.

to the total spin polarization, oscillating with different frequencies. In order to identify these contributions, again we will refer to the useful information retrieved from the experimental data as shown in Fig. 7.11.

The Larmor precession frequencies, for both contributions, vary linearly with applied magnetic field which is typical for electrons. However, for holes, it may show nonlinearities due to band mixing as reported in $In_xGa_{1-x}As/GaAs$ quantum wells [173]. The linear fit to these data yields the g -factors (absolute values) as labeled next to the corresponding data inside the panel. From the evaluated data it is clearly evidenced, that the signal with g -factor 0.438 ± 0.003 corresponds to the electron population from the bulk while the signal with small g -factor corresponds to the QW.

The spin dephasing times of electrons from both populations show a different behavior as shown in Fig. 7.11. The extracted T_2^* for the QW signal follow a linear increase (in few ps range) with growing magnetic field, however, the evaluated T_2^* for the bulk signal shows a strong reduction with applied field due to the spread of g -factor in the measured ensemble [119,120]. The size of inhomogeneity, Δg , can be obtained by fitting the data according to Eq. 2.70 as shown by the solid red line in Fig. 7.11. $\Delta g = 0.009$

FIGURE 7.11: (a) Larmor precession frequency and (b) the electron spin dephasing time dependence on the external magnetic field. The solid lines in panel (a) are linear fit to the data and in (b) is fit to the data according to Eq. 2.70.

was retrieved from the $1/B$ dependence [46, 103] which is only 2.05 % of the observed g value.

FIGURE 7.12: KR signal as a function pump-probe delay measured for the DQW (sample F) by tuning the laser wavelength to QW_2 in a magnetic field up to 6 T. The colored lines are experimental data while the red curves plotted on the top is a fit to the mono-exponential decaying cosine function. $T = 4$ K.

7.4.2 Magnetic field dependence of spin dynamics in QW₂

Figure 7.12 shows the TRKR signal recorded with and without applied magnetic field for sample F. The laser wavelength was tuned to QW₂ at liquid helium temperature. We do not observe here the signal from the bulk. All the curves show a single exponential decay and were fitted to a monoexponential decaying cosine function displayed in Eq. 3.31. Figure 7.13 shows the Larmor precession frequency as well as the spin dephasing time extracted from KR transients recorded at $B = (0-6)$ T. The Larmor frequency follows the linear dependence, $\omega_L = g\mu_B B/\hbar$, with $g = 0.315 \pm 0.001$. Again we did not see any influence of the external magnetic field on the spin dephasing time.

FIGURE 7.13: KR signal as a function pump-probe delay measured for the DQW (sample F) by tuning the laser wavelength to QW₂ in a magnetic field up to 6 T. The colored lines are experimental data while the red curves plotted on the top are a fit to the mono-exponential decaying cosine function. $T = 4$ K.

7.5 Spin dynamics in sample G

In this section, the spin dynamics in the DQW with a relatively thicker barrier of 5 nm between the wells, will be discussed. The sample has the same structure as of sample E but different Al contents in each well. QW₁ and QW₂ have $x = 11$ % and 16 % of Al contents respectively. The spin dynamics in this structure was studied as a function various applied magnetic field by tuning the laser energy to each quantum well separately.

7.5.1 Magnetic field dependence of spin dynamics in QW₁

Magnetic field dependence of the Kerr rotation signals, when laser was tuned to the quantum well with $x = 11\%$ of Al contents, is shown in Fig. 7.14(a). The application of magnetic field allows the observation of Larmor precession which speeds up with the growing magnetic field. One can clearly see that the amplitude of spin polarization degrades with increasing magnetic field. The evaluated values of Larmor frequency and spin dephasing times, in QW₁, are displayed in Fig. 7.14(b) and (c) as a function of external magnetic field. The dependence of frequency on magnetic field results $g = 0.153 \pm 0.005$. The spin dephasing time increases from 79.6 ps up to 151 ps by increasing the magnetic field from 0 to 6 T.

FIGURE 7.14: (a) Magnetic field dependence of Kerr rotation for sample G at $T = 4$ K. The curves are vertically offset for clarity. The data was recorded with experimental parameters given inside the panel. The magnetic field dependent of (b) the Larmor precession frequency and (c) the spin dephasing times.

7.5.2 Magnetic field dependence of spin dynamics in QW₂

Fig. 7.15 shows the magnetic field dependence of the Kerr rotation signal recorded by tuning laser wavelength in QW₂. On comparison with the signal from QW₁, one can see that the spin beat frequency is enhanced by increasing Al contents in QW₂. The resulting B dependence of the Larmor precession frequency and spin dephasing times are shown in Fig 15(a) and (b) respectively. The electron g-factor evaluated from the

FIGURE 7.15: *KR signal recorded for sample G at different magnetic fields. The red lines plotted on the top of experimental data are fits to Eq. 3.31.*

frequency of KR signal is 0.291 ± 0.001 . However, the applied magnetic field do not affect the spin relaxation time as shown in panel (b).

FIGURE 7.16: *The extracted values of (a) the Larmor precession frequency and (b) the ensemble dephasing time as a function of magnetic field. The solid red line in (a) is a linear fit to the data and in (b) is a guide to the eyes.*

FIGURE 7.17: The dependencies of the electron (a) g -factor and (b) spin dephasing time on the percentage of Al concentration inside each quantum well. The error bars depict the standard deviations.

7.6 Summary

In conclusion, we have studied the influence of Al contents on the spin dynamics of $Al_xGa_{1-x}As/AlAs$ single and double quantum wells. The spin beats frequency under applied magnetic field allows evaluating the electron g -factor which further helps to identify the signal from different electron populations. Compared to the bulk the contribution from the quantum well, to the total spin polarization, is robust against temperature in the single quantum well structure. The spin dephasing times and electron g -factors retrieved from all the samples are summarized in Fig. 7.17. One can clearly see that changing the Al contents inside the well can allow tuning of the g -factor as well as the spin dephasing time. The observed behavior of g -factor with Al concentration is in agreement with the data published in Ref. [174] where a small concentration of Al results in the increase of g -factor including the sign change. However, In the present study, we used only the absolute value of g -factor.

Chapter 8

Macroscopic transverse drift of long current-induced spin coherence in 2DEGs

In this chapter, we imaged the transport of current-induced spin coherence in a two-dimensional electron gas confined in a triple quantum well. Nonlocal Kerr rotation measurements, based on the optical resonant amplification of the electrically-induced polarization, revealed a large spatial variation of the electron g -factor and the efficient generation of a current-controlled spin-orbit field in a macroscopic Hall bar device. We observed coherence times in the nanoseconds range transported beyond half-millimeter distances in a direction transverse to the applied electric field. The measured long spin transport length can be explained by two material properties: large mean free path for charge diffusion in clean systems, and enhanced spin-orbit coefficients in the triple well.

