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Spindynamics of electrons and holes in diluted magnetic- and hybrid ferrometal-semiconductor quantum well structures

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Spindynamics of electrons and holes in diluted magnetic- and hybrid ferrometal-semiconductor quantum well structures

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August 2002

Report of a graduation project (August 2001 - August 2002) carried out at the Eindhoven University of Technology in the group Physics of Nanostructures (FNA)

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Abstract

Electron spin is becoming increasingly popular in electronics. The prospect of building electronic devices that use the spin orientation of carriers in addition to their charge, usually referred to as spintronics, has become a hot topic in physics nowadays. It has the potential advantages of non-volatility, higher storage density, faster read/write times and lower power consumption. Some technical issues as spin injection, transport and manipulation have to be resolved to successfully incorporate spins into semiconductors. In this report the dynamic spin behavior in semiconductors is studied by applying time-resolved magnetization modulation spectroscopy (TiMMS). This technique spin selectively excites electrons and holes with a circularly polarized laser pulse. The resulting spin imbalance returns to equilibrium due to several possible relaxation processes. The spin relaxation is measured by monitoring the magneto-optical response during the first few picoseconds after excitation by means of a second laser pulse.

Two types of structures have been investigated in this report, diluted magnetic semiconductor (DMS) quantum well (QW) structures (Cd_{1-x}MnxTe), and hybrid ferrometal-semiconductor quantum well structures (Fe on top of a GaAs/AlAs QW). Both structures show an increase in spin scattering compared to their non-magnetic counterpart. The influence of temperature, magnetic field, laser power and laser wavelength on scattering has been studied.

TiMMS measurements show that the magneto-optical signal of a bare GaAs QW is affected by a contribution from bulk GaAs. Furthermore, scattering of the electron and hole spin depends on the absorption and therefore on pump excitation energy and power. The spin scattering in the hybrid ferrometal-semiconductor is enhanced compared to the GaAs sample. In the diluted magnetic semiconductors a transverse applied magnetic field enables to distinguish between the contributions of holes, electrons and manganese to the magneto-optical signal and permits precise measurements of the enhanced electron g-factor. The results show that the electron spin relaxation is dominated by the exchange scattering with the Mn ions in the quantum well. Moreover, the electron spin relaxation time decreases as a function of temperature in contrast to what is expected.
Contents

1 Introduction .............................................. 5

2 Spin dynamics ........................................... 7
   2.1 Semiconductor band structure .......................... 7
   2.2 Spin selective excitation ............................... 8
   2.3 Spin relaxation processes ............................... 9
       2.3.1 Hole spin relaxation ............................... 9
       2.3.2 Electron spin relaxation ......................... 11

3 The magneto-optical Kerr effect .......................... 15
   3.1 Introduction ........................................ 15
   3.2 Description ........................................ 15
   3.3 Time-resolved Kerr effect ............................. 18

4 TiMMS ..................................................... 21
   4.1 TiMMS technique ...................................... 21
       4.1.1 Introduction ..................................... 21
       4.1.2 Analysis of TiMMS signals ....................... 22
   4.2 Experimental .......................................... 24
       4.2.1 Experimental setup ............................... 24
       4.2.2 Spectrometer .................................... 25
   4.3 Measurement procedure: basic concepts and interpretation ......... 26
       4.3.1 Time-resolved measurement ...................... 26
       4.3.2 Resonance spectrum ................................ 28

5 Hybrid ferrometal-semiconductors .......................... 31
   5.1 Introduction .......................................... 31
   5.2 Sample description .................................... 31
   5.3 Results GaAs QW sample ................................ 32
       5.3.1 Spectral characterization ........................ 32
       5.3.2 Spin dynamics .................................... 33
       5.3.3 Room temperature spin dynamics ................. 35
       5.3.4 Bulk influence .................................... 36
       5.3.5 Excitation density dependence .................. 38
       5.3.6 Temperature dependence .......................... 39
   5.4 Results Fe-GaAs QW sample ............................ 42
       5.4.1 Magnetic and structural characterization ......... 42
Chapter 1

Introduction

An electron does not only carry charge, but also has a spin degree of freedom. Its orientation can take two values, named spin up and spin down. Spintronics, electronics exploiting the relative spin orientation of the electron in addition to its charge, has become a highly interesting research topic in physics nowadays. This interest arises from the fact that the electron spin can store information, can be transferred as it is attached to mobile carriers and can be detected. In addition, the possibility of having long spin relaxation times in electronic materials hold the promise of realizing spintronic devices in the future.

Future spin devices may have many applications such as a spin-FET, a spin-polarized field effect transistor, which was proposed by Datta and Das [1] that is based on the injection of spin-polarized electrons from a ferromagnetic material into a semiconductor. Currently, the main disadvantage is the low spin injection efficiency, because of the Schottky barrier at the metal/semiconductor interface.

Schmidt and Molenkamp [2] used a dilute magnetic II-VI semiconductor as a spin aligner to inject highly spin polarized electrons into a GaAs light emitting diode. DMS’s are semiconductors with some of the original cations replaced by magnetic ions. The permanent magnetic moment can be used to manipulate spin and greatly enhances the electron g-factor. However, the effect of the magnetic ions is highest at low temperatures and high magnetic fields. For room temperature applications III-V DMS structures appear very promising, as they become ferromagnetic at high temperatures.

To successfully incorporate spins into semiconductors, some technical issues as spin injection, transport and manipulation have to be resolved. In this report the dynamic spin behavior in semiconductors is studied by means of optical spin injection. This method is stimulated by the development of new time resolved magneto-optical techniques, to investigate spin dynamics down to the subpicosecond domain. The applied technique, Time Resolved Magnetization Modulation Spectroscopy (TiMMS), uses a laser beam with a circular polarization that is modulated between left- and righthanded light. Due to the spin-orbit coupling in the semiconductors, electrons and holes with spin up and spin down are alternately excited. The resulting spin imbalance in the material returns to equilibrium by several relaxation processes. With the use of ultrashort optical pulses (100 fs), this photo-induced magnetization is transient and decays as a result of the relaxation processes. The induced magnetization modulation is detected by monitoring the corresponding modulation of the magneto-optical Kerr rotation (or ellipticity) of the sample experienced by a second linearly polarized laser beam with a variable time-delay. The spin behavior is studied by focusing on the spin life times.
and relaxation rates as a function of temperature, laser power, laser energy and magnetic field.

From a fundamental point of view, it is interesting to know the effect of a ferromagnetic layer on the electron spin dynamics in nearby semiconductors [3]. Therefore, TiMMS is applied to a hybrid ferrometal-semiconductor, that consists of a GaAs/AlAs QW with on top a Fe layer (Fe-GaAs). The use of QW structures allows tuning of the band gap and confinement energy. A GaAs QW grown in the same way is measured as a reference sample. Secondly, TiMMS is applied to study a DMS QW structure; Cd$_{1-x}$Mn$_x$Te. The interaction between band electrons and localized Mn states in these materials gives rise to a rich spectrum of magnetic behavior, which can be tuned by varying the Mn concentration. Moreover, DMS structures exhibit giant magneto-optical effects, large Faraday rotation and sharp spectroscopic features at the fundamental band gap, which makes them ideal candidates for dynamic spin behavior investigations.

The general structure of this report is as follows. Chapter 2 describes the band structure of a GaAs QW and a DMS QW, and continues with the optical injection and relaxation mechanisms in these structures. An introduction to the magneto-optical Kerr effect is given in Chapter 3. Chapter 4 presents a combined theoretical and experimental explanation of the TiMMS principle. The results on the hybrid ferrometal-semiconductor QW structure Fe-GaAs and the diluted magnetic semiconductor Cd$_{1-x}$Mn$_x$Te are presented in Chapter 5 and 6. Finally, general conclusions will be drawn and an outlook is given in chapter 7.
Chapter 2

Spin dynamics

This chapter describes the theory needed to interpret the Time Resolved Magnetization Modulation Spectroscopy (TiMMS) measurements. With TiMMS, a short circular polarized pump pulse spin selectively excites electrons and holes. The presence of several relaxation mechanisms will cause the induced spin imbalance to decay. In this report TiMMS is applied to two kinds of samples:

- Hybrid ferrometal-semiconductor quantum well (QW) structures, Fe-GaAs QW, to investigate the coupling between the electron spin in the quantum well and the spins of the ferromagnetic layer.

- Diluted Magnetic Semiconductor (DMS) quantum well structures, Cd$_{1-x}$Mn$_x$Te. This is a II-VI semiconductor, of which some cadmium cations (valence +2) have been replaced by magnetic ions with the same valence, in this case manganese, thereby adding local magnetic moments to the semiconductor.

Both are direct-band gap semiconductors and a simple description of their band structure is given in section 2.1. Next, the principle of optical excitation that generates electron-hole pairs in a semiconductor is explained. The last section is devoted to the spin relaxation mechanisms, by which the unbalanced population of spins is brought to equilibrium.

2.1 Semiconductor band structure

Semiconductors (SC) like GaAs and CdTe have a zinc-blende crystallographic structure of which the band structure is well known. The bottom of the conduction band and the top of the valence band are located at the center of the Brillouin zone (Γ-point). The s-like conduction band is onefold degenerate around the Γ point and has quantum numbers $J = 1/2$ and $m_j = \pm 1/2$. The p-like valence band is threefold degenerate. Due to spin-orbit coupling it is split into a P$_{1/2}$ split-off band and a degenerate P$_{3/2}$ band. The split-off band has quantum numbers $J = 1/2$ and $m_j = \pm 1/2$, and the P$_{3/2}$ band consists of the heavy-hole states with $J = 3/2$ and $m_j = \pm 3/2$ and the light-hole states with $J = 1/2$ and $m_j = \pm 1/2$.

In semiconductor quantum wells the P$_{3/2}$ band is also split. The heavy and light hole states are separated by the confinement. The quantum well forces quantization of the orbital angular momentum in the direction perpendicular to the well (z-axis). Compared to the light holes, the heavy holes have a larger out-of-plane component what is energetically unfavorable.
Figure 2.1: Band structure of semiconductor quantum well structures like GaAs and CdTe near the $\Gamma$ point.

Figure 2.1 shows a schematic picture of the band structure. The use of a quantum well has more advantages, because the band gap is tunable and it leads to sharp spectroscopic features around the band edge.

Absorption of a photon can also lead to the formation of an exciton, which is a bound electron-hole pair. The exciton binding energy for bulk GaAs is 4.7 meV [5] and for bulk CdTe 9 meV [4].

2.2 Spin selective excitation

Since the heavy hole and light hole states are split due to their confinement in the QW, the laser energy can be tuned near the direct band-gap energy such that excitations only take place from the heavy hole band. Part of the photons will be absorbed and optical transitions take place around the $\Gamma$ point between the top of the valence band (heavy holes) and the bottom of the conduction band.

Not all transitions from initial to final state are possible. According to Bohr [6], there is a definite energy associated with each stable orbit, and an atom radiates energy only when it makes a transition from one of these orbits to another. He found that the magnitude of the electron's angular momentum is quantized and that it must be an integral multiple of $h/2\pi$. A photon ordinarily carries off one unit of angular momentum $h/2\pi$ (its intrinsic spin angular momentum corresponds to $s=1$ [7]). The change in angular momentum of the electron must compensate for the angular momentum carried away by the photon. This leads to the requirements that in a transition the orbital angular momentum $l$ must change by 1 and the change in quantum number $\Delta m_j$ is 0 or ±1 [8].

Optical pumping with circularly polarized light increases or decreases the total angular momentum by one, depending on the helicity of the light polarization. When pumping with left-handed circularly polarized light only transitions from $m_j = 3/2$ to $m_j = 1/2$ are possible and thus only electrons with spin up are excited. Pumping with right-handed polarized light only transitions from $m_j = -3/2$ to $m_j = -1/2$ can occur and thus only spin down electrons are excited (figure 2.2). Circularly polarized light, therefore, is used to create an imbalance between spin up and down electrons in semiconductor quantum wells of 100%. In bulk
material where the heavy holes and light holes are degenerated, circularly polarized light will create a spin imbalance of only 50%. For higher photon energies also transitions between the top of the light holes band (or the split-off band) and the bottom of the conduction band are possible. However, the probability of a transition involving light holes is three times smaller than that of a transition involving heavy holes [9]. After absorption of the pump pulse, there are several competing processes on the ultrafast time scale. Thermalization processes take place at a time scale of approximately 1-2 ps [10], redistributing the induced carriers to the top/bottom of the valence/conduction band by electron-electron scattering. For pump frequencies well above the band gap also electron-phonon scattering contributes to the thermalization. Next, the fully spin-polarized electrons and holes will balance their total spin by several scattering mechanisms (ps-ns). Finally, electrons and holes will recombine at a time scale, which is usually much slower than the spin dynamics.

2.3 Spin relaxation processes

Spin relaxation refers to mechanisms that bring an imbalanced population of spin states into equilibrium. Electrons and holes can only lose their spin orientation by interaction with particles, like electrons, holes and phonons. The relaxation will cause the non-equilibrium part of the spin population to decay towards zero. A detailed understanding of the spin relaxation mechanisms is necessary for future device applications.

2.3.1 Hole spin relaxation

Spin-flipping of holes requires a change from $m_j = \pm 3/2$ (heavy holes) to $m_j = \pm 3/2$ (light holes). In bulk materials there is a strong mixing of the heavy- and light holes states near the $\Gamma$ point, leading to a rapid hole spin relaxation. In quantum wells (as studied in the present work), the degeneracy of the light and heavy holes at $k = 0$ is lifted (section 2.1, figure 2.1). After excitation the holes all thermalize to the heavy hole states and the light hole states are not available as they are at a different energy. Spin-flipping is not possible and spin scattering is decreased. However, there are some modified mechanisms that use the fact that at a certain $k$-value, the point of degeneration, the heavy hole states and light hole states will mix up, as depicted in figure 2.3. In practice the states will not cross, but new states are formed in which the heavy holes behave as light holes and the light holes as heavy holes. At these anti-crossing points, the light hole states become available again for scattering processes. Electrons and holes need an energy of a few meV [11] to get at this point, which
may be achieved by raising the temperature. So, most of the relaxation mechanisms based on this degeneracy are more effective at higher temperatures. One such mechanism is the

**Bir-Pikus (BP) mechanism.** The BP mechanism describes the interaction of holes with phonons. When holes flip their spin, the phonons slightly deform the crystal structure, which is described by the Bir-Pikus Hamiltonian. This process is expected to increase in strength at higher temperatures, due to non-zero \( k \)-values (around the point of degeneracy), and the increased amount of phonons. A more detailed description is given by Maialle *et al.* [12].

Another process is the **Bir-Aronov-Pikus (BAP) mechanism**, of which a schematic picture is shown in figure 2.4. In addition to the usual Coulomb interaction in crystals,

**Figure 2.3:** Schematic picture of the band structure of semiconductor quantum well structures like GaAs and CdTe near the \( \Gamma \) point. At nonzero \( k \)-values the heavy -and light hole states become degenerate and they mix up (solid line). In practice new states are formed with rapidly changing properties at this point.

**Figure 2.4:** The Bir-Aronov-Pikus mechanism. The exchange interaction between electron and hole spins causes electron-hole spin flipping. An electron and hole flip their spin at the same time and therefore maintain their total quantum numbers.

there exists also electron-hole exchange interaction, due to the Pauli exclusion principle. This principle states that the total wave function of a two-electron system should necessarily be antisymmetric. Thus, if the electrons have equal spin, the orbital wave function should be antisymmetric, and for opposite spins it has to be symmetric. Consequently, the energy of the
system depends directly on the spin. This can be written in the Heisenberg Hamiltonian [13]:

$$H = -2 \sum_{ij} J_{ij} \mathbf{s}_i \cdot \mathbf{s}_j,$$

(2.1)

in which the constant $J_{ij}$ is the strength of interaction between spin $i$ and $j$, related to the overlap of their charge distributions.

A significant property of the interaction is that it depends on the relative orientation of electron and hole spins and it acts on the spins as an effective magnetic field. The spin relaxation takes place as electron spins precess along this field [14], resulting in a simultaneous flip of electron spin and hole spin, for which the light hole $(m_j = \pm 1/2)$ states are needed. This mechanism plays an important role in semiconductors with a significant overlap between electron and hole wave functions, since the exchange energy is proportional to the integrated overlap of the electron and hole wave functions. The spin relaxation rate is proportionally to the square of the exchange energy $U$ for free electrons and holes: $1/\tau_{e/h} \propto U^2$. This is in agreement with the intuitive picture that the proximity of the electron and hole leads to an increase in the exchange energy and thus faster relaxation times. Moreover, although this mechanism works at non-zero $k$-values, the BAP mechanism will be less effective at high temperatures, because an increase in the temperature results in a weaker electron-hole exchange interaction, compared to the low temperature regime where they are bound in an exciton and their wave functions have a larger overlap. Varying the amount of holes or electrons by doping, increases this relaxation enormously.

Comparison of the Bir-Pikus and the Bir-Aronov-Pikus mechanisms.

The BP mechanism is based on hole-phonon scattering, this process becomes more important at higher temperatures, while the BAP mechanism is most effective when electrons and holes are bound in an exciton. In table 2.1 the main properties of the BP and BAP relaxation processes are given: kind of interaction, dependence on temperature and temperature regime where they are most effective.

<table>
<thead>
<tr>
<th>Relaxation mechanism</th>
<th>Interaction type</th>
<th>Relaxation rate $1/\tau_h$</th>
<th>Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>BP</td>
<td>hole-phonon</td>
<td>$\propto T$</td>
<td>$&gt; 50$ K</td>
</tr>
<tr>
<td>BAP</td>
<td>electron-hole</td>
<td>$\propto U^2$</td>
<td>$&lt; 50$ K</td>
</tr>
</tbody>
</table>

Table 2.1: Relative efficiencies of the Bir-Pikus and the Bir-Aronov-Pikus mechanisms.

2.3.2 Electron spin relaxation

Generally, electrons in semiconductors retain their spin information much longer than holes and are therefore more important for spintronic applications. Three mechanisms are commonly considered for electron spin relaxation in semiconductors: the BAP mechanism, which is already explained, the Dyakonov-Perel (DP) mechanism and the Elliot-Yafet (EY) mechanism. The DP, EY and BAP mechanisms are related to the spin-orbit splitting. Spin-orbit interaction occurs when an electron with non-zero spin moves with relativistic velocities in a static electric field [15]. In the rest frame of the electron the electrical field
transfers into a field that has a magnetic field component. This effective field both affects the dynamics of the spin and the total energy of the electron.

In the Elliot-Yafet mechanism, the lattice ions induce a local atomic electric field, which, via the spin-orbit interaction, will lead to a mixing of wave functions of opposite spin [16]. From the electron point of view, the nucleus is circling around it. This orbiting charge sets up a magnetic field in the electron's rest frame, which exerts a torque on the spinning electron, tending to align its magnetic moment along the direction of the field [8]. Acoustic phonons, boundaries, interfaces or impurity can induce transitions between the opposite spin states, leading to spin relaxation. As a result, in the process of momentum scattering the disorientation of electron spin also becomes possible (figure 2.5). Consequently, the spin flip length will depend linearly on the mean free path (or diffusion constant). The corresponding spin relaxation rate $1/\tau_e$ is proportional to $1/\tau_p$ ($\tau_p$ being the momentum relaxation time). At low temperatures, there are no phonons and the electrical resistivity of the material is dominated by collisions with impurity atoms. Yafet showed [17] that at these temperatures $1/\tau_e$ follows the temperature dependence of resistivity ($1/\tau_e \sim T^5$ at very low temperatures). The EY mechanism is generally considered weak in semiconductors like GaAs [18].

![Figure 2.5: The Elliot-Yafet mechanism. This mechanism is due to the interaction of the spins with the electric field of the nuclei.](image)

The Dyakonov-Perel mechanism is a third scattering process for the electron spins. In crystals without inversion center (such as zinc blende semiconductors), spin-orbit interaction lifts the degeneracy of the conduction band [19] at $k \neq 0$. Electrons with the same wave vector but with opposite spin orientations have different energies [17]). In the reported TiMMS measurements this splitting will not be visible, as it is assumed to be much smaller than the thermal energy $k_BT$. Spin splitting of the conduction band is equivalent to the presence of an effective internal magnetic field (spin-orbit interaction). The spin of an electron precesses along such a field, until the electron momentum changes by impurity, boundary or phonon scattering. Then the precession starts again, but along a different axis, which can be seen as a random walk in spin space. The electron spin relaxation occurs as a result of a number of these accidental small rotations, which appreciably lowers the spin relaxation rate (see figure 2.6). As the spin polarization changes during the precession, the scattering acts against the spin relaxation. The total time that it takes to lose spin information is proportional to $N$, the number of scattering events necessary to reach a broadening of the initial state of spin polarization in the order of $\pi$, multiplied by the time $\tau_p$ between these events. Assuming small random steps, this number for a random walk is $N \propto \left(\frac{\pi}{\Delta \theta}\right)^2$, with the precession angle $\Delta \theta = \omega \tau_p$ and $\omega$ the precession frequency. Accordingly, the DP spin relaxation rate is inversely proportional to the momentum relaxation rate ($1/\tau_e \sim \tau_p$).
2.3. SPIN RELAXATION PROCESSES

Figure 2.6: The D’Yakonov-Perel mechanism. In non-symmetric crystals, spin bands are no longer degenerate. The electron spin relaxation takes place as it randomly changes precession direction and frequency and is independent on the mean free path.

Comparison of the Elliot-Yafet, D’Yakonov-Perel and Bir-Aronov-Pikus mechanisms.

As stated by G. E. Pikus and A. N. Titkov [20] the BAP mechanism is most effective at low temperatures (< 50 K), where the electron-hole exchange interaction is the highest as they are bound in excitons. It is believed that the relaxation rate is almost constant until ± 100 K where the exciton no longer exists. For free carriers, the overlap of wave functions rapidly decreases with temperature, decreasing the exchange interaction. At higher temperatures the EY and DP become dominant. These mechanisms can be easily distinguished from each other: the DP spin relaxation rate is inversely proportional to the momentum relaxation rate, while in the EY mechanism this dependence is opposite. Therefore, when scattering on impurities dominates, the efficiency of the EY mechanism increases and that of the DP mechanism decreases with increasing impurity concentration. For electron scattering by phonons the spin relaxation rate \(1/\tau_e\) for both the EY and DP mechanism does not depend on the impurity concentration and rapidly increases with temperature.

In table 2.2 the main properties of these three mechanisms are summarized. It shows an overview of the temperature and \(\tau_p\) dependence of \(1/\tau_e\), and the temperature regime where a mechanism is most effective.

<table>
<thead>
<tr>
<th>Relaxation mechanism</th>
<th>Relaxation rate (1/\tau_e)</th>
<th>Temperature regime</th>
</tr>
</thead>
<tbody>
<tr>
<td>DP</td>
<td>(\propto \tau_p, \propto T)</td>
<td>&gt; 50 K</td>
</tr>
<tr>
<td>EY</td>
<td>(\propto \frac{1}{\tau_e}, \propto T)</td>
<td>&gt; 50 K</td>
</tr>
<tr>
<td>BAP</td>
<td>(\propto U^2)</td>
<td>&lt; 50 K</td>
</tr>
</tbody>
</table>

Table 2.2: Relative efficiencies of the Elliot-Yafet, D’Yakonov-Perel and Bir-Aronov-Pikus mechanisms.
Chapter 3

The magneto-optical Kerr effect

In the TiMMS experiments, the pump induced spin imbalance is detected by monitoring the corresponding modulation of the magneto-optical Kerr rotation (or ellipticity) of a second pulse, the probe, which follows the pump with a variable time-delay. This chapter gives an introduction to the magneto-optical Kerr effect.

3.1 Introduction

In 1877 The Kerr effect was discovered. If linearly polarized light reflects from the surface of magnetic (thus anisotropic) materials, the polarization state of the light changes. The Kerr effect can be characterized by two separate contributions: Kerr rotation and Kerr ellipticity. The Kerr rotation corresponds to a rotation of the direction of linearly polarized light, while Kerr ellipticity stands for the degree in which linearly polarized light becomes elliptical. The origin of these effects can be explained by regarding linearly polarized light as a superposition of two circularly polarized components with the same amplitude, but with opposite direction [21]. If a material has different reflection coefficients for these two components, then, after reflection, a relative phase shift between them causes their resultant to be rotated with respect to its original direction (Kerr rotation), while a relative change in amplitude between the two reflection components causes their resultant to become elliptical (Kerr ellipticity).

3.2 Description

The reflection of light on a surface is described by the Fresnel reflection coefficients. Writing the complex Fresnel reflection coefficient for right-handed circularly polarized light as \( \tilde{r}_+ = r_+ e^{i\phi_+} \) and for left-handed circularly polarized light as \( \tilde{r}_- = r_- e^{i\phi_-} \), the exact definitions of the Kerr rotation (\( \theta_K \)) and Kerr ellipticity (\( \varepsilon_K \)) are [22]:

\[
\theta_K = \frac{1}{2} (\phi_+ - \phi_-) \quad (3.1)
\]

and

\[
\varepsilon_K = \frac{r_+ - r_-}{r_+ + r_-} \quad (3.2)
\]

In polar geometry, when the magnetization is perpendicular to the sample plane, the relation between the reflection coefficient \( (r_\pm) \) and the complex index of refraction \( (n_\pm) \) is given by:
These definitions show that the Kerr effect exists only if the materials possesses optical anisotropy ($n_+ \neq n_- $).

The magneto-optical effects of an anisotropic material are usually described by the dielectric tensor $\bar{\varepsilon}$. For materials that are axisymmetric around the z-axis, the dielectric constant has to be invariant under a rotation around the z-axis. This means that the elements $\varepsilon_{xz}$ and $\varepsilon_{yz}$ of the tensor have to be equal to each other and $\varepsilon_{xy}$ has to be opposite to $\varepsilon_{yx}$. When the optical anisotropy is due to a magnetization or an applied magnetic field along the z-axis, the elements $\varepsilon_{zx}$, $\varepsilon_{yz}$, $\varepsilon_{zz}$ and $\varepsilon_{zy}$ are zero. The dielectric tensor can thus be written as [22]:

$$
\bar{\varepsilon} = \begin{pmatrix}
\varepsilon_{xx} & \varepsilon_{xy} & 0 \\
-\varepsilon_{xy} & \varepsilon_{xx} & 0 \\
0 & 0 & \varepsilon_{zz}
\end{pmatrix},
$$

where each of the tensor elements is complex and $\varepsilon_{zz}$ is not important for the Kerr effect. The diagonal terms are all even functions of the magnetization. The off-diagonal terms $\pm \varepsilon_{xy}$ are odd linear functions of the magnetic induction of the sample and only non-zero in case of optical anisotropy. This antisymmetric part thus gives rise to the magneto-optical Kerr effect.

The terms of the tensor can be explained by using a quantitative approach that includes spin-orbit interaction. Spin-orbit coupling describes the interaction between the spin and orbital angular momentum and thus connects magnetic and optical properties of materials. In this quantum mechanical calculation all resonances of the electron have to be taken into account, which means that all contributions depend on the quantum transition rates between the electron states. According to Benneman [23], the elements of the dielectric tensor are related to the quantum transition rates as follows:

$$
\varepsilon_{xx} = 1 + \frac{e^2}{\hbar m^2 V} \left( 2 \sum_{\beta} \sum_{\alpha} \frac{|\langle \beta | \sigma_x | \alpha \rangle|^2}{\omega_{\beta \alpha} (\omega_{\beta \alpha}^2 - \omega^2)} + \frac{i \sigma}{\omega^2} \sum_{\beta} \sum_{\alpha} |\langle \beta | \sigma_x | \alpha \rangle|^2 \delta(\omega_{\beta \alpha} - \omega) + \delta(\omega_{\beta \alpha} + \omega) \right),
$$

$$
\varepsilon_{xy} = \frac{e^2}{2\hbar m^2 V} \left( \frac{i}{\omega} \sum_{\beta} \sum_{\alpha} \left( |\langle \beta | \sigma^- | \alpha \rangle|^2 - |\langle \beta | \sigma^+ | \alpha \rangle|^2 \right) \delta(\omega_{\beta \alpha} - \omega) + \delta(\omega_{\beta \alpha} + \omega) \right) + \frac{-\sigma}{2\omega^2} \sum_{\beta} \sum_{\alpha} \left( |\langle \beta | \sigma^- | \alpha \rangle|^2 - |\langle \beta | \sigma^+ | \alpha \rangle|^2 \right) \delta(\omega_{\beta \alpha} - \omega) + \delta(\omega_{\beta \alpha} + \omega) \right),
$$

where $\alpha$ and $\beta$ are summation indices, ranging over the occupied and unoccupied states respectively, $\omega_{\beta \alpha}$ is the angular frequency that corresponds to the energy difference between states $\alpha$ and $\beta$ and $\sigma_x$, $\sigma^+$ and $\sigma^-$ are the momentum operators along the x-direction and for circular motion in the x, y plane. Furthermore V is the interaction volume. The real and imaginary part of both tensor elements are linked to each other by a Kramers-Kröning transformation. If the real part of the tensor element has a delta peak like behavior at the resonance frequency, the imaginary part shows a bipolar resonance structure and vice versa.

While describing the Kerr effect it is convenient to write the dielectric tensor in circular
coordinates rather than in linear coordinates:

\[
\bar{\varepsilon} = \begin{pmatrix}
\varepsilon_{xx} + i\varepsilon_{xy} & 0 & 0 \\
0 & \varepsilon_{xx} - i\varepsilon_{xy} & 0 \\
0 & 0 & \varepsilon_{zz}
\end{pmatrix}.
\]  \hspace{1cm} (3.6)

According to McGee [22] the complex refractive indices \( n_{\pm} \) for right-handed and left-handed circularly polarized light are related to the dielectric tensor by:

\[
n_{\pm}^2 = \varepsilon_{xx} \pm i\varepsilon_{xy}
\]  \hspace{1cm} (3.7)

Combining this equation with the expression describing the relation between the reflection coefficients \( r_{+/-} \) and the complex index of refraction \( n_{\pm} \) together with equations 3.1 and 3.2, an expression for the behavior of the magneto-optical signal in polar geometry can be derived. This expression can be simplified by assuming that the Kerr rotation \( (\theta_K) \) and the Kerr ellipticity \( (\varepsilon_k) \) are small, thus that the off-diagonal elements in the dielectric tensor are small compared to the diagonal ones [22]:

\[
\theta_K - i\varepsilon_K = -\frac{\varepsilon_{xy}}{\sqrt{\varepsilon_{xx}(\varepsilon_{xx} - 1)}}
\]  \hspace{1cm} (3.8)

Once again, we see that if the material does not have optical anisotropy \( (\varepsilon_{xy} = 0) \), then \( \theta_K \) and \( \varepsilon_k \) will be zero and the Kerr effect thus arises from a non-zero off-diagonal element of the dielectric tensor.

By combining equation 3.8 with equations 3.4 and 3.4 the dependence of the Kerr rotation and ellipticity on the light frequency can be evaluated. When the light frequency equals one of the structure’s resonance frequencies, the imaginary part of \( \varepsilon_{xy} \) \( (\varepsilon''_{xy}) \) goes to infinity. The real part of \( \varepsilon_{xy} \) \( (\varepsilon'_{xy}) \), related by the Kramers-Krönigs transformation, is zero at \( \omega_0 \). The ellipticity (imaginary part of equation 3.8) thus looks like a delta peak \( \sim \delta(\omega - \omega_0) \) and the rotation (real part) like a bipolar resonance structure. In figure 3.1 a theoretical prediction of the spectral behavior of the magneto-optical signal at the resonance frequency is shown.

![Figure 3.1: Theoretical calculation of the real (rotation) and imaginary (ellipticity) part of the magneto-optical signal of equation 3.8 at the resonance frequency \( \omega_0 \).](image)
3.3 Time-resolved Kerr effect

In a TiMMS measurement, a left-handed circularly polarized pump laser pulses only excites electrons with spin-up, while a right-handed pulse excites only spin-down electrons, inducing a spin imbalance (see section 2.2). This anisotropy in the sample induces a difference in the complex Fresnel reflection coefficients for right-handed and left-handed circularly polarized light. The linear probe pulse (consisting of these two contributions) therefore experiences Kerr rotation and/or ellipticity. For example, right after excitation by a left-handed pump pulse, the $m_j = -1/2$ level (spin down electrons) will be partially occupied, while the $m_j = 1/2$ level (spin up electrons) remains empty. A large part of the left-handed polarized light of the probe pulse will therefore be reflected, while the right-handed part will be absorbed. In time, spin relaxation removes this imbalance and a part of the left-handed light will also be absorbed until the imbalance is gone and $n_+ = n_-$. 

In the studied samples, the imaginary part of the refractive index is very small and approximately 3 for light frequencies around the band gap. Therefore $\varepsilon_{xx}$ is assumed to be real at these frequencies [24]. Right after arrival of the pump pulse with a photon energy near the band gap, the imaginary part of $\varepsilon_{xy}$ is at a maximum at the pump and probe frequency. The real part of $\varepsilon_{xy}$ is still zero, according to equation 3.5. When the excited carriers start relaxing towards lower energies, the imaginary part decreases while the real part starts to grow. The electrons/holes thermalize to the bottom/top of the conduction/valence band, with a total width of approximately $k_B T$ as in figure 3.2. When this new quasi-stable equilibrium has been reached, the population is almost completely independent of the frequency of the pump pulse. The profile of the imaginary part of $\varepsilon_{xy}$ looks like the new joined density of states. The Kramers-Krönigs transformation gives the real part of $\varepsilon_{xy}$ and is negative below and positive above the band gap energy. The TiMMS spectrum of the Kerr ellipticity (rotation) signal is given by the product of the absorption of the pump beam, which is a broadened step function around the band gap energy, and the imaginary (real) part of $\varepsilon_{xy}$. It thus looks like a Lorentzian, which is an admixture of a delta peak like function and a bipolar resonance structure. Since there is a difference in absorption below and above the band gap energy,

![Figure 3.2: Schematic picture of the QW band structure, showing spin-selective excitation and thermalization of carriers towards the bottom of the band.](image-url)
there is also a difference in amplitude between the negative and positive energy peak of the rotation signal.