8.1 Motivation

Future electronic technologies based on the spin degree of freedom will require maintaining long quantum coherence times during charge carrier transport in macroscopic devices [13, 175]. The successful performance of this fundamental task needs focused studies on drift and diffusion in test bench systems as, for example, electron spins in GaAs [176–179]. One study approach, early observed in bulk samples, is the drift of optically polarized spins by an in-plane electric field imaged by the age of bunches in

The results presented in this chapter are based on Ref. [2,3] on page 147

the spin polarization [180]. Beyond the simple acceleration of the electron's charge, the electric field changes the momentum dependent spin-orbit fields (B_{SO}) and manipulates the direction of their spins [181]. For two-dimensional electron gases (2DEGs) hosted in a semiconductor quantum well, several reports explored the spin-orbit interaction (SOI) tunability to produce an unidirectional B_{SO} for the diffusive generation of a spin helix [107, 182]. Very recently, the drift in those helical spin systems was also demonstrated showing remarkable properties as the enhancement of the spatial coherence and the electrical current control of the precession frequency [183–185].

A second possibility is the transport study of spins polarized by an electrical current [88–90, 186]. The generation of in-plane current-induced spin polarization (CISP) have been extensively studied in bulk samples [93, 187] as well as in p- and n-doped quantum wells [91, 92, 94, 188, 189]. The CISP has been associated to the spins spatially homogeneous alignment along B_{SO} . In a pioneering work using GaAs epilayers, V. Sih and collaborators studied the drift of electron spins that were polarized in the out-of-plane direction by the spin Hall effect (SHE) [87, 190]. Also in nonlocal experiments, they found that spin currents can be driven over tens of microns in transverse regions with the minimal electrical field, where the transverse spin drift velocities were similar to those for longitudinal charge transport. Furthermore, Y. K. Kato and collaborators also studied the CISP transverse transport in bulk InGaAs using an L-shaped channel [191]. Lately, The CISP has attracted large attention due to the feasibility to be electrically or optically controlled [83, 192] by electron and nuclear spin dynamics [193]. Nevertheless, the relationship between spin-orbit symmetry and electrical spin generation remains controversial and requires further work [151].

8.2 Experimental realization

Here, we studied the CISP transport in a triple quantum well (Sample B) containing a 2DEG. The multilayer system with several subbands occupied offers additional control knobs for the SOI as calculated [194, 195] and experimentally demonstrated in double quantum wells (DQWs) [83, 172]. For TQWs, it was predicted that the SOI could be smoothly tuned by the electron occupation, controlled by a gate voltage, and with a contribution arising from the linear Dresselhaus term being stronger than in DQWs [196]. Such structures have been extensively studied by magnetotransport [13, 175, 176] and also suggested for applications in the production of spin blockers and filters [44, 110]. We mapped the longitudinal and transverse drift of current-induced spin polarization using space-resolved Kerr rotation (KR) in a macroscopic Hall bar. By the periodic optical

control of the CISP [83], the data revealed transverse transport of spin coherence in the nanoseconds range over millimeter distances opening new paths for spintronic devices.

8.3 Time-resolved spin dynamics

The TQW sample was patterned in a macroscopic Hall bar with a width $w = 200 \mu m$, length separation (in the y-axis) between the side probes $L = 500 \mu m$ and $15 \mu m$ wide bridges connecting the main channel to the side regions as sketched in Fig. 8.1. First,

FIGURE 8.1: Scheme of the time-resolved KR in the Voigt geometry.

we studied the sample without the application of electric fields in order to determine the Landé g -factor at $v_d = 0$. We measured the electron spin dynamics using time-resolved KR in the Voigt geometry. We employed a tunable laser with pulse duration of 100 fs and repetition rate of $f_1 = 76$ MHz. The spin polarization is generated by a circularly polarized pump and its precession in a transverse magnetic field (B) was recorded by a linearly polarized probe laser. The pump beam polarization was modulated at a frequency $f_p = 50$ kHz for detection reference of the Kerr angle (Θ_K). Both pulses were focused to approximately $20 \mu m$. Figure 8.2(a) shows the KR as function of the time delay (Δt) between pump and probe pulses for several B at pump/probe power of 1 mW/300 μ W. In Fig 8.2(b), fitting the data with a typical oscillatory exponential decay (Eq. 3.31), we extracted the ensemble spin coherence time (T_2^*) and the Larmor frequency (ω_L). The linear dependence $\omega_L = g\mu_B B/\hbar$, where μ_B is Bohr magneton and $\hbar$ is the reduced Planck's constant, gives the Landé g -factor (absolute value) $g = 0.396$ (see section 8.4).

Furthermore, we found that $T_2^* = 0.5$ ns remains constant up to 1 T and then rapidly decreases due to the ensemble spread of the g -factor [119]. This time scale is limited by the spin-orbit coefficients, Rashba and Dresselhaus linear and cubic, through the Dyakonov-Perel spin relaxation mechanism which is dominant in 2DEGs [107, 108, 142].

FIGURE 8.2: *Optically-induced spin dynamics: (a) KR as function of Δt for different B . (c) Magnetic field dependence of the Larmor frequency (squares) and ensemble spin coherence time (circles).*

8.4 Wavelength dependence of the time-resolved spin dynamics

The TRKR was used to study the two-dimensional electron gas spin dynamics. Measurements were performed with and without external magnetic fields in order to characterize T_2^* and Landé g -factor (g) dependence on excitation wavelength (λ). Figure 8.3(a) shows TRKR scans for the different wavelength at $B = 0$ T. We found a change from an initial

FIGURE 8.3: *For $B = 0$: (a) Time-resolved KR signal for different wavelengths, (b) Amplitude at time delay $\Delta t = 0$ extracted from (a) plotted as function of wavelength.*

FIGURE 8.4: For $B \neq 0$: (a) Magnetic field dependence of T_2^* extracted by fitting the oscillatory part of the KR signal measured for different wavelengths, (b) Electron g -factor obtained from the linear dependence of ω_L on the applied magnetic field for different wavelengths. Pump/probe power of 1 mW/300 μ W and $T = 10$ K.

positive spin polarization to negative followed by an exponential decay. The TRKR amplitude at zero time delay (Δt) is plotted in Figure 8.3(b). The scans from 817 to 819 nm showed a positive component at shorter delay times but away from $\Delta t = 0$. This lineshape was previously associated with the electron spin dynamics in a GaAs/AlGaAs heterojunction system containing a high-mobility 2DEG [197]. With the application of an external transverse magnetic field, TRKR oscillations are observed arising from the precession of coherently excited electron spins about the in-plane field. To obtain the spin coherence time, the evolution of the Kerr rotation angle can be described by an exponentially damped harmonic as described in Eq. 3.31. The extracted magnetic field dependence of T_2^* and g -factor is shown in Fig. 8.4(a) and (b). We found T_2^* in the 0.5 ns range decreasing for fields larger than 1 T. Also, the electron g -factor presents a variation with the wavelength (from 811 to 819 nm) of 0.006 having a maximum at 817 nm.