Moreover, for very thin layers there are some additional reflections at the backside of the layer and reflections inside the layer that causes a phase shift.
Chapter 4

TiMMS

In this chapter the principles of the TiMMS technique are introduced. Also the experimental setup and procedure are discussed. Finally, the concepts needed to understand the results in chapter 5 and 6 are explained using measurements on a single gallium arsenide quantum well.

4.1 TiMMS technique

4.1.1 Introduction

The general lay-out for a TiMMS experiment is schematically illustrated in figure 4.1. Pump

![Schematic diagram explaining the TiMMS technique. A photo-elastic modulator (PEM) modulates the pump between left ($\sigma^+$) and right-handed($\sigma^-$) circular polarization. This results in a modulation of the magneto-optical signal, which is picked up by a lock-in amplifier.](image-url)

Figure 4.1: Schematic diagram explaining the TiMMS technique. A photo-elastic modulator (PEM) modulates the pump between left ($\sigma^+$) and right-handed($\sigma^-$) circular polarization. This results in a modulation of the magneto-optical signal, which is picked up by a lock-in amplifier.
and probe pulses are generated by a mode-locked laser with a repetition rate $F_{\text{rep}}=82 \text{ MHz}$. The linearly polarized pump beam passes a photo-elastic modulator (PEM) operating at a frequency of $50 \text{ kHz} < F_{\text{rep}}$ so the detected intensities can be considered as quasi-continuous. The PEM with its main axis at $45^\circ$ relative to the linear polarization of the pump, induces an oscillating phase variation between the two polarization components. Its Jones matrix is given by:

$$M(t) = \begin{pmatrix} 1 & 0 \\ 0 & e^{iA(t)} \end{pmatrix},$$

with $A(t) = A_0 \sin(2\pi Ft)$. For $A_0 = \pi/2$ the pump is modulated between left-handed and right-handed circularly polarized light. After focussing onto the sample, a spin imbalance is induced in the material as explained in section 2.2. This leads to a magneto-optical (MO) rotation and ellipticity of the linearly polarized probe beam (chapter 3) that is focussed on the same spot on the sample. The resulting Kerr (in reflection) or Faraday (in transmission) signal oscillates at the same frequency as the PEM and is measured as a function of the pump-probe delay time ($\Delta t$ in figure 4.1) by means of an analyzer, detector and lock-in amplifier. With this technique the time evolution of the induced spin imbalance can be mapped.

### 4.1.2 Analysis of TiMMS signals

The initial electric field of the pump ($E_1$) and the probe beam ($E_2$) are written as two-dimensional Jones vectors [24]:

$$E_1(t) = \frac{E_1(t)}{\sqrt{2}} \begin{pmatrix} 1 \\ 1 \end{pmatrix},$$

and

$$E_2(t) = E_2(t) \begin{pmatrix} 1 \\ 0 \end{pmatrix},$$

A common oscillating phase factor ($e^{i\varphi + i\varphi_\pm \varphi - \omega_\pm t}$) is implicitly assumed throughout the analysis. Using equation 4.1, the transmitted pump field after passing the PEM can be written as:

$$E_1'(t) = [A_+(t) \begin{pmatrix} 1 \\ i \end{pmatrix} + A_-(t) \begin{pmatrix} 1 \\ -i \end{pmatrix}] \frac{E_1(t)}{\sqrt{2}},$$

with $A_\pm(t) = \frac{1}{2}(1 \mp ie^{iA(t)})$, for left handed and right handed circularly polarized light. As explained in chapter 2, spin-selective excitation occurs when a circularly polarized photon is absorbed. The number of absorbed photons is proportional to the pump beam intensity, which is proportional to the square of the absolute value of the electric field. The difference between the amount of excited spin up ($N_+$) and spin down ($N_-$) electrons is then proportional to the difference in intensity of the components of the pump beam with left -and right-handed circular polarization, $N_+ - N_- \sim |A_+|^2 - |A_-|^2 = \sin(A(t))$. This induced imbalance introduces the off-diagonal element in the dielectric tensor, leading to the magneto-optical Kerr rotation of the reflected probe beam at time $t$ due to a pulse at time $t - \Delta t$ can be written as [24]:

$$\theta(t, \Delta t) = \Theta_{MO}(\Delta t) \sin[A(t - \Delta t)],$$

where $\Theta_{MO}$ is a response function, integrated over the duration of the laser, describing the relation between the pump beam intensity and the MO rotation of the probe beam (the signal
in averaged, to exclude variations over the cross section of the beam because of its intensity profile).

The electric field of the probe pulse after interaction with the sample in the limit $\Theta_{MO} \ll 1$ is approximately:

$$\vec{E}_2 = E_2 \left( \frac{1}{\Theta_{MO}} \right)$$

(4.6)

where the Kerr effect $\Theta_{MO}$ is written as: $\Theta_{MO} = \Theta'_{MO} + i\Theta''_{MO}$, according to equation 3.8. The polarization state of the reflected probe beam is measured by using an analyzer set at an angle of $90^\circ - \alpha$ with respect to the probe polarization, which detects the change in polarization due to the Kerr rotation.

Since the signal contains two sine functions ($\sin[\alpha_0 \sin(2\pi F(t-\Delta))]$), the detector signal is expanded in a Taylor series of Bessel functions [24]. The $dc$ signal ($V_{dc}$) and the signal at the fundamental ($V_{1F}$) and double ($V_{2F}$) modulation frequency on the first lock-in amplifier are:

$$\frac{V_{dc}}{V_0} = \sin^2 \alpha + \frac{1-J_0(2A_0)}{2} \cos^2 \alpha |\Theta_{MO}(\Delta t)|^2,$$

(4.7)

$$\frac{V_{1F}}{V_0} = J_1(A_0) \sin(2\alpha) \Theta'_{MO}(\Delta t),$$

(4.8)

$$\frac{V_{2F}}{V_0} = \frac{J_2(2A_0)}{2} |\Theta_{MO}(\Delta t)|^2.$$

(4.9)

Here $J_n$ is the $n$th order Bessel function, and $V_0$ is an overall scaling factor. For a Kerr rotation much smaller than the analyzer angle of $45^\circ$, the ratio between the signal at the (fundamental and second-harmonic) modulation frequency and the $dc$-signal can then be approximated by:

$$\frac{V_{1F}}{V_{dc}} = 2J_1(A_0) \Theta'_{MO}(\Delta t)$$

(4.10)

$$\frac{V_{2F}}{V_{dc}} = J_2(2A_0) |\Theta_{MO}(\Delta t)|^2$$

(4.11)

The $1F$ signal depends almost linearly on the real part of the Kerr rotation, while the $2F$ signal scales quadratic with the magnitude of the complex magneto-optical rotation. For small Kerr rotation angles, this signal is much smaller than the $1F$ signal and contains a significant amount of noise. The $2F$ signal is therefore not measured and equation 3.8 can not be used to calculate the ellipticity.

To be able to measure both rotation and ellipticity, a quarter wave plate (QWP), is inserted between the sample and the analyzer. By choosing $\beta$, the angle of the main axis of the QWP with respect to the initial horizontal probe polarization, $0^\circ$ or $45^\circ$ we select the real (rotation) or imaginary (ellipticity) component of the induced response [25]. The angle of the QWP with respect to the initial probe polarization is taken into account by rotating the axis system with this angle. The Jones matrix for a QWP with $\beta = 0^\circ$ can be written as [26]:

$$M = A \begin{pmatrix} 1 & 0 \\ 0 & i \end{pmatrix},$$

(4.12)

with $A$ some constant. The signal on the detector has changed compared to the signal without QWP. This can be seen as multiplying the electric field of equation 4.6 by $i$. Then $\Theta'_{MO}$ becomes the imaginary part of the Kerr signal, while $\Theta''_{MO}$ is the real part. The $V_{1F}$ signal measured at the detector is now ellipticity instead of rotation.
For $\beta = 45^\circ$, the electric field vector oscillating at an angle $\alpha$ with respect to the initial horizontal probe polarization, has to be converted to one that oscillates at an angle $(\alpha + 45^\circ)$. After the QWP it has to be converted back to the original axis. The total matrix of this operation becomes:

$$M = A \begin{pmatrix} 1 & i \\ i & 1 \end{pmatrix}.$$  \hspace{1cm} (4.13)

The signal on the detector for the QWP at this angle is the same as without QWP and rotation is measured.

## 4.2 Experimental

### 4.2.1 Experimental setup

The experimental setup for the TiMMS measurements is shown in figure 4.2. In the TiMMS experiments pulses from a mode-locked Ti:sapphire laser are used with a repetition rate of 82 MHz and 70 fs pulse duration. At the sample the pulses have a somewhat longer duration, because the setup does not contain a correction for group velocity dispersion. The photon

---

**Figure 4.2:** Schematic description of the setup used. Major components are the laser, the cryostat with the sample, the delay line (retroreflector), the polarizers and the analyzer. For modulation a chopper and a photo-elastic modulator are added.
4.2. EXPERIMENTAL

Energy is tunable from 1.46 to 1.69 eV, while the laser power varies from zero at the spectral limits to 0.8 W at a photon energy of 1.59 eV. The pulses are split with a beam splitter into pairs of pump and probe pulses with an intensity ratio of 5:1. The intensity and polarization of these beams can be varied by a combination of a λ/2-plate and a polarizer. The linearly polarized pump pulses pass a PEM, which introduces a circular polarization with a handedness oscillating at a frequency of $F_1 = 50 \text{kHz}$, well below the laser repetition frequency. There pulses are focussed on the sample with a 500 mm lens resulting in a spot diameter of $\sim 200 \mu\text{m}$ and a laser fluence of up to $5 \mu\text{J/cm}^2$.

A mechanical delay line generates time delays between pump and probe pulses, ranging from 0 to 600 ps. The time delayed probe pulses are focussed on the same spot on the sample. Both pulses are incident within a few degrees perpendicular to the sample surface, which is the same as the growth axis. The pulses are thus incident along the preferential quantization axis of the angular momentum. Spin flip due to spin-selective excitation by the pump beam, as described in chapter 2, requires a change in the quantum number of the spin component in this z-direction, as well as detection of the magneto-optical signal by the probe pulse. The polarization of the reflected probe beam is detected by a combination of a quarter wave plate, analyzer, detector, and a lock-in amplifier as explained in section 4.1.1.

To improve the signal-to-noise ratio, the probe beams are modulated with a mechanical chopper at a frequency of $F_2 = 60 \text{Hz}$. Possible signals due to pump light scattered in the direction of the detector are completely suppressed. Due to this double-modulation scheme, TiMMS is very sensitive and measurements contain almost no noise. The signal on the detector, given by equation 4.10, is the input for the first analogue lock-in amplifier, which gets its reference from the PEM and is set to a time constant $[10] \text{ms} \quad (\gg 1/F_1 \text{ and } \ll 1/F_2)$. Its (analogue) output is used as an input for a second lock-in amplifier, which gets its reference from the mechanical chopper and is set to a time constant of $100 \text{ms} \quad (\gg 1/F_2)$. The output of the second lock-in, proportional to the pump-induced change in probe polarization, goes to the computer system.

To enable measurements at different temperatures (5-300 K), the sample is mounted inside a variable temperature cryostat. A transverse magnetic field can be applied parallel to the sample surface, which is perpendicular to the direction of the laser pulses ($\vec{B} \perp \hat{e}$ and $\vec{B} \perp \hat{z}$, with $\vec{B}$ the applied magnetic field, $\hat{e}$ the growth axis normal to the quantum well plane, and $\hat{z}$ the direction of optical excitation). For Cd$_{1-x}$Mn$_x$Te samples, which have a high g-factor, this can be used to make spin-selectively excited electrons precess around this field. Electron precession, in its turn, will be a very interesting tool in characterizing the spin dynamics.

The setup can easily be modified to measure carrier dynamics, by inserting a polarizer directly after the PEM in the pump beam. The polarizer is set to a crossed orientation with respect to the original pump polarization. When the PEM retardation passes zero (and the pump pulse is thus linearly polarized), the beam is blocked. Only left- and right-handed circularly polarized light will partially be transmitted, leading to an intensity (instead of polarization) modulation of the pump beam at $2F$. The probe beam then measures the change in reflection, related to the number of excited electrons.

4.2.2 Spectrometer

An extra feature of the setup is an enhancement of the spectral resolution of the measurement by placing a spectrometer in the probe beam between the analyzer and the detector. The pulse width in the time domain in this energy range is around 70 fs, which corresponds in
the photon energy domain to a width of 0.01 eV, or 10~15 nm. The exciton binding energy for respectively bulk CdTe and a GaAs QW is approximately 9 meV [4] and 4.7 meV [5] (at 5 K), so the spectral resolution of the laser is not high enough. For this purpose a 15 cm single monochromator with a dispersion of 0.15 mm/nm and a 0.3 mm output slit is placed in the probe beam between the analyzer and the photodiode. The spectral resolution when the monochromator is in the probe beam is about 5 meV (2 nm), compared to 10-15 nm without monochromator. However, the 'uncertainty principle' applied to Gaussian pulses (Fourier transform) learns that the resolution in the time domain ($\Delta \tau_s$) decreases as the spectral resolution ($\Delta \omega_s$) increases according to: $\Delta \omega_s \Delta \tau_s \geq \frac{\hbar}{4\pi}$. Thus although the spectral resolution of the probe is now higher, the resolution in the time domain is lower. Besides the higher spectral resolution, an additional advantage of this setup is that the pump and probe pulses can be tuned separately from each other.

Furthermore, the monochromator selects a certain wavelength band from the laser pulse spectrum. When the monochromator position is set at the center wavelength of the Gaussian laser pulse, the amount of light reaching the detector is at a maximum. Tuning away from this position decreases the light amount, while the Kerr rotation is still the same. All measurements are therefore corrected for the mean intensity of the probe pulse after the monochromator. Another disadvantage is that the intensity on the detector is only 0.3 times the intensity without monochromator.

4.3 Measurement procedure: basic concepts and interpretation

In order to apply TiMMS, first of all a proper alignment is needed. The pump and the probe beam have to be focussed on exactly the same spot. For that purpose it was chosen to use separate lenses for both beams, to better control the focus of the beams. The initial overlap of pump and probe beam is done by eye and usually for a sample whose resonance frequency at a specific temperature is known. When a signal is obtained, the overlap can be optimized by maximizing the magneto-optical signal. Then, the reference sample can be replaced by the sample under investigation. In all experiments the change in Kerr ellipticity is measured (rather than rotation), except when explicitly stated otherwise. To be able to interpret the results described in chapters 5 and 6, first the basic concepts needed to study the dynamic spin behavior in semiconductor quantum wells must be explained.

Since the combined performance of spectroscopic- and time-resolved experiments allows to understand the spin dynamics, a time resolved measurement and a resonance spectrum for a GaAs/AlAs quantum well at a temperature of 5 K are shown (see section 5.1 for the sample structure).

4.3.1 Time-resolved measurement

The time that it takes for an unbalanced population of spin states to come to equilibrium at various temperatures is interesting for a detailed understanding of spin relaxation mechanisms. The life time is determined by scanning the pump-probe delay and looking at the time evolution of the magneto-optical signal. The pump pulse induces the imbalance and by measuring the Kerr rotation with the probe pulse at a certain time delay, the decay of this imbalance is determined. In figure 4.3, a time (delay) scan in the ellipticity configuration at
a frequency around the QW band gap (1.594 eV) is shown, together with a fit. At negative

time delays the probe pulse reaches the sample before the pump pulse does. Because there
is not yet an imbalance induced in the sample, the magneto-optical signal is zero. At zero
time delay, there is an almost instantaneous increase due to the induced spin imbalance by
the pump beam. After some initial dynamics related to the thermalization of electrons and
holes (1-2 ps), the magneto-optical signal starts decaying exponentially towards zero as the
imbalance is brought to equilibrium. From the TiMMS measurement it becomes clear that
there are two contributions to the magneto-optical signal, with approximately the same initial
magnitude. The signal consists of one fast contribution with a time constant of approximately
6 ps and a second slower component with a time constant of 70 ps. The first one is ascribed
to the relaxation of the hole spin, because holes loose their spin orientation generally much
faster than electrons as explained in section 2. The second exponentially decaying contribu­
tion to the signal is identified as the relaxation of the electron spin. The magnitude of
the magneto-optical signal as a function of time after excitation, \( \Psi(t) \), thus consist of two
contributions with approximately the same magnitude, but with a different time constant:

\[
\Psi(t) = A_e e^{-t/\tau_e} + A_h e^{-t/\tau_h},
\]

(4.14)

where \( A_e/h \) represents the amplitude and \( \tau_e/h \) the decay times of the excited electron/hole
spins. The first two picoseconds are not taken into account when determining the time
constant, because off thermalization effects immediately after absorption. A second reason is
that an interference effect is present at zero time delay, between a photon of the pump pulse
and a photon of the probe pulse. Under the influence of the resulting electric field, a third
photon from the pump beam can be deflected in the direction of the reflected probe beam,
which causes a very large signal, with a duration of the correlation function of pump and
probe pulses [24]. A TiMMS measurement in the rotation setup shows the same behavior,
with approximately the same time constant, which will be explained in the next chapter.
4.3.2 Resonance spectrum

To be able to interpret the dependence of the Kerr rotation and ellipticity on the laser pulse wavelength, first photoluminescence (PL) experiments are performed. The photon excitation energy in such experiments is much higher than the band gap energy \( [5] \). By absorption of a photon a electron-hole pair is created. After this excitation process, this pair is not a \( k = 0 \), but somewhere higher in the band. In less than a picosecond, this exciton will lose much of its energy by several relaxation processes (electron-phonon scattering). The electrons/holes relax to the bottom/top of the conduction/valence band and will recombine. Emission takes mainly place from the excited state that is lowest in energy (exciton level). Due to this exciton thermalization, the exciton is often at a lower energy in PL than in absorption (few meV). The energy of the quantum well band gap determined in the PL measurements was 1.57 eV.

So, now the magnitude of the magneto-optical signal is looked at. In section 3.3 a theoretical prediction of the spectral behavior of the magneto-optical signal around the resonance frequency is given. To investigate whether this is true, the magneto-optical signal is scanned as a function of the pump beam energy at a temperature of 5 K (figure 4.4), while the signal is probed 5 ps after arrival of the pump pulse. To distinguish between rotation and ellipticity the \( \lambda/4 \)-plate is respectively set at \( \beta = 0^\circ \) and \( \beta = 45^\circ \). Furthermore, the signal is normalized at the mean intensity of the probe pulse on the detector. The ellipticity signal has

![Figure 4.4: Photon energy dependence of the magneto-optical ellipticity and rotation signal of a GaAs QW at a temperature of 5 K and a fixed time delay of 5 ps.](image)

its resonance extremum at an energy of 1.594 eV. This is just a bit higher than the value of 1.57 eV, found in the PL experiments. The lower energy extremum of the rotation signal is at the same spectral position as the PL value \( [10] \). Due to the exciton thermalization, the PL spectrum is a bit red-shifted compared to the magneto-optical spectrum.

The spectra for rotation and ellipticity obey the Kramers-Krönigs relation, i.e., the rotation signal shows a sign reversal where the ellipticity reaches an extremum, as expected. Right after arrival of the pump pulse with a photon energy near the band gap, \( \varepsilon_{xy}'' \) is at a maximum at the pump and probe frequency, while \( \varepsilon_{xy}' \) is zero. After the first relaxation, the imaginary part decreases while the real part starts to grow and is negative below and positive above the band gap energy. The TiMMS spectrum of the Kerr ellipticity (rotation) signal
is given by the product of the absorption of the pump beam and the imaginary (real) part of $\varepsilon_{xy}$. The ellipticity spectrum thus looks like a delta peak function and the rotation as a bipolar resonance structure. Since there is a difference in absorption below and above the band gap energy, there is also a difference in amplitude between the negative and positive energy peak of the rotation signal.
Chapter 5

Hybrid ferrometal-semiconductors

5.1 Introduction

A large potential of industrial applications is attributed to spintronics, electronic devices that exploit the spin orientation of the electron in addition to its charge. Basic requirements are an efficient injection of spin polarized electrons into a semiconductor and a sufficient life time of the spin information. As an example, Datta and Das [1] proposed a spin-polarized field effect transistor, which is based on the injection of spin-polarized electrons from a ferromagnetic material into a semiconductor. Since this principle is anticipated to work at room temperature without magnetic fields, it would be an ideal candidate for applications.

At the moment a lot of research is done to inject spins into a semiconductor. Spin injection can be done by optical means as explained in this report, by tunnelling of carriers from a ferromagnetic material into a non-magnetic material or by passing current from a ferromagnetic region into a non-magnetic region [15]. The electrical spin injection has not proven to be very successful yet. A problem might be that a metal/semiconductor usually forms a Schottky barrier. The nature of the electronic states at this barrier is poorly understood.

Another interesting research area is to study the interaction between a ferromagnetic layer and the spin polarized carriers in a quantum well, by means of optical injection. In this chapter TiMMS is therefore applied to hybrid ferrometal-semiconductor quantum wells (semiconductors with a ferromagnetic layer on top). By looking at the time evolution of the induced spin imbalance of the excited electrons and holes as a function of temperature, laser power and wavelength, the scattering mechanisms responsible for the relaxation can be distinguished.

5.2 Sample description

A GaAs/AlAs semiconductor quantum well with on top an iron layer to study the effect of the ferromagnetic layer on the spin dynamics of the semiconductor QW, is grown by molecular beam epitaxy at IMEC (Leuven). The use of a quantum well system allows tuning of the band gap and leads to sharp spectroscopic features around the band edge. Furthermore it leads to localization; as electrons and holes are excited in the quantum well, the distance between the carriers and the ferromagnetic layer is almost exactly known.

The sample throughout this report referred to as Fe-GaAs, consists of a 7 nm wide GaAs/AlAs (100) quantum well, which is sandwiched between a 4 and a 10 nm aluminium
arsenide barrier on a GaAs substrate. On top of the quantum well a 5 nm GaAs spacer layer and a 8 nm iron layer are deposited, and finally a 2nm GaAs cap to protect the sample for oxidation (see figure 5.1. To be able to interpret the results correctly a reference sample is grown is exactly the same way, but without the iron layer.

Figure 5.1: Schematic description of a GaAs/AlAs quantum well (sample 1) and a GaAs QW with on top a Fe layer (sample 2).

5.3 Results GaAs QW sample

5.3.1 Spectral characterization

A resonance spectrum shows the magneto-optical response as a function of the laser pulse energy as described in section 3.3. In this spectrum, excited electrons and holes in the QW, bulk, barrier or substrate can be distinguished by the energy at which resonance features appear. For understanding the spin dynamic behavior not only the time evolution, but also the spectroscopic characterization is important. Figure 5.2 therefore shows the photon energy dependence of the magneto-optical ellipticity signal at room temperature and cryogenic
5.3. RESULTS GAAS QW SAMPLE

The signal is probed 5 ps after arrival of the pump pulse. At a temperature of 5 K, the ellipticity signal consists of two features, respectively at an energy of 1.52 eV and 1.594 eV. Since photoluminescence experiments showed that the energy of the quantum well band gap at 5 K is 1.57 eV (see section 4.3.2), the resonance structure at this energy is due to the quantum well. The other contribution to the magneto-optical signal at 1.52 eV has a relative phase difference of 90° compared to the QW signal. Moreover it is reasonably well separated from the QW contribution. Since the QW is grown on a GaAs substrate and the energy of the band gap of bulk GaAs is 1.52 eV [27], this signal is ascribed to the spins of bulk GaAs electrons and holes. At this low temperature, it is believed that the electrons and holes are all in the exciton state.

The spectrum of the magneto-optical signal at room temperature does not show a contribution of bulk GaAs like in the 5 K case, because the band gap energy of room temperature bulk is 1.41 eV, which lies outside the laser range of 1.46 to 1.69 eV.

The resonance extremum of the quantum well feature going from 5 to 293 K shifts gradually from a photon energy of 1.594 eV to an energy of 1.51 eV as shown in figure 5.3. When the temperature is raised the crystal lattice expands. This leads to an increased lattice constant and therefore decreased band gap energy. The resonance features of the magneto-optical spectrum are thus expected to shift down in energy with an increasing temperature.

It was verified that the rotational spectra, related to the ellipticity by the Kramers-Kröning transformation, experience the same shift in energy with temperature.

5.3.2 Spin dynamics

To obtain the spin relaxation time of the induced spin imbalance of bulk and QW carriers at cryogenic temperatures, TiMMS scans are performed at the laser photon energy of the GaAs QW band gap (1.594 eV) and bulk GaAs band gap (1.52 eV). The magneto-optical ellipticity signal as a function of pump-probe delay at 5 K is shown in figure 5.4. To investigate whether carrier dynamics is interfering with spin dynamics in this sample, non-magnetic transient reflection experiments as described in section 4.2 have been performed. As we are only
interested in the spin dynamic behavior of carriers in the QW, the lasers is set at the QW gap. These measurements are also plotted in figure 5.4. The maximum amplitude of the

bulk signal is scaled to the same amplitude as the maximum of the QW signal for better comparison. It is clear that the spins of bulk carriers behave differently than the quantum well carriers what probably has something to do with the confinement of the quantum well, which is not present in bulk GaAs. Moreover, none of these signals goes to zero at long time scales. Neglecting this, the measurements are fitted with two exponents, one fast contribution ascribed to the hole spin decay and a slower contribution of the electron spins. For the QW the hole spins decay in \( \pm 6 \) ps and the electron spins in \( \pm 60 \) ps. For bulk GaAs these values are higher; \( \pm 16 \) ps and \( \pm 200 \) ps.

Next, the influence of recombination on the spin behavior is looked at. It is important to know the electron-hole recombination time to interpret the TiMMS measurements properly, since the measured total imbalance in the sample is the spin imbalance multiplied by the amount of excited electrons and holes. If recombination times are much longer than the spin decay, the magneto-optical signal can be written as in equation 4.14, but when recombination takes place at comparable time scales, the total magneto-optical signal has to be formulated as:

\[
\Psi(t) = (A_e e^{-t/\tau_e} + A_h e^{-t/\tau_h}) \cdot e^{-t/\tau_{rec}},
\]  

in which \( \tau_{rec} \) represents the recombination time.

Recombination of excited QW holes and electrons takes place at a timescale of \( \pm 250 \) ps, while the spin decay constant of the magnetic measurement is approximately \( 60 \) ps. If the effect of the recombination on the time evolution of the spins is included, by applying equation 5.1, the relaxation time of the electron spins becomes \( \pm 80 \) ps. The influence of the
amount of free carriers on the spin relaxation time is thus \( \pm 20\% \). Therefore, all measurements have to be corrected for the recombination times. With this correction, the relaxation time at a temperature of 5 K for QW electrons then becomes 80 ps. The influence on hole spin relaxation is only 2.5 \% and \( \tau_h \) stays 6 ps. The correction for recombination of carriers is assumed throughout this report, but will not be written explicitly anymore.

5.3.3 Room temperature spin dynamics

Room temperature measurements of the dynamic spin behavior are very important, as future application will work at room temperature. Figure 5.5 shows a time resolved scan taken at room temperature and different pump laser energies. The amplitude of the induced time resolved ellipticity signal changes while scanning through the band gap. At a photon energy of the band gap it is at its maximum, while slightly higher and lower energies give smaller amplitudes, as expected from the resonance spectrum. To compare the time evolution of the spins, the maximum amplitude of all signals is scaled to the same amplitude as the maximum of the QW signal.

![Figure 5.5: Time resolved measurements at room temperature and an energy of the pump beam of 1.509 eV (QW band gap), 1.499 eV and 1.496 eV.](image)

Recombination of carriers is of minor importance at room temperature, since the recombination rate was found to be less than 1/1200 ps and will have an influence smaller than 2 \% on the measurement. It might seem strange that recombination does not go faster at higher temperatures. However, at low temperatures almost all excited electrons and holes are at \( k = 0 \) and recombination can take place quite easily. At higher temperatures, the carriers have a higher energy and are not anymore at \( k = 0 \). For recombination to occur \( k \) has to be conserved (\( \Delta k = 0 \)). Therefore recombination goes slower at elevated temperatures (up to \( \pm 150 \) K). At temperatures above 150 K, recombination start to go faster with increasing temperature.

For all laser beam energies, the signal reverses sign at positive time delays and only approaches zero at a longer timescale. This behavior, although much less pronounced, was also observed in the 5 K TiMMS experiment. This makes it impossible to fit these measurements according to equation 4.14. Moreover, there is a significant wavelength dependence of the
magneto-optical signal. The measurement with the laser photon energy set at 1.496 eV (close to the bulk gap) shows the most extreme features. It seems that the magneto-optical signal consists of a fast negative contribution of the quantum well and a slower and smaller positive contribution.

5.3.4 Bulk influence

Model description

Here we propose a model to explain the observed time evolution of the spin imbalance. Apparently the magneto-optical signal consists of the signal due to the spin of electrons and holes of the quantum well, with a reasonable fast relaxation time and another signal with opposite sign, a smaller amplitude and a somewhat longer relaxation time. Combined they form the measured time resolved scans. This phenomenon is probably due to the spins of bulk GaAs electrons and holes. The relaxation time of the electron spins of bulk GaAs at 5 K is approximately 200 ps, which is reasonably slower than the 80 ps found for the quantum well. The bulk component at an energy of 1.52 eV has a relative phase difference of 90° compared to the QW well signal (see figure 5.2). Although the resonance features of bulk and QW seem well distinguished at 5 K, they not entirely separated, as can be seen from the time-delayed signal which experiences a sign reversal in figure 5.5. At higher temperatures, the bulk and QW ellipticity peak broaden in the spectral domain and the mixing of bulk and QW becomes larger. At room temperature the bulk peak has shifted to an energy of 1.41 eV and is not possible to measure due to the laser range (1.46 eV-1.69 eV). Since the signal of the resonance spectrum does not go to zero away from the quantum well energy (figure 5.2), a tail of the bulk peak may still overlap with the QW peak. Thus, even when it is not possible to see a separate contribution of bulk anymore at room temperature, there is still an influence of the bulk electron/hole spins.

Accordingly, the magneto-optical signal can be written as:

\[ \Psi(t) = A_{e}e^{-t/\tau_e} + A_{h}e^{-t/\tau_h} + A_{bulk,e}e^{-t/\tau_{bulk,e}} + A_{bulk,h}e^{-t/\tau_{bulk,h}}, \]

with \( A_{e/h} \) the amplitudes and \( \tau_{e/h} \) the decay times of the excited electron/hole spins of the quantum well, and \( A_{bulk,e/h} \) and \( \tau_{bulk,e/h} \) the amplitudes and the decay times of the bulk electron/hole spins. When the pump laser photon energy is fixed near the QW band gap, some assumptions can be made. First of all, the amplitude of the bulk signal is assumed to be independent of the wavelength and smaller than the QW signal. This is reasonable, since the bulk band gap lies relatively far away from the QW. Secondly, as the relaxation time of bulk hole spins is very fast with a maximum value of 16 ps at 5 K, the hole spins are neglected, while the bulk electron spin relaxation time is assumed to be constant. Furthermore, the relaxation time of quantum well electron spins is constant for a fixed pump energy.

Comparison of model and measurements

To be able to compare the measurements with the proposed hypothesis, the spectrometer is placed in the probe beam. In this way a higher spectral resolution is achieved and it becomes possible to tune the probe and pump beam separately. According to the model, when the pump laser photon energy is fixed at the QW band gap and the probe energy is scanned, only the amplitude of the excited electron and hole spin signal should change, because they have different absorption rates below and above the gap.
Consequently, room temperature TiMMS measurements are performed at one photon excitation energy of the pump pulse (1.509 eV), while the probe pulse measures at 7 different energies ranging from 1.499 eV up to 1.521 eV (figure 5.6). Also a measurement is done without the monochromator by removing the exit slit. In the right figure, these data are fitted according to the model. It seems that there is a significant wavelength dependence of

\[
\begin{align*}
0.7 & \quad 0.6 \quad \ldots \quad 0.5 \quad \ldots \quad 0.4 \quad \ldots \quad 0.3 \quad \ldots \quad 0.2 \quad \ldots \quad 0.1 \quad \ldots \\
-0.1 & \quad 0.0 \quad \ldots \quad 0.1 & \quad \ldots \quad 0.2 & \quad \ldots \quad 0.3 & \quad \ldots \quad 0.4 & \quad \ldots \quad 0.5 & \quad \ldots \quad 0.6 & \quad \ldots \quad 0.7
\end{align*}
\]

\[
\begin{align*}
50 & \quad 100 & \quad 150 & \quad 200 & \quad 1.500 & \quad 1.505 & \quad 1.510 & \quad 1.515 & \quad 1.520
\end{align*}
\]

\[
\begin{align*}
\text{Time after excitation (ps)} & \quad \text{Probe pulse energy (eV)}
\end{align*}
\]

\[
\begin{align*}
A_{\text{QW}} & \quad \text{mean } A_{\text{QW}} & \quad A_{\text{ba}} & \quad \text{mean } A_{\text{ba}} & \quad A_{\text{QW without mono}} & \quad \text{mean } A_{\text{QW without mono}}
\end{align*}
\]

Figure 5.6: TiMMS data on the GaAs QW at room temperature. The left figure shows an experiment performed without monochromator, where the pump and probe measure at the same energy. Also plotted are data of measurement with monochromator at a fixed photon excitation energy of 1.509 eV. The probe pulse measures at 7 different energies from 1.499 eV up to 1.521 eV. The right figure shows the amplitudes of the separate contributions to the magneto-optical signal of QW and bulk GaAs.

the probe beam present, but when the measurements are all scaled to the same amplitude they do not differ a lot. These data are now fitted according to equation 5.2 and results are plotted in figure 5.6. For a measurement with the monochromator at a fixed pump laser photon energy (1.509 eV), the amplitude of the bulk contribution is kept constant, smaller and with opposite sign compared to the quantum well. Also the relaxation time of the bulk electrons is kept constant at a value of ± 200 ps. At room temperature the hole spins are extremely fast and thus not taken into account. The data can be well described using a single value of the quantum well electron spin relaxation time \( \tau_e = 28 \) ps, regardless of the wavelength of the probe beam. Only the amplitudes change because of the different absorption rates below and above the gap. It is also possible to fit the measurements without the spectrometer in the same way. At a photon energy of the pump beam of 1.509 eV, again a time constant of 28 ps is found at room temperature. The model seems right as the mean amplitude of the TiMMS signals measured with monochromator is almost the same as the amplitude of the signal without.
5.3.5 Excitation density dependence

Pump laser wavelength dependence

From figure 5.5, showing different features for different pump beam energies, one may suspect different electron/hole spin dynamics at different wavelengths. To see if this is indeed the case, time resolved RT measurement are performed at three different excitation energies of the pump beam. The electron spin relaxation times found for these measurements (data fitted according to the model), are listed in table 5.1. There is evidently a systematic wavelength dependence of the electron relaxation time. With increasing wavelength (decreasing excitation energy) around the QW band gap energy, the electron spin relaxation time increases.