8.5 Longitudinal spin transport

Now, we switched from optically-induced spin polarization to current-induced spin polarization. Figure 8.5 shows the experimental geometry with $\vec{E} \perp \vec{B}$ and $\vec{B}_{SO} \parallel \vec{B}$. We replaced the optical pump pulse by an AC voltage with tunable rms amplitude V_{AC} and fixed modulation frequency $f_2 = 1.1402$ kHz for Θ_K lock-in detection. The probe

FIGURE 8.5: *Experimental geometry for the optical amplification of the CISP.*

pulse was kept at the same power, wavelength and focus used in the previous section for maximum signal (see section 8.6).

In the longitudinal configuration, we tested the CISP response by applying V_{AC} in ohmic contacts at the central channel (1-2) and measuring the KR as function of B . Figure 8.6(a) shows the amplification of the KR at certain resonant fields due to the constructive interference of the CISP dynamics when it is controlled by optical periodic excitation. As sketched, the electron spins in the 2DEG drift parallel to the in-plane $\vec{E}$ and feel

FIGURE 8.6: *Current-induced spin polarization - Longitudinal configuration. (a) KR as function of B measured for several V_{AC} . (b) Local current and voltage across the sample as a function of the applied voltage. $T = 5$ K.*

a k -dependent spin-orbit field. The spin polarization becomes aligned along $\vec{B}_{SO}$ in a direction perpendicular to $\vec{E}$. The optical pulse train then hits the sample along the out-of-plane direction and rotates the spin polarization towards that direction (detected by polar Kerr rotation).

In a time scale faster than the kHz voltage modulation, the precession (with a GHz frequency) of those electrically-polarized spins is amplified by the pulse train if the ensemble coherence time is long enough to persist between pulses with MHz repetition frequency. Such resonant spin amplification (RSA) of the CISP follows the condition $\Delta B = (\hbar f_1)/g\mu_B B$ and it was previously reported on double quantum well samples [83]. While larger voltages enhance the CISP amplitude, very high voltages are detrimental to the spin coherence due to heating effects. The largest V_{AC} retaining the formation of the RSA pattern thus depends on the electrons temperature (see section 8.6 for similar data at 1.2 and 10 K). Figure 8.6(b) shows a mA current flow while the voltage across the sample is only about 10 % of V_{AC} due to the high mobility. The local resistance $R_L = V_L/i_L$ increases linearly with V_{AC} from 50.35 Ω at 0.5 V to 87.71 Ω at 5 V.

8.6 Wavelength and temperature dependence of the optically controlled CISP

In the same wavelength range of section 8.3, we investigated the amplitude dependence for the RSA pattern formation at a fixed voltage. The data is shown in Figure 8.7. We found a strong dependence on λ with the maximum signal at 817 nm. For the spin transport studies, we chose this wavelength as it strongly shows the current effect on the spin coherence added to the results in section 8.10. We fixed $\lambda = 817$ nm for the following discussions. Next, we explore the temperature influence on the formation of the RSA pattern as function of the applied voltage.

We compared the data at 1.2 and 10 K in Figure 8.8. Clearly, the application of high voltages induces heating which in turn results in strong spin decoherence. This unwanted effect can be delayed by lowering the sample temperature. For $T = 1.2$ K, the spin polarization amplitude increases with the applied voltage reaching a maximum at 3.5 V followed by a decrease. Furthermore, the resonances have a larger amplitude for larger fields, for example at 2.5 V. This is a known indication of long coherence time for the hole spins involved in the generation of the electron spin coherence [165]. For $T = 10$ K, we cannot observe any current-induced spin coherence beyond 2.0 V. Also note the drastic change of the resonances width related to loss of spin coherence when increasing the temperature.

FIGURE 8.7: Magnetic field scans of the KR signal measured for different probe wavelengths at a fixed power of $300 \mu\text{W}$. $V_{AC} = 3 \text{ V}$ and $T = 1.2 \text{ K}$.

FIGURE 8.8: Magnetic field scans of the KR signal measured for different applied voltages at $T = (a) 1.2 \text{ K}$ and $(b) 10 \text{ K}$.

8.7 Transverse spin transport

Next, we analyze the transverse transport of spin coherence in regions where the electric fields are considerably reduced. Figure 8.9 shows the reflectivity map of the device displaying the central channel and side voltage probes. The low intensity (red) regions indicate the gaps between the conducting areas, the white lines are the edges of those regions and the solid lines are the possible current paths. The horizontal features at $y = 0$ mm are the gaps separating the contacts 6-5 and 3-4.

FIGURE 8.9: Reflectivity map of the device.

First, we measured the dependence of the CISP amplitude and coherence on V_{AC} using contacts 5-4 and setting the probing spot away at $(x,y) = (0,-0.4)$ mm. The result is plotted in Fig. 8.10(a). We will focus on the spin coherence time for $B = 0$ which can be directly evaluated from the width of the zero-th resonance using a Hanle model as described in eq 4.3. The dashed line guides the field position of the zero-th peak. There, a constant null total magnetic field, arising from the addition of $\vec{B}$ and $\vec{B}_{SO}$, requires the shifting of the Hanle peak towards $B = -B_{SO}$ [198]. The fitted curve was plotted on top of the experimental data (red line) and the extracted parameters are presented in Fig. 8.10(b). In contrast to the longitudinal case, where the spin polarization is measured in a region with a local current flow, the RSA holds over a large voltage range with a V_{AC} enhancement of the spin coherence reaching 5.27 ns at 3 V. T_2^* decreases for higher voltages but still remains in the nanoseconds range up to 5 V. Complementary, the current-controlled spin-orbit field attains more than 5 mT at 3 V. This field agrees with the theoretical value $\langle \vec{B}_{SO}^* \rangle$ calculated for $v_d = 0.5$ nm/ps in [31]. Nevertheless,

FIGURE 8.10: B scans of KR at $(x,y) = (0,-0.4)$ mm applying different V_{AC} in contacts 5-4. The dashed line is a guide to the eyes for the zero-field resonance position at $B = -B_{SO}$. The red line is a Hanle model from where T_2^* (squares) and B_{SO} (circles) were extracted and plotted in (b).

the expected drift velocity for the longitudinal configuration should be much larger and equal to $V_d = I/(en_s w) = 18.1$ nm/ps for a current $I = 5$ mA at $V_{AC} = 3$ V (see Fig. 8.6) with an electric field of $E = V_L/L = 0.9$ kV/m. Therefore, we found that v_d is almost two orders of magnitude smaller in the transverse regions far from the current flow in comparison with longitudinal sections (inside the flow).

The device geometry allow us to examine the variation of the spin coherence transport as function of the width of the region that connects the current path to the transverse zones. For instance, when V_{AC} is applied along x , the transverse region along y consists of the central channel. Reciprocally, when V_{AC} is applied along y , the transverse transport will be into the lateral arms of the Hall bar connected to the current path by narrow bridges giving a strong drift constriction in the x axis. We fixed $V_{AC} = 3$ V for the largest spin-orbit field, CISP amplitude and spin coherence time. In Fig. 8.11, we used contacts 5-4 approximately $250 \mu\text{m}$ away from the measuring spots to emulate a nonlocal geometry. We applied V_{AC} in the x axis and scanned the probe position in the y axis (at $x = 0$ along the central channel) while sweeping the magnetic field. The CISP transverse transport was observed again in a scan of about $\Delta y = 0.5$ mm. Nevertheless, the CISP decay now became visibly weaker with distance.

FIGURE 8.11: *Transverse spin drift along the $(0,y)$ mm axis with $V_{AC} = 3$ V. The Hall bar displays the sweeping direction (arrows) and the contacts used for V_{AC} application. The closed circles show the spatial variation of electron g-factor.*

A remarkable result is the change of the ΔB period in the RSA pattern observed for both x and y scans. In Fig. 8.11, this effect is seen by the shift of the outer resonances towards higher fields as function of y. On the other side, the zero-th resonance position seems to remain constant given by V_{AC} . Our experimental technique separates the influence of the B_{SO} and g-factor changes: while the first behaviour depends exclusively on the

FIGURE 8.12: *Transverse spin drift along the $(x,-0.35)$ with $V_{AC} = 3$ V. The Hall bar displays the sweeping direction (arrows) and the contacts used for V_{AC} application.*

g -factor, the second is related only to B_{SO} (see also Fig. 8.10(a) and (b)). The spatial dependence of g is displayed by solid circles in Fig. 8.11. The modification of the g -factor by an in-plane electric field was reported recently. For bulk InGaAs epilayers [199], they measured $\Delta g = 0.0053$ and found that g increases with the drift velocity.

For [110] oriented QWs [200], the electrical variation of the g tensor was also found to depend on the external magnetic field and to increase with the current [200]. Our data agrees with those reports as the g -factor decreases for electrons that drifted far from the current path. Nevertheless, we reached a variation of $\Delta g = 0.02$ which could indicate a larger change on the drift velocity because $\Delta g \propto V_d^2$ [199]. We noted that, at the longest transverse distance from the current path ($y = 0.4$ mm), we recovered the g -factor value measured at $V_d = 0$ in Fig. 8.2(b). However, the data does not show clearly the expected variation of B_{SO} with y (or V_d). This may be explained considering the additional experimental difficulty originated from the fact that the variation with the drift is smaller for B_{SO} than for g , since B_{SO} depends only linearly on V_d (see section III.c of Ref. [31]).

In Fig. 8.12, we applied V_{AC} in the y axis (contacts 1-2) and scanned the probe position in the x axis (at constant $y = -0.35$ mm) from the left to the right side arms (5-4) passing by the central channel also. The x -scan thus allows us to map simultaneously the longitudinal and transverse transport. Inside the central channel, we reproduced the data from Fig 8.6 where the high current leads to low spin polarization due to heating (white shade from $x = 0$ to ± 0.1 mm). Entering the transverse side regions, the polarization was observed in long ranges of $\Delta x = \pm 0.5$ mm. Furthermore, the spin

FIGURE 8.13: (a) Amplitude of the current-induced spin polarization and (b) the spin coherence transported along y extracted from Fig 8.11. The fitting of spin polarization decay (solid line) gives ℓ_s as parameter. All the error bars correspond to the fitting standard error. $T = 5$ K.

transport extension can be inferred from the KR dependence on the spatial parameter (y or x) estimated by $\exp(-y/\ell_s)$ where ℓ_s is the CISP transport length.

Fig 8.13(a),(b) and Fig 8.14(a),(b) show the KR amplitude and coherence time following a resonance peak in Fig 8.11 and Fig 8.12, respectively. The lengths obtained from the exponential decay fitting (solid line) of the spin polarization are $\ell_s = 0.171$ mm for scans along x and 0.685 mm for y. Figure 8.13(b) displays the surprising result that the CISP transverse drift can drive constant spin coherence of about 6 ns by almost half millimeter. Furthermore, for the x-scan, the spin coherence is lose from a value close to 5 ns to around 1 ns in a similar displacement. Those values for ℓ_s and T_2^* are independent of the field resonance chosen for the analysis within the experimental error (see section 8.8). The measured ℓ_s asymmetry is likely a result of the device geometry. The anisotropy of B_{SO} related to the orientation of the Hall bar with the crystallographic axes was not experimentally evaluated. Our calculation indicated that both in-plane orientations are equivalent (see discussion in Ref. [31]).

FIGURE 8.14: (a) Amplitude of the current-induced spin polarization and (b) the spin coherence transported along x extracted from Fig 8.12. The fitting of spin polarization decay (solid line) gives ℓ_s as parameter. All the error bars correspond to the fitting standard error. $T = 5$ K.

8.8 Field dependence of the transport length for spin coherence

In the section 8.7, we showed the spin transport length ℓ_s extracted from the decay of a single resonance peak at a given magnetic field. This is a condition of approximately

FIGURE 8.15: KR amplitude and coherence time as function of y for different magnetic fields.

FIGURE 8.16: KR amplitude and coherence time as function of x for different magnetic fields.

constant field since the peak shifts due to the period modification with the g -factor variation in space. Figures 8.15 and 8.16 show fittings, similar to the Figure 8.13 and 8.14 in the section 8.7, for different conditions of high and low magnetic field as well as different field polarity. All the results are consistent within the experimental error. We conclude that the obtained ℓ_s is robust and the long spin coherence is maintained for mT fields (in agreement with Figure 8.4(a)).

8.9 Nonlocal charge transport

In this section, we investigate the characteristic length for transverse charge transport in our device. The observed long spin transport length requires not only large spin-orbit coefficients but also needs a large mean free path for charge diffusion. In the clean system, the large charge diffusion will extend the regions where the current induces spin polarization towards transverse directions. On the other side, the large calculated spin-orbit coefficients will then enhance and maximize the efficiency of the polarization generation. We also explored if the large spin drift of the current-induced spin polarization can inversely produce a charge current in close contacts. In the spin Hall effect regime, it is expected that long spin diffusion leads to nonlocal charge transport by means of the inverse SHE [201, 202]. In the Rashba-Edelstein frame, also an inverse effect was proposed for clean electron gases when electron-impurity scattering is very weak in a nonlinear regime [203]. The nonlocal transport can be characterized by the transresistance R_{NL} defined by the ratio between the nonlocal response voltage V_{NL} (in 5-4) and the applied current i_L (in 6-3) for a given V_{AC} .

The experimental configuration for the measurement of the nonlocal voltage is shown as an inset of Fig. 8.17. In this configuration, we note that $\vec{E} \parallel \vec{B}$ and $\vec{B}_{SO} \perp \vec{B}$ which could produce an enhancement of spin polarization for fields larger than zero resembling an antisymmetric Hanle curve [93] or asymmetric if $\vec{B}_{SO}$ is not exactly perpendicular to $\vec{B}$ [204]. For $B = 0$, we observed a nonlocal voltage on the order of tens of μV increasing with V_{AC} until saturation. Measuring the local current at the source contacts (6-3), we found a maximum R_{NL} of 0.037Ω for $V_{AC} = 3 V$.