<table>
<thead>
<tr>
<th>Pump beam energy (eV)</th>
<th>Electron spin relaxation time(ps)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.503</td>
<td>28.6 ± 0.2</td>
</tr>
<tr>
<td>1.509</td>
<td>28.1 ± 0.2</td>
</tr>
<tr>
<td>1.518</td>
<td>27.5 ± 0.3</td>
</tr>
</tbody>
</table>

Table 5.1: Spin relaxation times of the electrons, for three different pump beam energies at room temperature.

Pump laser power dependence

The wavelength dependence observed in table 5.1 is not necessarily an intrinsic effect, but might be due to fluctuations of the absorbed laser power. The laser power changes with the laser wavelength. For each laser there is a specific wavelength that results in the highest laser power. This difference in absorbed laser power could cause a difference in spin relaxation as follows:

- The magnitude of the magneto-optical signal is proportional to the laser power, because an increase in the amount of excited electrons and holes cause an increase in spin imbalance of the same amount. As was observed by Beschoten [28], a larger amount of free carriers increases the spin relaxation rate.

- An increase in laser power can cause heating of the sample and influence the spin relaxation times as they are dependent on the temperature.

Therefore, the electron spin relaxation time as a function of pump beam power at room temperature is plotted in figure 5.7. The pump beam energy is set at 1.509 eV (QW band gap) and its power is varied by a combination of a λ/2-plate and a polarizer (see section 4.2). With increasing pump beam power, the relaxation time of the electron spins decreases.

In the studied wavelength regime (~ 800 nm), the pump laser power is almost constant. So, the observed wavelength dependence (1 ps) can not be explained by fluctuations of the power of the pump pulse. Combining the results of table 5.1 and figure 5.7 leads to the conclusion that the amount of absorption in the sample causes the difference in relaxation, because increasing the photon excitation energy leads to higher absorption. When the wavelength of the pump pulse is set such that it excites electrons and holes somewhat below the band gap of the quantum well, the absorption is smaller than when excitation is done well above the band gap. Thus increasing the absorption by increasing the laser power of the pump beam...
5.3. RESULTS GAAS QW SAMPLE

or by moving the excitation energy to the band gap, yields an increase in carrier density and therefore an increase in spin relaxation rate.

This also explains that there is always a small difference in relaxation times of ± 2 ps, when experiments are performed in the ellipticity or rotation configuration. The laser wavelength is a bit different in both cases, ± 2 nm, leading to a different amount of absorption and thus in relaxation.

5.3.6 Temperature dependence

To reveal the dependencies of the scattering mechanisms on the temperature, the electron and hole spin relaxation as a function of temperature are studied.

Hole spin relaxation times

Holes can change their spin due to electron-hole scattering (BAP) or hole-phonon scattering (BP) (see section 2.3). The BAP mechanism will be more effective at low temperatures (up to 50 K). At low temperatures the amount of excitons will be larger than the amount of free electrons and holes. When carriers are bound in an exciton their wave functions have a larger overlap. This leads to a stronger electron-hole exchange interaction and faster relaxation. The probability of the BP process to occur becomes higher at non-zero k-values and is highest at the anti-crossing point, where the heavy hole states and light hole states are closest to each other. At this point of degeneracy, the light hole states become available again for scattering processes. Electrons and holes need energy (few meV [11]) to get at this point that may be achieved by raising the temperature. So, the BP relaxation mechanism becomes more effective at higher temperatures. The measured relaxation time (τ_h) and rate (1/τ_h) of the spin imbalance of excited holes as a function of temperature is depicted in figure 5.8. The figures show that the hole spin relaxation time is only 6 ps at 5 K and shows a strong decrease upon increasing temperature. At temperatures above 100 K it is not possible to distinguish any contribution of holes to the magneto-optical signal. At 5 K, almost all holes are near k=0.

Figure 5.7: Dependence of the electron spin relaxation time on the pump beam power at RT. The pump laser excitation energy is 1.509 eV.
and spin scattering through electron-hole scattering was thought to be the main relaxation mechanism [12]. However, the electron spin relaxation time was found to be 80 ps at 5 K. For the BAP mechanism the electron and hole spin relaxation time should have been the same. So, this fast relaxation is not only due to electron-hole scattering. Therefore, the BP process already has a large influence in the low temperature and increases relaxation at higher temperatures, due to thermal disorder.

Electron spin relaxation times

The temperature dependence of the decay of the spin imbalance of the excited electrons is more difficult to understand. Pikus and Titkov [20] performed similar experiments. They suggested that the responsible mechanisms (see section 2.3) are the BAP-mechanism (electron-hole scattering), D'Yakonov-Perel mechanism (momentum scattering) and Elliot-Yaffet mechanism (impurity/phonon scattering). The BAP will be most effective at low temperature. Above 50 K the DP process is believed to be the main relaxation mechanism. In these samples, scattering at impurities is negligible compared to scattering at phonons, therefore the DP mechanism is predominant rather than the EY mechanism and rapidly increases with temperature. In this momentum scattering process the electron spin looses its information by randomly changing precession direction and frequency. The spin relaxation rate \(1/\tau_e\) for the DP mechanism in III-V compounds should rise proportional to \(T^3 T_p\), where the power dependence on temperature of the carrier momentum relaxation rate \(T_p\) is taken into account: \(1/\tau_e \sim T^{3+n}\), in which \(n\) is a phenomenological parameter [20]. The temperature dependence of \(n\) is caused by the variation with temperature of the relative efficiencies of electron scattering by phonons, which has the effect of slowing down the spin-relaxation rate at higher temperatures. At lower temperatures the DP process becomes slower and at temperatures below 50K the electron spin relaxation rate deviates from the DP prediction. In this temperature regime there are almost no phonons and the DP process is believed to be smaller than the BAP process.

To check whether this theory also applies for this sample, the electron spin relaxation rate \(1/\tau_e\) and time \(\tau_e\) as a function of temperature are plotted in figure 5.9, together with \(1/\tau_h\)
5.3. RESULTS GAAS QW SAMPLE

\( \tau_h \). At cryogenic temperature the relaxation rate is 1/80 ps and only shows a small increase as the temperatures is raised to 50 K. The exciton binding energy in bulk GaAs is ± 4.7 meV. If this energy was entirely of thermic origin, the temperature involved to get free electrons and holes would be about 54 K. Since there is a sharp increase at approximately 50 K, it indeed seems that electrons and holes are bound in an exciton up to this temperature and relaxation in this regime is most probably due to electron-hole scattering. At temperatures above 50 K the DP mechanism is most important and the spin relaxation rate of the electrons show a sharp increase. Between 50 and 100 K the slope of the experimental curve is the highest, because \( \tau_p \) increases, whereas at higher temperatures where \( \tau_p \) decreases the slope becomes smaller. At room temperature the spin relaxation rate has increased to 1/30 ps. Due to the more intensive electron scattering at lower temperatures (< 50 K), the decrease of \( \tau_p \) causes the efficiency of the DP mechanism also to decrease and starts deviating from the DP prediction. The measurements are in good agreement with the theory.

**Figure 5.9:** Relaxation rate and time of the electron and hole spin as a function of temperature. The dotted lines are a guide for the eye.
5.4 Results Fe-GaAs QW sample

To examine the influence of the ferromagnetic layer on the coherent electron spin dynamics in a gallium arsenide semiconductor quantum well, the Fe-GaAs Qw is studied.

5.4.1 Magnetic and structural characterization

To confirm that there is indeed a ferromagnetic layer on top of the Fe-GaAs sample, a hysteresis curve obtained by MOKE experiments is shown in figure 5.10. The Magneto-optical Kerr Effect is a change in polarization of light when reflected from a ferromagnetic material. For the Fe-GaAs QW sample, the magnetization is measured versus the field applied along the easy axis of the iron layer. For large applied fields, the sample is completely magnetized along the direction of the field. From the obtained hysteresis loop it becomes evident that there is indeed a magnetic layer on top of the sample.

However, there might be interdiffusion of the iron layer into the sample. In a sample where the iron layer was grown directly on top of AlAs, the QW had vanished due to diffusion of the iron layer. Therefore, photoluminescence experiments are performed to see if the QW still exists. PL measurements at 5 K clearly showed a peak at 1.52 eV (GaAs substrate) and a peak due to the QW at 1.638 eV.

5.4.2 Spectral characterization

5 K resonance spectrum

The dependence of the magneto-optical signal of the Fe-GaAs QW sample on excitation energy at a temperature of 5 K is shown in figure 5.11. The signal is probed 5 ps after arrival of the pump pulse. This resonance spectrum looks similar to the spectrum of the GaAs sample in figure 5.2. However, the signal experiences a phase shift of 90° compared to the GaAs sample. The magneto-optical rotation shows a delta peak like function, while ellipticity shows a bipolar resonance structure. The change in phase is due to the iron layer, since reflections at the backside of the layer and reflections inside the layer cause a phase shift (see chapter 3.2). The rotation signal consists of two features, respectively at an energy of 1.52 eV (bulk GaAs) and 1.646 eV (GaAs QW). This is in agreement with the photoluminescence.
5.4. RESULTS FE-GAAS QW SAMPLE

Figure 5.11: Photon energy dependence of the magneto-optical ellipticity and rotation signal of the Fe-GaAs QW sample at a temperature of 5 K and a fixed time delay of 5 ps.

experiments. The PL peak due to the QW at 5 K is 1.638 eV, which is at the same energy as the lower energy extremum of the ellipticity signal. The bulk structure is again reasonably well separated from the QW contribution.

Temperature dependence

The resonance extremum of the quantum well feature going from 5 to 293 K shifts gradually from a photon energy of 1.646 eV to an energy of 1.55 eV as shown in figure 5.12. This is due to the fact that as the temperature is raised, the lattice constant increases due to the expanding crystal lattice and therefore the band gap energy decreases. The resonance features

Figure 5.12: Energy of the extremum of the resonance spectrum plotted as a function of temperature for the Fe-GaAs QW and GaAs QW sample. Also the bulk GaAs contribution is shown.
of the magneto-optical spectrum are thus expected to shift down in energy with an increasing temperature.

The difference in band gap energies between the two samples is caused by somewhat different growing conditions.

5.4.3 Spin dynamics

To obtain the temperature dependence of the spin relaxation time of the induced spin imbalance of QW carriers, TiMMS scans are performed at cryogenic and room temperature (figure 5.13). In these experiments, the change in Kerr rotation is measured, because the magneto-optical signal shows a peak when scanning through the band gap in this configuration. This shift will not influence spin dynamics. According to section 5.3.5, the relaxation times of the decay of the induced spin imbalance will not be different for the rotation or ellipticity configuration, when measured at one excitation energy. The magneto-optical rotation signal as a function of pump-probe delay at 5 K and RT is shown in figure 5.13. The laser photon energy is set at 1.646 eV (5 K QW gap) and 1.55 eV (RT QW gap). To investigate whether carrier dynamics is interfering with spin dynamics in this sample, non-magnetic transient reflection experiments as described in section 4.2 have been performed and are also plotted in the figure.

The TiMMS data at a temperature of 5 K are fitted with two exponents, one with a decay constant of 2 ps ascribed to hole spin relaxation, and one with ± 30 ps ascribed to electron spin relaxation. Recombination of carriers at this temperature is in the order of 100 ps. So it has approximately the same influence on the magneto-optical signal as for the GaAs QW sample; 28% on the electron spin relaxation and less than 2% on the hole spin relaxation. The electron hole spin relaxation then becomes ± 60 ps.

At room temperature the experiments can only be fitted with a single exponent. The holes relaxation is very fast (already 2 ps at 5 K) and not distinguishable anymore. The

Figure 5.13: Time resolved rotation measurement of the Fe-GaAs QW sample, with their fits. One measurement is performed at 5 K with a pump pulse excitation energy of 1.646 eV (left figure) and the other experiment is done at RT and an energy of 1.55 eV (right figure). The maximum amplitudes of the TiMMS data and non-magnetic transient reflection data are scaled to the same value for better comparison.
5.4. RESULTS FE-GAAS QW SAMPLE

electron spins relax in ± 10 ps. Recombination takes places with a decay time of more than 200 ps and is again slower at higher temperatures (see section 5.3.3). Its influence on spin relaxation is negligible.

5.4.4 Influence of FM layer on QW spins

To compare the relaxation of the GaAs quantum well and the Fe-GaAs sample, both their magneto-optical signal as a function of the delay are measured at 5 K and at room temperature (excitation at the QW energy for both samples) in figure 5.14. The MO-signal of the Fe-GaAs sample is much smaller than the GaAs QW sample. A large part of the probe beam reflects at the iron layer and experiences no rotation. The magneto-optical signal of the QW is only due to the small amount of light that is transmitted. Accordingly, the dc-intensity of the signal on the detector is almost the same compared to the signal of the sample without the iron layer, but the magneto-optical signal of the quantum well is much smaller. Therefore, both measurements are normalized to the same value. Moreover, since the iron layer reflects much light, it decreases the absorption of the sample and thus decreases scattering (see section 5.3.5) by an decreased carrier density. This will have a maximum influence of ± 4 ps.

As shown in figure 5.14, adding the iron layer to the GaAs QW indeed increases the spin relaxation rate. For the GaAs QW sample electron spin relaxation times were found of 80 ps at 5 K and 30 ps at RT, which is reasonably slower than the values for the Fe-GaAs sample. As the spacing between the iron layer and the QW is 9 nm, it is assumed that this increase is not caused by a QW wave function that leaks into the metal layer. Furthermore, the amount of increase can also not be ascribed to the absorption of the iron layer. So, there is definitely an influence of the ferromagnetic layer on the spin relaxation in the well.

The influence of the iron layer on the spin dynamics in the GaAs QW can be seen as an additional scattering channel. The electron spin relaxation rate can be written as:

\[ \frac{1}{\tau_e} = \frac{1}{\tau_{e,FM}} + \frac{1}{\tau_{e,intr}} \]

(5.3)
in which the electron spin relaxation rate of the sample \( (1/\tau_e) \) is the sum of the intrinsic electron spin relaxation rate of the GaAs QW \( (1/\tau_{e,\text{intr}}) \) and the additional effect of the ferromagnetic iron layer \( (1/\tau_{e,\text{FM}}) \).

For temperatures ranging from 5 K to room temperature, the electron spin relaxation times of both samples are plotted in figure 5.15, together with the spin relaxation rates. All electron spin relaxation times show a decrease with increasing temperature. The influence of the ferromagnetic layer on scattering increases with temperature. The relative increase in scattering with temperature in the Fe-GaAs sample is therefore larger than for the GaAs sample.

### 5.4.5 Monochromator

To study the effect of a ferromagnetic layer on the spin behavior in more detail, the Fe-GaAs QW sample is also investigated with the spectrometer. Figure 5.16 shows TiMMS monochromator scans performed at a temperature of 5 K and a fixed photon excitation energy of the pump pulse of 1.646 eV. The probe pulse measures at 6 different energies ranging from 1.634 eV up to 1.656 eV. The maximum amplitudes of the signals of the mono scans are scaled to the same value. There is a large difference in relaxation. For the GaAs QW sample the mono scans, when scaled to the same value, where almost exactly alike (see section 5.3.4) and it was possible to fit the measurements according to the bulk theory. However, for the Fe-GaAs QW sample it seems that the spin relaxation times change at a different probe pulse energy. These data can not be fitted according to the bulk approach or any other model whatsoever.