In order to differentiate the contributions from spin mediated transport and classical charge diffusion, the Ohmic component in the nonlocal resistance for a narrow strip can be estimated as $R_{NL} = R_L \exp(-y/\ell_C)$ where $\ell_C = w/\pi$ and $y = L$ is the distance from the V_{AC} contacts [205, 206]. For our device, we obtain a large transport length $\ell_C = 64 \mu m$ implying that the charge mediated mechanism should be important for R_{NL} . It also may hinder the spin mediated phenomena even for $y = L = \ell_S$ due to the large mean free path for charge in the clean system [201]. Determining the local resistance, see

FIGURE 8.17: Nonlocal transport: Nonlocal voltage (circles) and transresistance (squares). The inset shows the local resistance and the experimental configuration. The theoretically estimated Ohmic contribution to the R_{NL} is shown by the dashed line. $B = 0$ T.

inset in Fig. 8.17, we calculated the expected R_{NL} given by the Ohmic contribution and plotted it with a dashed line. Both curves for R_{NL} , experimental and theoretical, present similar peak position but with an amplitude difference of 10%. Also, the experimental curve is broader keeping the maximum value constant in a larger voltage band. More work is needed to study wire structures in order to isolate the spin and charge-related conductance contributions in all-electrical measurements.

8.10 Magnetoresistance in perpendicular magnetic fields

Magnetotransport measurements in triple quantum wells are well described in the recent literature. See References [125, 127, 207] for further details of transport studies on similar samples. Figure 8.18 shows the longitudinal (R_{xx}) and transverse (R_{xy} - Hall) magnetoresistance in a perpendicular field. The observed oscillations in R_{xx} consist of the interplay between two types of oscillating phenomena. In 2DEGs, Shubnikov-de Hass (SdH) oscillations occur due to a periodic modulation of electron scattering as the Landau levels consecutively pass through the Fermi level. Additionally, in quantum wells with two or more occupied subbands, the magnetoresistance exhibits another type of oscillations, the so-called magnetointersubband (MIS) oscillations.

FIGURE 8.18: Normalized longitudinal (R_{xx}) and Hall (R_{xy}) resistances with perpendicular field. $T = 1.2$ K.

MIS oscillations occur because of a modulation of the probability of transitions between the Landau levels belonging to different subbands which is periodic in the field. The MIS oscillation peaks correspond to the maximal scattering of electrons between the Landau levels when the subband separation equals a multiple of the cyclotron energy. For our high-density 2DEG, we can measure only high Landau levels for moderate fields. The

FIGURE 8.19: The SdH oscillations: (a) R_{xx} as function of $1/B$, (b) Fourier transform of (a).

quantization of R_{xy} associated with the minima in R_{xx} is also displayed in Figure 8.18. The SdH oscillations are periodic in $1/B$ (see Figure 8.19(a)). From that periodicity, one can obtain the subbands density n_i according to $1/f_i = (2e)/(hn_i)$ where f_i is the frequency of the oscillation in the $1/B$ plot. In Figure 8.19(b), the Fourier transform results in 3 large peaks arising from the 3 subbands. The peak with lower frequency corresponds to the lower density in the third subband, and the other two correspond to similar densities in the first and second subband. The first two subbands are almost degenerate ($\Delta_{12} \sim 0$) as confirmed by calculation [31]. We found that Δ_{12} is very sensitive to the width of the lateral wells (increasing rapidly for smaller widths).

8.11 Summary

In this chapter, we present the study of the current-induced spin polarization in a 2DEG confined in a triple quantum well. We found that the TQW has exceptional properties for the current-control of spin-orbit fields, as we calculated and later experimentally observed. The data showed long coherence time for the spin ensemble in the nanoseconds range.

Surprisingly, we observed the drift transport of such current-induced spin polarization over macroscopic distances in a direction transverse to the applied electric field. During the transport, the spin polarization retain its long-lived nanosecond coherence, and the polarization amplitude decay was found to be limited by the device geometry.

The drifting electrons acquire a variation of the Landé g -factor as a function of the velocity controlled by the proximity to the current path. A spin-orbit field was tuned by the applied voltage reaching several mT. The transverse spin transport length was found to be one order of magnitude larger than the Ohmic charge diffusion in the studied configuration.

The observed long spin transport length can be explained by two material properties in the TQW: large mean free path for charge diffusion in the clean systems, and large spin-orbit coefficients. Future studies in narrow wire channels are still required to distinguish charge mediated from spin-mediated transport in all-electrical measurements. Additional measurements with high spatial resolution are still required to verify the calculated proximity of the left and right subbands to the crossed persistent spin helix regime.

The current investigation in a macroscopic device illuminates a path for practical applications using other complex material systems including ferromagnet/semiconductor hybrids, metallic and magnetic thin-films [208–210]. The presented experimental method may be relevant for electrical switching of the direct and inverse spin Hall and spin-galvanic effects [211–213].

Chapter 9

Conclusions

The last chapter of the thesis in hand is dedicated to summarizing all the experimental results obtained on two-dimensional electron gases hosted in GaAs/AlGaAs and $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{AlAs}$ quantum wells, doped beyond metal-insulator transition, with and without external magnetic field.

9.1 Thesis summary

The central theme of the thesis at hand is to shed light on the generation, detection, and manipulation of electron spin coherence in semiconductor nanostructures. For the successful implementation of new functionalities using the spin degree of freedom in technological platforms, the selection of an appropriate structure with a lowest possible spin dephasing rate is an essential requirement. According to previous literature, the longest spin dephasing time for Si-doped n-GaAs has been found at the doping concentration close to metal-insulator transition [46–48]. All the experimental work reported in this thesis used GaAs/AlGaAs or $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{AlAs}$ QWs with doping concentration beyond metal-insulator transition. The spin dynamics in these materials have been widely studied using single QW structures [99, 101], however, by exploring the multi-layer system (double and triple QWs), we have enhanced the list of available material systems.

The thesis began with a general introduction in the chapter 1 followed by a background theory for the understanding of the results (chapter 2). There we introduced basic properties of GaAs, 2DEG heterostructure, spin precession around magnetic field and Landé g -factor and briefly introduced metal-insulator transition. The main focus was put on the origin of spin-orbit coupling and an overview of the principal mechanism responsible for spin dephasing in semiconductor nanostructures.

To study the dynamics of optically generated spins, we successfully introduced the pump-probe techniques: time-resolved Kerr rotation and Resonant spin amplification, which are based on the optical pumping and probing (chapter 3). Additionally, we introduced the current-induced spin polarization technique where the AC voltages with tunable rms amplitude V_{AC} were applied to the sample to inject spin polarization and probe laser pulses were used for the readout of the spin polarization. Furthermore, an overview of the optical set-up to perform these measurements was given.