It might be that this is related to some artifact due to the experimental procedure. When the pump beam energy is fixed at the QW band gap and the probe beam measures in discrete steps, the \( V_{1F} \) signal as measured without spectrometer can be written as a summation of

![Figure 5.15: Dependence of the electron spin relaxation time of a GaAs QW \( (\tau_{e,\text{intr}}) \) and Fe-GaAs QW sample \( (\tau_e) \) on temperature. The influence of the ferromagnetic layer \( (\tau_{e,\text{FM}}) \) is also shown. The right figure shows the relaxation rates.](image-url)
5.4.RESULTS FE-GAAS QW SAMPLE

Figure 5.16: The figure shows TiMMS scans performed with the spectrometer on the Fe-GaAs QW sample at 5 K, together with a time scan performed without spectrometer. The pump beam energy is fixed at 1.646 eV, while the probe pulse measures at 7 different energies from 1.634 eV up to 1.656 eV.

These monochromator scans:

\[ V_{1F} = \sum_i \frac{\Delta \omega}{\omega_{\text{res}}} V_{1F,i}, \]

(5.4)

with \( i \) the number of scans, \( \Delta \omega \) the size of the discrete step in probe energy (1.9 nm) and \( \omega_{\text{res}} \) the spectral resolution (2.5 nm). If there is something wrong with the experimental procedure this relationship will not be valid. So, the summation of the data of the above monoscan according to equation 5.4 is plotted in figure 5.17 and compared with the time scan of the measurement without monochromator.

Figure 5.17: The figure shows TiMMS data as measured without spectrometer (\( V_{1F} \)) together with the summation of the mono scans (\( \sum_i \frac{\Delta \omega}{\omega_{\text{res}}} V_{1F,i} \)). The pump beam energy is fixed at 1.509 eV, while the probe measures at 7 different energies from 1.499 eV up to 1.521 eV. The temperature is 5 K.

Apparently there is nothing wrong with the performed experiments, as this is almost a perfect fit. Thus, the TiMMS scans really show a difference in relaxation at different probe
energies and it is not possible to fit the data.

This indicates that the measurements performed without monochromator, do not show the real behavior of the Fe-GaAs QW. So, this sample will not give a quantitative insight in how a ferromagnetic layer influences the spin dynamics, but only a qualitative comparison can be made.

5.5 Conclusions and outlook

We have investigated a GaAs QW and a GaAs QW with a ferromagnetic layer on top. The magneto-optical response from the bare QW was affected by a small signal due to a contribution of bulk GaAs, which could be corrected for. The observed spin dynamics depends on excitation density; for higher pump intensities a faster relaxation of the electron spins is found. This also explains the wavelength dependence of $\tau_e$, since the laser power depends on the wavelength and also because the absorption increases with pump pulse excitation energy. The temperature dependence of this sample is in agreement with the literature. The hole spins loose their spin information by electron-hole scattering and hole-phonon scattering. Relaxation times are in the order of 6 ps at 5 K. With increasing temperature the hole spin relaxation time decreases, as the hole-phonon scattering becomes largest. At temperatures above 100 K the hole spin relaxation is extremely fast and it is not possible to distinguish any contribution of holes to the magneto-optical signal. The electron spin relaxation is due to electron-hole scattering and the DP mechanism. The relaxation time of the electron spins is $\pm 80$ ps at cryogenic temperature and decreases to 30 ps at room temperature. This is explained by the fact that at temperatures up to 50 K electron-hole scattering (BAP mechanism) is of major importance, while at higher temperature the momentum scattering (DP mechanism) increases the spin relaxation rate.

The spin scattering in the hybrid ferrometal-semiconductor (8 nm iron on a GaAs QW) is enhanced compared to the GaAs sample. This increase in relaxation is probably due to dipole-dipole interaction and not due to exchange interaction between electron spins of the well and electron spins of the FM layer. Exchange interaction depends on the overlap of wave functions and since the Fe layer is at a distance of 9 nm from the QW, there is no overlap in wave functions. This sample could only give quantitative information. It seems that the relaxation rate $1/\tau_{em}$ increases with temperature. The spin dynamics of the carriers in the QW is thus indeed influenced by the ferromagnetic iron layer. Suggestions for future samples are that they should be n-doped. With a well chosen doping level (order of $10^{-17}$ cm$^{-3}$), the intrinsic relaxation rate of the QW can be tuned to near the laser repetition time (12 ns) and $\tau_{fm}$ is expected to dominate the relaxation (equation 5.3). Then it is possible to sensitively measure the effect of the ferromagnetic layer on the electron spin dynamics in the quantum well. Moreover, to be sure that the effect is caused by the ferromagnetism, the reference sample should also have an additional metal layer on top, which is non-ferromagnetic but with almost the same dielectric constant as the ferromagnetic layer. Other options are for example a GaAs quantum well sample with a ferromagnetic wedge on top, or samples with varying distances between QW and FM layer. Furthermore, Kawakami et al. [3] have observed a very surprising effect; the spin dynamic behavior in the measured hybrid ferrometal-semiconductors was unexpectedly dominated by hyperpolarized nuclear spins that align along the ferromagnet's magnetization. This is very interesting for future fundamental studies.
Chapter 6

Diluted Magnetic Semiconductors

6.1 Introduction

In non-magnetic semiconductors, the spin relaxation is mediated by the spin-orbit interaction. A direct way to influence the spin relaxation is by doping the semiconductor with magnetic ions. Electron and hole spin relaxation times in these Diluted Magnetic Semiconductors (DMS) quantum well materials are found to be much faster than in their non-magnetic counterparts. Consequently, the permanent magnetic moments can be used to manipulate spin and greatly enhances the electron $g$-factor at low temperatures.

Schmidt and Molenkamp [2] used a dilute magnetic II-VI semiconductor as a spin aligner to inject highly spin polarized electrons into a GaAs light emitting diode. For device applications this is very promising, as the magnetic properties can be controlled and modulated through electric fields. They are not ideal for device applications, since the effect of the magnetic ions in II-VI DMS’s is highest at low temperatures and high magnetic fields. However, they are considered interesting test systems for fundamental spin-dynamical investigations, as localized spins can be introduced and controlled independently. III-V DMS structures appear very promising for room temperature applications, as they become ferromagnetic at high temperatures.

In this chapter, TiMMS is applied to study the spin dynamics of II-VI Diluted Magnetic Semiconductors (DMS) quantum wells, by investigation of the time evolution and $g$-factor of the induced spin imbalance of the excited electrons and holes as a function of temperature, applied magnetic field and manganese concentration.

6.2 DMS theory

The studied DMS material is Cd$_{1-x}$Mn$_x$Te. This is a II-VI semiconductor, where some cadmium cations (valence $+$2) have been replaced by (para)magnetic ions with the same electrovalence ($x$ is the percentage of manganese). Manganese has a half filled 3$d$-electron shell with 5 electrons. All five spins are parallel in this orbital (according to Hund’s rule the total energy is minimal when the spins are aligned), leading to a total spin quantum number of 5/2 [29] [30]. Incorporating manganese (3$d^5$) in Cd$_{1-x}$Mn$_x$Te does not affect the semiconductor band structure, but because of its magnetic moment the electron motion is influenced via the exchange interaction between the manganese 3$d$-electron spins and valence and conduction band electron spins. The coupling between the electron/hole spin of position
$r$ and $S_i$ and the Mn spin $S_i$ localized at $R_i$ can be explained by introducing an Heisenberg type $s$-$d$-Hamiltonian $H$ as [31]:

$$H = \sum_i J(r - R_i)S \cdot S_i,$$

(6.1)

with $J$ the relative strength of interaction between the spins. This effective Hamiltonian acts on the envelope function $\Psi$. The exchange integral $\alpha$, parameter of interaction of electrons and holes with Mn ions can be written as: $\alpha = \langle \Psi | J | \Psi \rangle$.

The range of useful manganese concentration is limited by intrinsic anti-ferromagnetic exchange interactions between nearest neighbors. At low temperatures ($< 20$ K), a fraction of the manganese ions forms pairs with strong anti-ferromagnetic ordering and these blocked manganese pairs produce no magnetic moment. The fraction increases with the amount of manganese in the sample. The remaining “free” ions align in the field $(B)$ according to a modified Brillouin function $B_S$ [32] with an effective temperature that is higher than the real temperature. The magnetization, can accordingly be modelled as [33]:

$$M(B, T) = S g_{Mn} \mu_B N_0 x_{eff}(x) B_S \left( \frac{S \mu_B B}{k_B(T + T_{af})} \right),$$

(6.2)

with the manganese spin $S = \frac{5}{2}$ and the Lande-factor $g_{Mn} = 2$. $k_B$ is the Boltzmann constant, $\mu_B$ the Bohr magneton and $N_0$ the number of unit cells per unit volume (cation density). The anti-ferromagnetic interaction between the Mn spins is taken into account with two empirical parameters $x_{eff}(x)$ and $T_{af}$, which are for Cd$_{1-x}$Mn$_x$Te [34]:

$$x_{eff}(x) = x(0.26 e^{-4.3 x} + 0.73 e^{-6.2 x} + 0.01)$$

(6.3)

and

$$T_{af}(K) = 58x - 150x^2.$$  

(6.4)

In all II-VI DMS materials, the exchange constant has approximately the same value and is in Cd$_{1-x}$Mn$_x$Te $N_0 \alpha \approx 0.22$ eV [35] [36].

The presence of the magnetic Mn$^{2+}$ moments enhance the $g$-factor of the electron spins by the $sp$-$d$ exchange interaction. The electron spins experience an effective $g_{eff}$-factor, which is a sum of an intrinsic factor $g_{in}^*$ and a contribution due to $s$-$d$ exchange interaction $g_{exch}^*$ [37]:

$$g_{eff}^* = g_{in}^* + g_{exch}^*.$$  

(6.5)

The exchange part is proportional to the exchange integral $\alpha$ and the manganese magnetization $M(B, T_{Mn})$, leading to:

$$g_{eff}^* = g_{in}^* + \alpha M(B, T_{Mn})/(g_{Mn} \mu_B^2 B),$$

(6.6)

with $g_{Mn}$ the manganese ions $g$-factor. Consequently, as the exchange field acting on the electrons depends on the manganese magnetization, the electron spin $g$-factor can be modified by varying the amount of manganese, the temperature and the magnetic field (only for high fields, because for small fields $M \propto B$).

Incorporating magnetic moments into the lattice, provides an extra spin-scattering source that can also be tuned by varying the manganese concentration. In electron $s$-$d$ interaction, electrons loose their spin orientation by scattering at a manganese $d$-electron. The relaxation rate is proportional to the volume concentration of manganese substitutions in the structure.
Calculations by Bastard [38] resulted in electron spin relaxation times in the order of 20 ps for a concentration of only 5% manganese in Cd$_{1-x}$Mn$_x$Te, compared to 1 ns of their non-magnetic counterparts. The holes can also change their spin by scattering at manganese ions, via the hole $p$-$d$ exchange interaction [39]. As the light hole states are needed for this mechanism, it becomes more important near the anti-crossing point of the heavy hole and light hole band. Relaxation due to this process increases thus with temperature.

### 6.3 Samples description

In the following experiments, different samples have been studied. All samples were grown by MBE at the Polish Academy of Sciences. Two samples are investigated in this report. A schematic description of structure of the samples is depicted in figure 6.1. The basic structure for both samples consists of a GaAs and Cd$_{0.75}$Mg$_{0.25}$Te layer forming the substrate, on which a buffer layer with the same zinc blende structure as cadmium manganese telluride and cadmium telluride is grown.

The first sample has ten identical quantum wells in order to gain a high signal-to-noise ratio and a Mn fraction $x$ of 4%. These Cd$_{1-x}$Mn$_x$Te QW's are sandwiched between Cd$_{0.85}$Mg$_{0.15}$Te barriers on top of this buffer. Finally a Cd$_{0.85}$Mg$_{0.15}$Te cap is deposited to protect the structure from oxidation.

**Sample 1:**
- 0.5 mm GaAs + 4.2 µm Cd$_{0.75}$Mg$_{0.25}$Te substrate
- 0.3µm Cd$_{0.85}$Mg$_{0.15}$Te buffer
- 10 x (80 Å Cd$_{1-x}$Mn$_x$Te well + 250 Å Cd$_{0.85}$Mg$_{0.15}$Te barrier)
- 0.1 µm Cd$_{0.85}$Mg$_{0.15}$Te cap

The second sample has almost the same structure, but only a single QW and a manganese concentration of 0.5%.

**Sample 2:**
- 0.5 mm GaAs + 4.2 µm Cd$_{0.75}$Mg$_{0.25}$Te substrate
- 0.3µm Cd$_{0.85}$Mg$_{0.15}$Te buffer
- (80 Å Cd$_{1-x}$Mn$_x$Te well + 250 Å Cd$_{0.85}$Mg$_{0.15}$Te barrier)
- 0.1 µm Cd$_{0.85}$Mg$_{0.15}$Te cap

Figure 6.1: Schematic description of the Cd$_{1-x}$Mn$_x$Te DMS sample is shown.
Moreover, these samples will be compared with previous measurements performed by C. Smits [4] on other samples: a bulk CdTe sample, an identical sample to the 4 % sample, only with 7 % Mn and a sample with a slightly different structure containing 10 % Mn in the barrier.

Sample 3: 0.5 mm GaAs + 3.0 μm CdTe substrate
   500 Å Cd₀.₉Mn₀.₁Te buffer
   10 x (CdTe well + 100 Å Cd₀.₉Mn₀.₁Te barrier)
   500 Å Cd₀.₉Mn₀.₁Te cap

6.4 Spectral characterization

Important property for characterization of the quantum wells is the band gap. Table 6.4 gives the band gaps for all measured quantum well structures by their photoluminescence at a temperature of 5 K. The band gap becomes larger with increasing amount of manganese in the quantum well. Next, the dependence of the magneto-optical signal of sample 1 and 2 on excitation energy at a temperature of 5 K is shown in figure 6.2. The signal is probed 5 ps after arrival of the pump pulse. This resonance spectrum looks similar to the previous

<table>
<thead>
<tr>
<th>Sample</th>
<th>E (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>bulk CdTe</td>
<td>1.592</td>
</tr>
<tr>
<td>10 % Mn in barrier</td>
<td>1.609</td>
</tr>
<tr>
<td>0.5 % Mn in well</td>
<td>1.637</td>
</tr>
<tr>
<td>4 % Mn in well</td>
<td>1.709</td>
</tr>
<tr>
<td>7 % Mn in well</td>
<td>1.756</td>
</tr>
</tbody>
</table>

Table 6.1: Energy of the band gap of the quantum well structures, as well as of bulk cadmium telluride, measured via their photoluminescence.

Figure 6.2: Photon energy dependence of the magneto-optical ellipticity and rotation signal of sample 1 and 2 at a temperature of 5 K and a fixed time delay of 5 ps.
spectra of the GaAs and Fe-GaAs sample. The magneto-optical ellipticity signals show a delta peak like function, while the rotation signals show a bipolar resonance structure. The magneto-optical spectra are in agreement with the performed PL measurements, which are always at a slightly lower energy.

6.5 Magnetic field dependence

6.5.1 Distinguishing between contributions

The circularly polarized pump pulse excites electrons and holes in the quantum well with their spins initially oriented normal to the quantum well plane. If a magnetic field is applied perpendicular to the direction of the laser beam, i.e. in the plane of the quantum well, the electron spins precess about the applied field. At low temperatures, the holes are all at the top of the valence band and consequently only the heavy hole states with $m_j = \pm 3/2$ are available for transitions. Therefore, the hole spins are not able to precess about the applied field. The manganese spins are, in the absence of a pump pulse, preferentially aligned along the direction of this magnetic field, where their net magnetization is not visible to the Kerr rotation probe (which measures the $z$ rotation). After spin-selective excitation, scattering of the spin-polarized holes with the manganese moments will induce a net transverse magnetic moment in the manganese system. Therefore, the manganese spins will also start to precess around the applied field. In figure 6.3 a time scan for the sample containing 4% of manganese in the QW with an applied magnetic field of 0.65T is shown. The magneto-optical signal consist of a fast oscillating signal with a frequency of 600 GHz, and a non-oscillating signal. The oscillatory contribution is ascribed to the electron spins with a corresponding $g$-factor of 48. Compared to a 'normal' electron with $g$-factor of two, this extremely high $g$-factor in DMS materials is due to the $sp$-$d$ exchange interaction. The relaxation time of the electron

![Figure 6.3: Magneto-optical signal for the sample containing 4% manganese in the QW at a temperature of 5 K, with application of a magnetic field of 0.65 T perpendicular to the laser beam. The laser photon energy was set at 1.712 eV, the band gap energy of the quantum well. The right figure shows the same time scan at a longer time scale.](image-url)
spin imbalance is 2.6 ps, which is very fast (compared for example with the 70 ps for GaAs at this temperature) and due to the additional s-d scatter mechanism. The non-oscillating part is attributed to the holes and their relaxation time is 1.2 ps. After the MO-signal from the electron and hole spin imbalance is damped out, a weak oscillatory signal remains (figure 6.3. This signal oscillates with a frequency of approximately 20 GHz and is due to the precession of manganese d-electrons, with a \( g \)-factor of 2.

The TiMMS signal of the DMS samples thus consists of three different contributions to the magneto-optical signal; two oscillating signals for the electrons and manganese ions, and one non-oscillating component for the holes:

\[
\Psi(t) = A_e e^{-t/\tau_e} \sin(2\pi f_e t + \phi_e) + A_h e^{-t/\tau_h} + A_{\text{Mn}} e^{-t/\tau_{\text{Mn}}} \sin(2\pi f_{\text{Mn}} t + \phi_{\text{Mn}}),
\]

with \( \Psi(t) \) the magnitude of the magneto-optical signal as a function of time after excitation. \( A_i \) represents the amplitude due to the excited electron/hole spin in the QW and the manganese d-electron spin respectively, while \( \tau_i, f_i, \phi_i \) describe the decay times, frequencies and phase of the oscillating components of the signal. The frequency of oscillation of the electron spins, \( f = \frac{g_e}{2\pi \tau_e^3} B \), determined by the spin splitting in transverse fields, enables very precise measurements of the enhanced electron \( g \)-factor in these magnetic quantum wells (\( g^* \)-factor further denoted as \( g \)-factor).

Consequently, application of a magnetic field enables to distinguish between the electron, hole and manganese contributions and permits precise measurements of the enhanced \( g \)-factor.

### 6.5.2 \( g \)-factor

To check if the magnetic field does not influence the dynamic spin behavior in sample 1, time resolved scans were performed at a temperature of 5 K and at different magnetic fields. The photon energy of the pump beam was tuned to 1.712 eV, the band gap of the quantum well. From the time scans, the precession frequencies for manganese d-electrons and the initially excited electrons are calculated and plotted as a function of the amplitude of the applied transverse magnetic field (figure 6.4). The electron precession frequency follows the relation:

\[
\text{Electron precession frequency (GHz)} = \frac{g_e^*}{2\pi \tau_e^3} B
\]

The pump beam is set at the band gap energy of the quantum well (1.712 eV).

![Figure 6.4: Dependence of the precession frequency of the excited electrons and manganese d-electrons on the amplitude of the applied magnetic field at 5 K for sample 1. The pump beam is set at the band gap energy of the quantum well (1.712 eV).](image)
6.5. MAGNETIC FIELD DEPENDENCE

Brillouin like sample magnetization [40]. The slope of the curves related to $g$ with $g$ given by equation 6.6, is linear in $B$ for small magnetic fields, implying that $g$ is constant. The measured $g$-factor for the manganese $d$-electrons at a temperature of 5 K is 2 and for the electron spins 48. The theory stating that at low temperatures anti-ferromagnetic coupled manganese pairs are blocked (equations 6.6 and 6.2), predicts a value of 54 for the electron spins, while the paramagnetic theory [32] gives a value of 120. Thus it seems reasonable to assume that the manganese ions has formed pairs with strong anti-ferromagnetic ordering and does not contribute to the magnetic moment of the sample.

At high magnetic fields, a deviation from linearity can be observed due to the fact that the exchange contribution to the (effective) $g$-factor saturates with $B$, as the magnetization $M$ does (equation 6.6). In very high magnetic fields ($> 1$ T) the intrinsic contribution $g_{in}$ dominates. In this report only measurements up to 1 T are described, where the effect of the magnetic field is negligible.

6.5.3 Relaxation time

To examine whether the electron and hole spin relaxation times are influenced by the applied field in sample 1, they are plotted as a function of the amplitude of the applied transverse field in figure 6.5. The hole spins relax in approximately 1 ps, while the electrons are somewhat slower, 2.6 ps. With increasing transverse field the relaxation time of the holes decreases. This can be caused by band mixing effects. With the application of a sufficiently large transverse magnetic field, the holes experience a reorientation of their total angular momentum from the growth direction to the field direction. In magnetic quantum wells the strong $sp$-$d$ interaction leads to very large Zeeman energies which rapidly overcome the large heavy hole/light hole splitting for fields of the order of 1 T [40]. The coupling of the bands leads to an increased spin relaxation rate, as explained in chapter 2. The electrons lose their spin also faster, which could be due the fact that as the light hole states become available at these high fields also electron-hole scattering (BAP mechanism) may be more effective. At higher fields up to
4 T a larger effect on spin dynamics is expected. However, with this setup it is only possible to perform measurements at magnetic field up to 1 T. Accordingly, the following experiments have been performed at the same applied magnetic field of 0.8 T.

6.6 Temperature dependence

6.6.1 Band gap

Excited electrons and holes of the QW, bulk, barrier or substrate can be distinguished by the energy at which resonance features appear. For a detailed understanding of the temperature dependent spin dynamic behavior, not only the time evolution is important but also the spectral evolution. With an increasing temperature the band gap energy decreases. The resonance features of the magneto-optical spectrum are thus expected to shift down in energy with an increasing temperature. In previous work performed by C. Smits [4], a difference was found between spin relaxation of exciton bound carriers and free carriers. This was concluded from the observation that measurements performed on sample 1 showed two bipolar resonances, which gave different time constants. To check if there might be a difference for free and bound carriers, the monochromator is also placed in the probe beam, between the analyzer and the detector. The binding energy of excitons is 9 meV and a separation between the exciton level and the bottom of the conduction band is distinguishable when the spectrometer is used.

The temperature dependence of the resonance spectrum for both samples is measured. The energy of the resonance extremum for both samples decreases with increasing temperature. However, the data of sample 1 display some unusual oscillatory behavior as a function of temperature. In figure 6.6 the photon excitation energy dependence of the magneto-optical ellipticity signal of sample 1 is plotted. The spectroscopic features do not only change as a function of temperature, but at the same time display a continuous rotation in the complex plane, which means that the signal is oscillating between the imaginary and real part of the

![Figure 6.6](image-url)
6.6. TEMPERATURE DEPENDENCE

magneto-optical response. The initial Lorentzian spectrum at an energy of 1.707 eV (maximum 1 in the figure) changes in a bipolar resonance curve and back in to a peak at an energy of 1.669 eV (maximum 2 in the figure), when increasing the temperature. This is visualized in figure 6.7, where the energy of the extrema of the Lorentzian curve is plotted as a function of temperature for both samples. The sample with a single QW containing 0.5% manganese shows the expected behavior as the energy needed to excite the electrons in the quantum well decreases with increasing temperature. The other sample also shows a decrease in energy with temperature, but clearly there are some strange jumps. This might be understandable by an interference effect (see figure 6.8). There are two contributions to the magneto-optical signal.

First of all there is a signal of the ordinary reflection magneto-optical Kerr effect. Secondly, there is a contribution of an additional magneto-optical Faraday rotation, due to the trans-
parent CdMgTe buffer, which causes a reflection at the CdMgTe/GaAs interface. This signal experiences an additional magneto-optical Faraday rotation (signal 3 in the figure) that builds up in a cumulative way over 2 times 10 quantum wells before leaving the sample. Therefore, there is a competition between these two contributions to the magneto-optical signal. For the Kerr signal (signal 2 in the figure) an oscillatory response is obtained as a function of the number of quantum wells. The Kerr signal is proportional to \( \sum_n e^{2n\pi k d_{spac}} \), with \( n \) the number of wells, and is maximal when \( 2kd_{spac} = \pi \), with \( d_{spac} \) the distance between two QW’s. The wavelength in the material is approximately 700/3 nm, leading to a maximum in Kerr signal when the total thickness of the layer with the QW’s is \( l \) times 58 nm, with \( l \) is 1, 2, 3 etc.. The magneto-optical response for ten QW’s with a total layer thickness \( d = 330 \) nm is thus close to zero for this structure. Because the magneto-optical reflection signal at CdMgTe/GaAs interface is dependent on temperature and wavelength, the oscillating response as a function of temperature is probably due to interference between this Faraday rotation and the ordinary non-magnetic reflection at the outer surface (signal 1 in the figure).

As an approximation, the optical phase difference, \( \Delta \), between the non-magnetic reflected beam and the beam reflected at the CdMgTe/GaAs interface can be written as the product of the optical path difference \( \Delta_p \) and the wave propagation constant \( k \) as:

\[
\Delta = \Delta_p = (2nD)(\frac{2\pi}{\lambda}),
\]

(6.8)

where \( n \) is the refractive index of the material (± 3 in these kind of structures) and \( D \) is the total thickness of the layer (4.83 \( \mu \)m). Constructive interference (\( \Delta = 2\pi m \)) from successive orders are achieved for \( \lambda \approx 730 \) nm (\( m = 39.7 \)), and for \( \lambda \approx 712 \) nm (\( m = 40.7 \)). The corresponding wavelength change between two orders is thus approximately 18 nm. The measurements indeed show that moving from maximum one to two (\( 2\pi \) phase change), the wavelength changes in the order of 10 - 20 nm. Thus, the period is correct in comparison with the thickness of the layer.

In the case of a single QW, the magneto-optical Kerr reflection will be much larger than the reflection at the CdMgTe/GaAs interface and this transmitted Faraday signal will only contribute twice instead of 20 times. Indeed, the resonance spectrum of the other sample with just a single quantum well and a manganese percentage of 0.5 \% does not experience a phase change. This confirms the hypothesis that the strange temperature dependence of the resonance spectrum of the first sample is indeed caused by interference. We tried to correct the data of the 4 \% sample for this effect, assuming that the measured dielectric constant consists of the real dielectric constant multiplied by a wavelength (and thus temperature) dependent phase-factor. However, it was found difficult to obtain reliable results.

So, due to the interference effects, excitons and free electrons and holes can not be distinguished in the 4 \% sample. Moreover, the energy of the extremum of the resonance spectrum of the 0.5 \% sample moves to lower energies as a function of temperature and does not reveal a difference between free and exciton bound carriers (see also figure 6.2).

Thus, the conclusion by C. Smits [4] that there is a separation between excitons and free carriers, based on the observation that measurements done on sample 1 in the rotation configuration showed two bipolar resonances, which gave different time constants, is not right. Since ellipticity measurements with the monochromater on the single QW sample just showed one Lorentzian peak, probably those two bipolar features were also due to the interference effect. Moreover, in the studied wavelength regime (\( \sim 720 \) nm), the laser power is not constant. So, the difference between the relaxation times of the two features in his
measurements is most probably due to the previous observed (pump laser) excitation density dependence as explained in section 5.3.5. The relaxation time decreases as the excitation intensity increases. The excitation intensity in his measurements was the highest for the lower energy resonance structure and therefore led to a smaller relaxation time. Most likely, when these measurements are repeated at a fixed excitation intensity, the relaxation will be the same.

6.6.2 g-factor

The effective $g$-factor in (weak) magnetic fields is dominated by the $s$-$d$ exchange interaction between excited electrons and the momenta of the manganese $d$-electrons. Thus, as the exchange field acting on electrons depends on the manganese magnetization, the effective $g$-factor is strongly temperature dependent (equation 6.6). Accordingly, the $g$-factor as a function of temperature for sample 1 is plotted in figure 6.9. The $g$-factor of sample 2 is too small to be distinguishable. The $g$-factor is again calculated from time delayed scans, with an applied transverse magnetic field. The photon energy of the pump beam was set at the band gap of the quantum well.

![Figure 6.9: Measured electron $g$-factor as a function of temperature of sample 1. The photon energy of the pump beam was set at the band gap of the quantum well. The dotted line represents the paramagnetic theory [32] and the solid line the anti-ferromagnetic theory according to equation 6.6.](image)

At low temperatures the $g$-factor is very high, approximately 48, due to the high exchange field acting on the electrons. With increasing temperature, the manganese is more randomly ordered and the magnetic moment of the polarized region surrounding an electron decreases. As the magnetization decreases so does the exchange field and therefore the electron $g$-factor decreases. This is in accordance with the Brillouin function, because the electron precession frequency slows considerably at elevated temperatures, which leads to a smaller $g_{\text{exch}}$. At 150 K the electron spin precession is very slow and is not possible to distinguish anymore.

In the figure the paramagnetic and the anti-ferromagnetic Mn pairs theory are also plotted. The $g$-factor deviates considerably from the paramagnetic theory. Because this theory predicts somewhat larger values, the manganese is not entirely paramagnetic. The anti-ferromagnetic
theory is totally in agreement with the measurements. Apparently, the manganese ions indeed have formed pairs with strong anti-ferromagnetic ordering, which do not contribute to the magnetic moment.

Since, the exchange field depends on the Mn magnetization, the $g$-factor can also be modified with a varying amount of manganese (equation 6.6). An increase in manganese increases the magnetization and thus the exchange field, leading to a higher $g$-factor. For the sample with 0.5% of manganese in the QW, it was not possible to derive the effective $g$-factor as it was very small. Both theories predict very small values (15 and 16 at cryogenic temperature), which are hardly visible in the measurement.

### 6.6.3 Relaxation time

To determine the responsible mechanisms for relaxation in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$, a temperature dependent study of the relaxation time of the electron and hole spin is performed. Because recombination of electrons and holes might interfere with the spin relaxation, non-magnetic transient reflection experiments are performed. At all temperatures the recombination time was found to be much higher ($>200$ ps over the complete temperature domain) than the relaxation time. The electron-hole pairs exist long enough not to interfere with the experiments to determine the decay of the spin imbalance. As a first impression, two time scans performed at a room temperature and 5 K for sample 1 are plotted in figure 6.10. At 5 K the hole spins relax in 1.2 ps, while the electron spin relaxation time is 2.6 ps. At room temperature, the hole spin is very fast and it is not possible to distinguish the hole spin contribution to the signal anymore. The electron spins do not precess anymore and there is no reason to apply a magnetic field. Their relaxation time is 6 ps. It becomes clear that the electron spin relaxation slows considerably at elevated temperatures.
6.6. TEMPERATURE DEPENDENCE

Hole spin relaxation

The scattering mechanisms in which holes can flip their spin are the \( p-d \) exchange interaction, the electron-hole (BAP) mechanism and the hole-phonon (BP) mechanism. Hole-phonon and \( p-d \) exchange interaction become more important at higher temperatures due to band-mixing effects. The hole scattering rate \( \frac{1}{\tau_h} \) can be decomposed into those three contributions:

\[
\frac{1}{\tau_h} = \frac{1}{\tau_{e-h}} + \frac{1}{\tau_{h-Mn}} + \frac{1}{\tau_{h-ph}},
\]

in which \( \tau_{e-h}, \tau_{h-Mn} \) and \( \tau_{h-ph} \) represents respectively the relaxation times due to electron-hole (BAP), hole-manganese (\( p-d \) interaction) and hole-phonon (BP) scattering. In figure 6.11 the temperature dependence of the hole spin relaxation is plotted. At all temperatures a very rapid hole spin relaxation is found, between 0 and 1.5 ps. The figure shows that the hole spin relaxation time is only 1.2 ps at 5 K and shows a strong decrease upon increasing temperature. At temperatures above 100 K it is not possible to distinguish any contribution of holes to the magneto-optical signal. At 5 K, almost all holes are near \( k=0 \) and spin scattering through electron-hole scattering was thought to be the main relaxation mechanism [12]. However, the electron spin relaxation time was found to be 2.5 ps at 5 K. For the BAP mechanism the electron and hole spin relaxation time should have been the same. So, this fast relaxation is not only due to electron-hole scattering. The \( p-d \) exchange interaction and hole-phonon mechanism already have a large influence in the low temperature regime and they both increase relaxation at higher temperatures due to a larger thermal disorder.

Electron spin relaxation

The DMS samples have been grown without intrinsic doping what means that they have an \( n \)-type doping of \( 10^{17} \text{cm}^{-3} \) and thus contain, compared to the GaAs QW sample, a lot of impurities. This means that the EY mechanism overcomes the DP mechanism, as it is dependent on the mean free path (section 2.3). Consequently, the electrons can flip their
CHAPTER 6. DILUTED MAGNETIC SEMICONDUCTORS

spin by scattering on manganese (e-Mn) due to s-d interaction, electron-phonon/impurity scattering (e-ph/imp) and electron-hole scattering (e-h). The electron spin relaxation rate $1/\tau_e$ can thus be written as:

$$1/\tau_e = 1/\tau_{e-h} + 1/\tau_{e-Mn} + 1/\tau_{e-ph,imp}$$ (6.10)

In figure 6.12, the electron spin relaxation time as a function of temperature is plotted for both samples, together with data of experiments performed by C. Smits [4] on three other samples: bulk CdTe, sample 3 containing ten quantum wells and 7 % manganese and a sample with ten CdTe wells and 10 % Mn in the barrier. In bulk CdTe no influence of manganese and confinement exists and the electron spin relaxation is due to the BAP and EY mechanism, leading to a time constant of 40 ps at cryogenic temperature. In the samples containing manganese, scattering is greatly enhanced. A Mn percentage of only 0.5 already reduces the relaxation time to 12 ps. This fast relaxation is due to the s-d exchange interaction between the electrons and the Mn ions, and therefore electron-hole scattering and scattering at impurities play a minor role in the Mn doped samples. Moreover, the figure also shows that the scattering depends on the amount of incorporated Mn. Increasing the amount of manganese increases the exchange field experienced by the electrons, leading to even faster relaxation; a 4 % Mn QW has a time constant of 2 ps. The Mn concentration increases with a factor 8, while the relaxation decreases with a factor 6. Taken into consideration that a fraction of the Mn ions has formed anti-ferromagnetic pairs, these ratios are in good agreement.