In chapter 4 the observation of long-lived nanosecond spin coherence in (001) oriented GaAs/AlGaAs quantum well structures was studied in dependence of various experimental parameters. The varied parameters were the applied magnetic field, excitation wavelength, time delay between pump and probe and excitation power and sample temperature. In the first part of the chapter, we studied the spin dynamics of the double quantum well (sample A). The electron spin dephasing time of 4.4 ns was measured. To achieve the longest electron spin dephasing time a modified form of the pump-probe Kerr rotation technique known as resonant spin amplification was used.

Additionally, we measured the time evolution of Kerr rotation signal for two triple quantum wells (sample B and C) with different well width. Due to long spin coherence time in sample B, the decay of KR envelope was very weak, and it was difficult to retrieve T_2^* by fitting. The resonant spin amplification was measured which yield $T_2^* > 10$ ns. The observed results are among the longest time reported in the literature for the samples of similar doping levels [51, 116]. In contrast, sample C displayed fast decoherence due to the presence of spin-orbit coupling.

Next, in chapter 5 of this thesis, a large spin relaxation anisotropy up to a factor of 10 has been observed for the in- and out-of-plane spin orientations. We believe that this anisotropy stems from the presence of an internal magnetic field and the inhomogeneity of electron g -factor. Different experimental parameters such as sample temperature, pump-probe delay, and excitation power were varied to explore their effect on the spin relaxation anisotropy. The analysis of the data yields the magnitude of the internal magnetic field of the order of few mT which has been found to decrease strongly with the sample temperature and excitation power.

Chapter 6 of the thesis presents the influence of sample temperature on the spin dynamics recorded in sample B. The main investigation methods used for this study were time-resolved Kerr rotation and resonant spin amplification. Two wavelengths with larger spin relaxation time were chosen for this study. TRKR data revealed the robustness of the carrier's spin precession against growing temperature and was traced in a broad range up to 250 K. Long-lived spin coherence with dephasing time $T_2^* > 14$ ns was found at $T = 5$ K for $\lambda = 823$ nm. It has been found that T_2^* decreases with the growth of temperature

due to the efficient Dyakonov-Perel mechanism and become much shorter of the order 0.27 ns at $T = 250$ K. The g -factor evaluated from TRKR data, by fitting the spin precession with decaying cosine function, was found to grow linearly with temperature.

Chapter 7 of the thesis corresponds to the effects of Al contents on spin dynamics. We investigated four different samples, one single and three double quantum wells, of the equal well width of 14 nm but different Al contents ($8.2\% < x < 16\%$) inside each well. In the single quantum well sample ($x = 10\%$) and DQW with $x = 8.2\%$, we observed a signal from the bulk which was identified by its g -factor. Comparison of the evaluated parameters in all the samples reveals that the spin dephasing time and electron g -factor changes remarkably with the Al contents inside the wells.

Finally, in last part of the thesis (chapter 8), we explored the drift of long current-induced spin coherence. We studied the current-induced spin transport of a high mobility dense two-dimensional electron gas confined in GaAs/AlGaAs triple quantum well (sample C). Optically-controlled CISP was performed as a function of laser detuning and sample temperature from where we found that signal was stronger at $\lambda = 817$ nm and lower temperature.

By longitudinal spin transport, we confirmed that the voltage across the sample is only 10 % of the applied V_{AC} due to high mobility of the sample. Furthermore, the transverse transport of spin coherence was analyzed where we observed the drift transport of current-induced spin polarization over the macroscopic range. The results of the experiments on the studied structure showed that transverse drift of current-induced spin polarization could drive a constant spin coherence of around 6 ns by almost half a millimeter. Additionally, the spatial dependence of the Landé g -factor was found.

The obtained results have enlightened the initialization, manipulation, and detection of spin coherence in semiconductors which are three of the main prerequisite for practical applications in both spintronics and quantum information processing.

Publications

Parts of this work are/will be published as follows:

1) *“Long-lived nanosecond spin coherence in high-mobility 2DEGs confined in double and triple quantum wells”*

Saeed Ullah, G. M. Gusev, A. K. Bakarov, and F. G. G. Hernandez

Journal of Applied Physics **119**, 215701 (2016)

2) *“Macroscopic transverse drift of long current-induced spin coherence in two-dimensional electron gases”*

F. G. G. Hernandez, **Saeed Ullah**, G. J. Ferreira, N. M. Kawahala, G. M. Gusev, A. K. Bakarov

Phys. Rev. B 94, 045305 (2016)

3) *“Macroscopic transport of a current-induced spin polarization”*

Saeed Ullah, G. J. Ferreira, G. M. Gusev, A. K. Bakarov, and F. G. G. Hernandez

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4) *“Large anisotropic spin relaxation time of exciton bound to donor states in triple quantum wells”*

Saeed Ullah, G. M. Gusev, A. K. Bakarov, and F. G. G. Hernandez

Submitted to Journal of Applied Physics. arXiv: 1703.07156 [Cond-mat.mes-hall] (2016).

5) *“Robustness of spin coherence against high temperature in multilayer system”*

Saeed Ullah, G. M. Gusev, A. K. Bakarov, and F. G. G. Hernandez

In preparation

List of participation in conferences

- 1) *“The 16th Brazilian Workshop on Semiconductor Physics”*, Itirapina- SP- Brazil, May 5-10, 2013.
- 2) *“XXXVII Brazilian Meeting on Condensed Matter Physics”*, Costa do Sauipe, Bahia-Brazil, May 12-16, 2014.
- 3) *“The 8th International Conference on Physics and Applications of Spin Phenomena in Solids (PASPS VIII)”*, Washington D.C., USA, July 28-31, 2014.
- 4) *“The 17th Brazilian Workshop on Semiconductor Physics (BWSP-17)”*, Uberlandia/MG, Brazil, May 03-08, 2015.
- 5) *“The 8th International School and Conference on Spintronics and Quantum Information Technology (Spin Tech VIII)”*, Basel, Switzerland, August 10-13, 2015.
- 6) *“XXXIX Brazilian Meeting on Condensed Matter Physics (XXXIX ENFMC)”*, Natal-Rio Grande do Norte-Brazil, September 03-07, 2016.