Contrary to the hole spin (and the electron spin in GaAs), the electron spin relaxation time $\tau_e$ is found to increases with increasing temperature in the high-temperature regime ($T > 50$ K). This is a very remarkable behavior as spin scattering is expected to be enhanced at elevated temperatures for the usual BAP and EY mechanism. Most striking is that even the bulk CdTe sample shows this behavior, implying that the observed effect does not depend
on the incorporated Mn ions. Apparently, the decrease of spin scattering with temperature for these CdTe structures is caused by some other relaxation mechanism.

It was suggested by Y. Ohno [41] [42] to plot the data double-logarithmic as done in figure 6.13. For all samples, the electron spin relaxation saturates below $T = 50$ K. In the

![Image](image-url)

**Figure 6.13:** Electron spin relaxation time of all samples as a function of temperature. The laser beam energy was set at the quantum well band gap. The lines show the temperature power dependence ($\tau_e \propto T^\alpha$).

In the high-temperature regime ($T > 50$ K), the electron spin relaxation time $\tau_e$ increases with increase of $T$, according to a power law as $\tau_e \propto T^\alpha$. It seems that for all samples, the electron spin relaxation temperature dependence is approximately the same. Sample 2 deviates considerably, but this could be due to the fact that measurements were only performed up to 200 K and this might introduce a large error.

The temperature dependence of the electron spin relaxation in terms of $\alpha$ for these five different structures is given in table 6.2. The power of the electron spin relaxation temperature dependence has a constant value of 0.5 for all samples (except the one with Mn in the barrier and sample 2, which is probably not very accurate), and the observed effect indeed does not depend on the amount of incorporated manganese.

<table>
<thead>
<tr>
<th>Structure</th>
<th>$\alpha$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk CdTe</td>
<td>0.49 ± 0.04</td>
</tr>
<tr>
<td>10 % Mn in barrier</td>
<td>-0.19 ± 0.03</td>
</tr>
<tr>
<td>0.5 % Mn in well</td>
<td>0.3 ± 0.03</td>
</tr>
<tr>
<td>4 % Mn in well</td>
<td>0.47 ± 0.03</td>
</tr>
<tr>
<td>7 % Mn in well</td>
<td>0.57 ± 0.1</td>
</tr>
</tbody>
</table>

**Table 6.2:** Temperature dependence of the electron spin relaxation time in terms of $\alpha$ for five different samples.
6.7 Discussion

The relaxation time of the electron spin of bulk CdTe, where no influence of manganese and confinement exists is much larger that the samples containing manganese, where scattering is greatly enhanced due to the s-d exchange interaction between the electrons and the Mn ions and electron-hole scattering is relatively small.

Above 50 K, all samples (except the one with manganese) show a decrease in scattering with temperature, contrary to what was expected. Since the bulk CdTe sample shows the same decreasing relaxation of the electron spin with increasing temperature as the QW's containing Mn ions, there is apparently a mechanism that shows a strong decrease of scattering at higher temperatures and is independent of the manganese concentration. The possibility of the EY, DP or BAP mechanism being the dominant spin relaxation mechanism above 50 K can be ruled out, because these mechanisms become more effective at higher temperatures. None of the stated mechanisms (see section 2.3) does account for the increase in scattering time at higher temperatures.

Measurements performed by Y. Ohno [41] [42] also showed similar behavior of the electron spin relaxation time as a function of temperature. In GaAs (110) QW's (where the DP mechanism is suppressed due to the modified symmetry) the relaxation time of the electron spin increased with temperature contrary to the relaxation in GaAs (100) QW's. Y. Ohno proposed two theoretical possibilities; BAP interaction based on the spin-flip scattering of free electrons in a sea of spin-depolarized holes via exchange interaction, and exciton spin relaxation. In the latter mechanism, the electron and hole bound in the exciton flip their spin simultaneously. The two mechanisms can be distinguished by comparison of the electron and hole spin relaxation times. Ohno found electron and hole spin relaxation times of the same order. In our case the hole spin relaxation time is much smaller than that of the electrons at all temperatures, and therefore relaxation due to the exciton mechanism can be ruled out. However, the BAP mechanism cannot account for the positive temperature dependence either, as the temperature dependence of $\tau_e$ is usually predicted as: $\tau_e \propto 1/T^\alpha$, with $\alpha > 0$ [14].

The observed positive temperature dependence ($\alpha < 0$) cannot be explained by a simple model, but there might be another scenario in which spin scattering is due to exchange interaction: excitonic electron-hole exchange interaction. This principle is based on the fact that as the temperature is raised, the thermal ionization of excitons takes place more rapidly (the exciton lifetime thus decreases with temperature), resulting in a weaker electron-hole exchange interaction. Since the wave functions have a smaller overlap, the electron spin scattering by this interaction with the hole spins is reduced.

The responsible mechanisms for the electron spin relaxation in Cd$_{1-x}$Mn$_x$Te (100) and bulk CdTe, are thus believed to be the temperature independent s-d exchange interaction leading to fast relaxation, and the temperature dependent excitonic electron-hole exchange interaction. This also explains the temperature dependence of sample 3 that contains 10 % Mn in the barrier. At low temperatures, the exciton is almost completely in the well and there is no overlap with the Mn of the barrier, leading to slow relaxation. As the temperature increases, the wave functions of the Mn ions and the electrons in the well start to overlap, increasing the relaxation as observed.

To be able to understand exactly what is happening, a QW without manganese should be grown. Because it contains no manganese, the electron spin relaxation would mainly be due to this exciton behavior. When the electron-hole scattering as a function of temperature of
such a sample is known, it is possible to compare with these measurements and draw some final conclusions.

6.8 Conclusions

Because of the high $g$-factor in Cd$_{1-x}$Mn$_x$Te, an application of a magnetic field enables to distinguish between the separate contributions of holes, electrons and manganese to the signal. Furthermore it permits precise measurements of the enhanced electron $g_{eff}$-factor, which depends on the magnetization of the Mn ions. At the used weak magnetic fields the frequency scales linearly with the applied magnetic field and the effect on the electron and hole spin relaxation times is very small.

At low temperatures the $g$-factor is very high, due to the high exchange field acting on the electrons. With increasing temperature, the manganese is more randomly ordered and the total magnetic moment decreases. When the magnetization decreases so does the exchange field and therefore the electron $g$-factor decreases. This is in accordance with the Brillouin function, because the electron precession frequency slows considerably at elevated temperatures, which leads to a smaller $g_{exch}$.

The $g$-factor deviates considerably from the paramagnetic theory. Because this theory predicts somewhat larger values, the manganese is not entirely paramagnetic. The antiferromagnetic theory is totally in agreement with the measurements. Apparently, the manganese ions indeed have formed pairs with strong antiferromagnetic ordering, which do not contribute to the magnetic moment.

An increase in the amount of manganese increases the exchange field, leading to a higher $g$-factor. For the sample with 0.5 % of manganese in the QW, it was not possible to derive the effective $g$-factor as it was very small.

Hole spins relax relatively fast (0.5-2 ps for 0.5 % Mn in the QW) at all temperatures. Since electron spins relax at a somewhat longer time scale, this fast relaxation is not only due to electron-hole scattering. The $p$-$d$ exchange interaction and hole-phonon mechanism already have a large influence in the low temperature regime and they both increase relaxation at higher temperatures due to a larger thermal disorder. Above 100 K, the hole contribution to the magneto-optical signal can no longer be distinguished.

The electron spin relaxation is very fast due to the $s$-$d$ interaction. As this interaction is increased by incorporating more manganese in the quantum well, the relaxation rate increases. The relaxation time for a quantum well containing 4 % manganese at a temperature of 5 K is 2.6 ps and it increases to 6 ps at room temperature, by which the DP and EY process can be ruled out as the dominant mechanism. It seems that the electrons loose their spin first of all through $s$-$d$ exchange interaction. Secondly, it appears that there exists a mechanism that goes slower at higher temperatures, and is of the same order as the $s$-$d$ interaction. We propose that the excitonic electron-hole exchange interaction [41] causes this anomalous behavior. A future sample should have a quantum well with no manganese at all, to study only the electron-hole scattering. Once the electron-hole scattering as a function of temperature is known, it would be possible to compare with the already obtained data. Then it could be checked if our conjecture is right.
Chapter 7

Conclusions and Outlook

In this report TiMMS is applied to two kinds of samples, hybrid ferromagnetic-semiconductors to study the influence of a ferromagnetic layer on the electron spins in the quantum well, and diluted magnetic semiconductors, which have magnetic moments incorporated in their structure that affect the spin dynamics.

The TiMMS measurements on the GaAs QW sample are understood quite well. It is shown in section 5.3.5 that the relaxation time of the electron spin depends on the absorption and thus on pump excitation energy and power. The ellipticity and rotation configuration give the same relaxation time if the excitation energy is the same. The temperature dependence of this sample, section 5.3.6, is in agreement with the literature. The relaxation time of the electron spins is $\pm 80$ ps at cryogenic temperature and decreases to $30$ ps at room temperature. This is explained by the fact that at temperatures up to $50$ K electron-hole scattering (BAP mechanism) is of major importance, while at higher temperature the momentum scattering (DP mechanism) increases the spin relaxation rate. Hole spin relaxation is much faster, in the order of $6$ ps at $5$ K. With increasing temperature the hole spin relaxation time decreases due to the band-mixing effect, that increases the BAP and BP scattering. Above $100$ K, the hole contribution to the magneto-optical signal can no longer be distinguished.

The spin relaxation in the hybrid ferrometal-semiconductor (8 nm iron on a GaAs QW) is faster than in the GaAs sample. However, because it was not possible to fit the measured data accurately enough, this sample could only give qualitative information. Suggestions for future samples are: n-doped GaAs samples, samples with a ferromagnetic wedge or a sample with varying distances between QW and FM layer. Moreover, to confirm that the observed effect is caused by the ferromagnetism, a reference sample with a non-ferromagnetic layer should be grown. The observation of the effect that hyperpolarized nuclear spins align along the ferromagnet's magnetization [3] is very interesting for future fundamental studies.

In the diluted magnetic semiconductors a transverse applied magnetic field enables to distinguish between the contributions of holes, electrons and manganese to the magneto-optical signal and permits precise measurements of the enhanced electron $g$-factor (section 6.5). The hole spin relax very fast. For a sample with 0.5 % Mn in the QW $\tau_h$ varies from 0.5 ps to 2 ps. Relaxation is due to the $p-d$ exchange interaction, BAP and BP mechanisms (section 6.6). Hole-phonon and $p-d$ interaction decrease the hole spin relaxation time at higher temperatures. It is again not possible to distinguish any contribution of holes to the magneto-optical signal above $100$ K.

Due to $s-d$ interaction, the electron spin relaxation is very fast in the magnetic QW's.
Increasing the amount of manganese, increases this interaction and thus the electron spin relaxation. The relaxation time for a quantum well containing 0.5 % manganese at a temperature of 5 K it is 13 ps, while 4 % Mn leads to 2.6 ps (section 6.6.3). As a function of temperature the relaxation decreases. The time constant of the 4 % sample increases to 6 ps at room temperature. It seems that the electrons loose their spin first of all through $s$-$d$ exchange interaction. Secondly, it appears that there exists a mechanism that goes slower with increasing temperature, and is of the same order as the $s$-$d$ interaction. It is proposed that this behavior is probably due to the excitonic electron-hole exchange interaction [41]. A future sample should have a quantum well with no manganese at all, to study only the electron-hole scattering. Once the electron-hole scattering as a function of temperature is known, it would be possible to compare with the already obtained data, to check this proposal.
Chapter 8

Technology Assessment

Conventional electronics rely on the charge of electrons for storage and transport of information. Electronics exploiting the relative spin orientation of the electron in addition to its charge, has become a highly interesting topic in physics nowadays. This research is important for commercially applications and for fundamental studies.

8.1 Commercial applications

The discovery of the giant magneto resistance (GMR) in 1988 marks the beginning of this new field called spintronics. A spin-valve, a GMR-based device, consists of a thin non-magnetic layer sandwiched between two ferromagnetic layers. The electrical resistance strongly depends on the relative alignment of the ferromagnetic layers, which can be changed by an external applied magnetic field. As the magnetization changes from parallel to antiparallel alignment, the resistance rises from 5 to 10 % [43]. The first large scale industrial application of the giant magneto resistance (GMR) devices was as magnetic field sensors in read heads of magnetic recording discs.

In 1995 Moodera et al. [45] fabricated the first magnetic tunnel junctions (MTJ), in which two ferromagnetic electrodes are separated by a thin insulator layer. Electrons have a finite probability to tunnel across the insulator. In the same way as the spin-valve resistance, the tunnelling resistance is modulated by the magnetic field. Since the tunnelling current density is usually small in MTJ’s, they have high resistances that rises from 20 to 40 %.

There are various applications of GMR and TMR based devices. One of the most promising is for non-volatile memory storage. Non-volatile means that information stored is not lost when power is removed from the system [44]. Magnetic random access memory (MRAM) has the potential to be a competitor of existing CMOS based FLASH memories. The memory element in MRAM’s is a magnetic element that can be written by applying a current and can be read by measuring the resistance. Advantages are a lower power consumption, higher storage density and fast read/write times combined with the non-volatility. At present MRAM’s are more expensive, slower and has lower storage density, but they have potential. TMR is technological more likely to be used for MRAM’s than GMR.
8.2 Future devices

Both GMR and TMR are two-terminal devices. The Johnson transistor, a four-terminal device for combined carrier and spin transport has been proposed [46]. This device consists of a paramagnetic layer sandwiched between to ferromagnetic layers. A battery is connected to the paramagnetic layer and one of the ferromagnetic layers. The output voltage is measured between the second ferromagnetic layer and a non-magnetic counter-electrode. The magnetic field dependent charge build up in the gate shifts the transistor threshold voltage, which can be used to measure magnetic fields [47]. Thus, although a four terminal device, it does not allow direct electric control, but relies instead on the application of an external magnetic field.

Datta and Das [1] suggested another device, the so called spin-polarized field effect transistor. This spin-FET device consists of a two dimensional electron gas (2DEG) with ferromagnetic electrodes (source and drain). The asymmetric QW induces a Rashba-type [15] of spin orbit interaction, which will induce spin precession. By application of an external gate voltage, the (a)symmetry and thus the spin-orbit coupling can be varied. The spin precession can therefore in principle be controlled by the gate. It seems feasible to make a 'spin-device' with a field effect transistor-like behavior, in which the total channel conductance will depend on the applied gate voltage.

Future spin devices may have many uses, but probably the most interesting device applications are not discovered yet.

8.3 Fundamental studies

From a fundamental point of view there are several research areas. The first one, is the search for 100% spin polarized materials. Second area is that of spin injection and third the understanding of spin transport in semiconductors. Spin injection can be done by optical means as explained in this report, by tunnelling of carriers from a ferromagnetic material into a non-magnetic material or by passing current from a ferromagnetic region into a non-magnetic region [15]. Most difficult, but perhaps most important case, is the electrical injection of spins into a semiconductor. This has not proven to be very successful yet. A problem might be that a metal/semiconductor usually forms a Schottky barrier. In comparison with a tunnelling barrier, the nature of the electronic states at this barrier is poorly understood.

In this report, the dynamic spin behavior of two kind of structures is studied. In addition to the Datta and Das spin-FET, a hybrid ferrometal-semiconductor quantum well (QW) structure, Fe-GaAs QW, is examined in chapter 5 to investigate the coupling between the ferromagnetic layer and the spin polarized carriers in the quantum well. Since this principle works at room temperature and low magnetic fields, it is an ideal candidate for applications. A disadvantage as mentioned above is the currently low spin injection efficiency.

Schmidt and Molenkamp [2] used a dilute magnetic II-VI semiconductor as a spin aligner to inject highly spin polarized electrons into a GaAs light emitting diode. The permanent magnetic moments can be used to manipulate spin and greatly enhances the electron $g$-factor as done in chapter 6. For device applications however, this is not the best candidate as the effect of the magnetic ions is highest at low temperatures and high magnetic fields. For room temperature applications III-V DMS structures appear very promising, as they become ferromagnetic at high temperatures.

Other materials combining properties of ferromagnets and semiconductors are the real
ferromagnetic semiconductors like europium sulphide, with a Curie temperature of 16K. As this material has different barrier heights for spin-up and down, it can be used to spin-selectively inject spins into semiconductors. First studies of this material are now performed in different groups.
Chapter 9

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