Appendix A

A Mathematical Proofs

A.1 Landau quantization

Consider the quantum mechanical problem of the motion of an electron in 2DEG in a homogeneous magnetic field directed perpendicular to the plane of 2DEG i.e., $\vec{B} = (0, 0, B)$. The Hamiltonian for such a system is given by

$$H = \frac{1}{2m^*} \left(\vec{p} - e\vec{A} \right)^2, \quad (\text{A.1})$$

so that the Schrödinger equation for the electron in 2DEG has the following form:

$$\left[\frac{1}{2m^*} \left(\vec{p} - e\vec{A} \right)^2 \right] \psi(x, y) = E\psi(x, y) \quad (\text{A.2})$$

where $\vec{A}$ is the vector potential associated with magnetic field $\vec{B} = \vec{\nabla} \times \vec{A}$, and $\vec{p} = -i\hbar\vec{\nabla}$ is the momentum operator. The so called Landau gauge is a convenient choice for the vector potential, where, $\vec{A}$ can be chosen such that $\vec{B}$ remains unchanged. For instance, choosing Landau gauge, $\vec{A} = B(0, x, 0)$ the Schrödinger equation takes the following form:

$$\left[\frac{1}{2m^*} (p_x^2 + (p_y + eBx)^2) \right] \psi(x, y) = E\psi(x, y) \quad (\text{A.3})$$

As the vector potential is independent of y, suggesting that the wave function should be a product of an unknown function of x with a plane wave of y.

$$\psi(x, y) = \varphi(x)\chi(y) = \varphi(x) \frac{1}{\sqrt{L}} \exp(iky) \quad (\text{A.4})$$

Substituting this equation in Eq. A.3 we can rewrite it as

$$\left[\frac{1}{2m^*} (p_x^2 + (\hbar k_y + eBx)^2) \right] \varphi(x) = E\varphi(x) \quad (\text{A.5})$$

Using $\vec{p} = -i\hbar\vec{\nabla}$, the above equation yields

$$\left[\frac{p_x^2}{2m^*} + \frac{1}{2}m^*\omega_c^2 \left(x + \frac{\hbar k_y}{eB} \right)^2 \right] \varphi(x) = E\varphi(x), \quad (\text{A.6})$$

$$\left[\frac{p_x^2}{2m^*} + \frac{1}{2}m^*\omega_c^2 (x + x_k)^2 \right] \varphi(x) = E\varphi(x) \quad (\text{A.7})$$

One can easily see that this is nothing but the Schrödinger equation for the harmonic oscillator on the x-axis shifted from the origin by $x_k = \hbar k_y / eB$ with cyclotron frequency $\omega_c = eB / m^*$. This problem is solved with

$$\varphi(x) = \phi_n(x - x_k) = \frac{1}{2^n n! \sqrt{\pi} \ell_B} \exp \left[-\frac{(x - x_k)^2}{2\ell_B^2} \right] H_{n-1} \left(\frac{x - x_k}{\ell_B} \right) \quad (\text{A.8})$$

Where $n = 1, 2, 3, \dots, x_k = \hbar k_y / eB = \ell_b^2 k_y$ is the centre of cyclotron orbit, where we have introduced the so called magnetic length: $\ell_B = \sqrt{\hbar / eB} = \sqrt{\hbar / m^* \omega_c}$ and H_{n-1} is the $(n-1)^{th}$ Hermite polynomial. For the electron moving in xy-plane in the magnetic field, we can, therefore, write the wave function and energies as eignvalues of Eq. A.3.

$$\psi_n(x, y) = \phi_n(x - x_k) \exp(ik_y y) \quad (\text{A.9})$$

$$E_n = \hbar\omega_c \left(n + \frac{1}{2} \right) \quad (\text{A.10})$$

For the electrons moving in cyclotron orbits, the density of states $D(E)$ is no longer constant, but obeys an additional quantization giving rise to a series of delta-like energy levels, the so called Landau levels, spaced by $\hbar\omega_c$ (See Fig. A.1). The step-like $D(E)$ associated with 2DEG is given by

$$D(E) = \frac{g_s}{2\pi\ell_B^2} \sum_{i,n} \delta(E - E_{i,n}) \quad (\text{A.11})$$

Where

$$E_{i,n} = E_i + \hbar\omega_c \left(n + \frac{1}{2} \right) \quad (\text{A.12})$$

Where $i, n = (0, 1, 2, 3, \dots)$ with sub-band index i and Landau quantum number n , g_s is the spin degeneracy and δ is Dirac function. In real systems the LLs are slightly broadened due to scattering events caused by crystal defects and incorporated impurities.

FIGURE A.1: The electron energy states in k -space with (a) $B = 0$ and (b) $B \neq 0$. Density of state $D(E)$ vs energy E for 2DEG at (c) $B = 0$ and (d) $B \neq 0$. Broadening of δ -like LL peaks in $D(E)$ with increasing magnetic field.

As a consequence, we observe the SdH oscillations. Therefore the $D(E)$ can be written as

$$D(E) = \frac{g_s}{2\pi\ell_B^2} \sum_{i,n} \Gamma(E - E_{i,n}) \quad (\text{A.13})$$

Where $\Gamma = \hbar/2\tau_q$ is the broadening in LLs due to crystal defects and impurities with quantum scattering time τ_q . In order to get the number of states in LL, consider a rectangular sample with dimensions L_x and L_y . The wave vector k is quantized in multiples of $\Delta k = 2\pi/L_y$. The maximum value of the wave vector k_{max} can be found from the condition that the position of the center of oscillator x_k takes the value L_x i.e. $L_x = \hbar k_{max}/eB$. Thus the number of states in LL is given by

$$N = \frac{k_{max}}{\Delta k} = \frac{eB}{2\pi\hbar} L_x L_y \quad (\text{A.14})$$

The degeneracy of a LL defined as the number of states in LL per unit area can be obtained as

$$N_L = \frac{N}{A} = \frac{eBA}{2\pi\hbar A} = \frac{eB}{2\pi\hbar} = \frac{1}{2\pi\ell_B^2} \quad (\text{A.15})$$

Thus, in term of N_L Eq. A.13 can be written as

$$D(E) = g_s N_L \sum_{i,n} \Gamma(E - E_{i,n}) \quad (\text{A.16})$$

The filling factor ν is a useful measure for the number of occupied Landau levels which can be defined as the ratio of electron density to the Landau level density of states:

$$\nu = \frac{n_s}{N_L} = 2\pi\ell_B^2 n_s = \frac{\hbar n_s}{eB} \quad (\text{A.17})$$

It's clear from the above relation, that for a given n_s , the increase in magnetic field results in the decrease of filling factor.

A.2 Derivation of RSA formula

In Sec. 3.6 we discussed the precession of spin ensembles due to single excitation around an external magnetic field given by

$$\Theta_K(\Delta t, B) = A \exp\left(\frac{-\Delta t}{T_2^*}\right) \cos\left(\frac{|g|\mu_B B}{\hbar} \Delta t\right) \quad (\text{A.18})$$

where T_2^* is the ensemble dephasing time, A is the initial amplitude, and $g = \hbar\omega_L/\mu_B B$ is the electron g -factor with reduced Planck constant $\hbar$, Larmor frequency ω_L , Bohr magneton μ_B , and external magnetic field B.

In this section, we proceed the other way round by taking into account the repetitive spin injection by successive laser pulses at fixed repetition period t_{rep} . The derivations follow the one presented in Ref. [214]. To calculate the net magnetization by summing up all previous excitation build up by a repetitive injection one gets

$$\Theta_K(\Delta t, \omega_L) = \sum_{n=1}^{\infty} A \exp\left(-\frac{(\Delta t + nt_{rep})}{T_2^*}\right) \cos(\omega_L(\Delta t + nt_{rep})) \quad (\text{A.19})$$

with $\Delta t \in [t_{rep}; 0]$ and $t_{rep} > 0$. By writing the cosine term in exponential form

$$\cos[\omega_L(\Delta t + nt_{rep})] = \frac{1}{2} [\exp(i\omega_L(\Delta t + nt_{rep})) + \exp(-i\omega_L(\Delta t + nt_{rep}))] \quad (\text{A.20})$$

Using Eq. A.20 in A.19 we can rewrite it as

$$\Theta_K(\Delta t, \omega_L) = \sum_{n=1}^{\infty} A \exp\left(-\frac{(\Delta t + nt_{rep})}{T_2^*}\right) \frac{1}{2} [\exp(i\omega_L(\Delta t + nt_{rep})) + \exp(-i\omega_L(\Delta t + nt_{rep}))] \quad (\text{A.21})$$

Using $\varrho = \left(\frac{1}{T_2^*} - i\omega_L\right)$ and $\varrho^* = \left(\frac{1}{T_2^*} + i\omega_L\right)$ in the above equation and rearranging yields:

$$\Theta_K(\Delta t, \omega_L) = \frac{A}{2} \left[\exp(-\Delta t \varrho) \sum_{n=1}^{\infty} \exp(-nt_{rep}\varrho) + \exp(-\Delta t \varrho^*) \sum_{n=1}^{\infty} \exp(-nt_{rep}\varrho^*) \right] \quad (\text{A.22})$$

For integral number $n \in \mathbb{N}$

$$\exp(-nx) = \prod_{i=1}^n \exp(-x) = [\exp(-x)]^n \quad (\text{A.23})$$

Thus Eq. A.22 takes form

$$\Theta_K(\Delta t, \omega_L) = \frac{A}{2} \left[\exp(-\Delta t \varrho) \sum_{n=1}^{\infty} \exp(-t_{rep}\varrho)^n + \exp(-\Delta t \varrho^*) \sum_{n=1}^{\infty} \exp(-t_{rep}\varrho^*)^n \right] \quad (\text{A.24})$$

For $t_{rep} > 0$, $T_2^* > 0$ the infinite geometrical series converges $|z| < 1$

$$\sum_{n=1}^{\infty} x^n = -\frac{x}{x-1} \quad (\text{A.25})$$

For real number x , $|\exp(ix)| = 1$ and also for $t_{rep}/T_2^* > 0$, the sum of geometrical series in Eq. A.24 is guaranteed

$$|\exp(-t_{rep}\varrho)| = \left| \exp \left[-t_{rep} \left(\frac{1}{T_2^*} - i\omega_L \right) \right] \right| = \left| \exp \left(\frac{-t_{rep}}{T_2^*} \right) \right| |\exp(-it_{rep}\omega_L)| < 1. \quad (\text{A.26})$$

Therefore, Eq. A.24 can be simplified to:

$$\Theta_K(\Delta t, \omega_L) = \frac{A}{2} \left[\frac{\exp(-\Delta t \varrho)}{\exp(t_{rep}\varrho) - 1} + \frac{\exp(-\Delta t \varrho^*)}{\exp(t_{rep}\varrho^*) - 1} \right] \quad (\text{A.27})$$

Taking LCM leads to

$$\Theta_K(\Delta t, \omega_L) = \frac{A}{2} \left[\frac{\exp(-\Delta t \varrho) \exp(t_{rep}\varrho^* - 1) + \exp(-\Delta t \varrho^*) \exp(t_{rep}\varrho - 1)}{[\exp(t_{rep}\varrho) - 1][\exp(t_{rep}\varrho^*) - 1]} \right] \quad (\text{A.28})$$

Further simplification implies

$$\Theta_K(\Delta t, \omega_L) = \frac{A}{2} \left[\frac{\exp(-\Delta t \varrho) \exp(t_{rep}\varrho^* - 1) + \exp(-\Delta t \varrho^*) \exp(t_{rep}\varrho - 1)}{\exp[t_{rep}(\varrho + \varrho^*)] - [\exp(t_{rep}\varrho) + \exp(t_{rep}\varrho^*)] + 1} \right] \quad (\text{A.29})$$

By introducing the values of ϱ and ϱ^* in Eq. A.29 it can be written as

$$\Theta_K(\Delta t, \omega_L) = \frac{A}{2} \left[\frac{\exp(-\Delta t/T_2^*) [\exp(-i\omega_L \Delta t) (\exp(t_{rep}/T_2^* - i\omega_L t_{rep} - 1))]}{\exp(2t_{rep}/T_2^*) + 1 - 2\operatorname{Re}(\exp(t_{rep}/T_2^* + i\omega_L t_{rep} - 1))} + \frac{\exp(i\omega_L \Delta t) (\exp(t_{rep}/T_2^* + i\omega_L t_{rep} - 1))}{\exp(2t_{rep}/T_2^*) + 1 - 2\operatorname{Re}(\exp(t_{rep}/T_2^* + i\omega_L t_{rep} - 1))} \right] \quad (\text{A.30})$$

$$\begin{aligned} \Theta_K & \quad (\text{A.31}) \\ &= \frac{A}{2} \exp\left(\frac{-\Delta t}{T_2^*}\right) \left[\frac{\exp(t_{rep}/T_2^* - i(\Delta t + t_{rep})\omega_L) + \exp(t_{rep}/T_2^* - i(\Delta t + t_{rep})\omega_L)}{\exp(t_{rep}/T_2^*) [\exp(t_{rep}/T_2^*) + \exp(-t_{rep}/T_2^*)] - 2\exp(t_{rep}/T_2^*) \cos(\omega_L t_{rep})} \right. \\ & \quad \left. - \frac{[\exp(i\omega_L \Delta t) + \exp(-i\omega_L \Delta t)]}{\exp(t_{rep}/T_2^*) [\exp(t_{rep}/T_2^*) + \exp(-t_{rep}/T_2^*)] - 2\exp(t_{rep}/T_2^*) \cos(\omega_L t_{rep})} \right] \end{aligned}$$

$$\Theta_K = \frac{A}{2} \left[\frac{\exp(-\Delta t/T_2^*) [\exp(t_{rep}/T_2^*) 2\cos(\omega_L(t_{rep} + \Delta t)) - 2\cos(\omega_L \Delta t)]}{\exp(t_{rep}/T_2^*) [2\cosh(t_{rep}/T_2^*) - 2\cos(\omega_L t_{rep})]} \right] \quad (\text{A.32})$$

Thus, we have gotten the required Eq. 3.33.

$$\Theta_K(B, \Delta t) = \frac{A}{2} \exp\left(-\frac{(\Delta t + t_{rep})}{T_2^*}\right) \left[\frac{\cos(\omega_L \Delta t) - \exp(t_{rep}/T_2^*) \cos[\omega_L(\Delta t + t_{rep})]}{\cos(\omega_L t_{rep}) - \cosh(t_{rep}/T_2^*)} \right] \quad (\text{A.33})$$

with $\Delta t \in [0; t_{rep})$.

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