Coherent control of electron spin dynamics in nano-engineered semiconductor structures
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Chapter 1

Introduction
1.1 Electron spin and polarization of light

The electron spin and polarization of light belong to the most fundamental concepts that are used in the physical description of materials and radiation, and both have been intensively investigated ever since their discovery. Nevertheless, our understanding of the behavior spin and radiation—and their interaction—in materials and devices is far from complete, and modern technology does not yet use the full potential they offer. This thesis presents research that is part of a broad field that simultaneously aims at improving this fundamental understanding and realizing proof-of-principle demonstrations of new control schemes and functionalities that better exploit the electron spin and light polarization.

As often happens, the formal description and the name for spin and light polarization came later than the first experiments which could not be explained without these concepts. The name "polarization" for light was introduced by Etienne Louis Malus in 1808, while the first scientific description of an effect due to light polarization (double refraction) was already published by Danish mathematician Rasmus Bartholin in 1669 [1]. The current understanding of the phenomenon is formalized together with the electromagnetic theory of light. The polarization of electromagnetic waves is very widely used. However, optical communication mostly still uses the detection of light intensity, regardless its polarization. This means that in the ultimate limit of such an approach we can transmit one bit of information with one photon successfully transmitted through a network. It is a great achievement compared to what man could do a hundred years ago, but if the polarization states of the photon would be included, each transmitted photon could carry as many bits as one would be able to discriminate different polarization states. This can give a huge increase in the throughput of the network without an increase in the modulation frequencies.

The concept of spin was proposed by Uhlenbeck and Goudsmit [2, 3] in 1925, while the first experiments which needed the spin for their explanation include the observation of the anisotropic magnetoresistance [4] in 1857, and the famous experiment by Stern and Gerlach [5] in 1922. In the widespread information processing with electronics, the role of spin in electrical signals is similar to the role of polarization in optical signals. Here, information is mostly associated the presence or absence of charge or an electron current, regardless of its spin. The utilization of the intrinsic property of electron spin as being a superposition of two states would upscale one bit of useful information for a transmitted electron to a continuum. The practical limitation would be set by the ability of the detector
1.2 Motivation

The quantum description of the interaction of light with matter connects these two concepts. An optical photon in a coherent superposition of polarization states is highly suited for transferring quantum information, while it can also have a selective coupling to the different spin states of conduction electrons in solid state. Altogether this makes this pair (optical photons and electron spins in solid state) a very promising candidate for the realization of new information processing schemes that have been proposed, such as spintronics and quantum information processing [6].

1.2 Motivation

The research presented in this thesis aimed at improving the understanding of, and abilities to control the evolution of spin ensembles in solid state systems. Various fundamental aspects of spin ensemble evolution like dephasing, decoherence, relaxation, diffusion and drift are addressed. All experiments use GaAs or GaAs/AlGaAs-based semiconductor materials with optical control and readout. These GaAs-based materials provide interesting test systems since they combine the highest available material quality, a very advanced level of possibilities for device processing and engineering, and strong optical transitions across the semiconductor bandgap that obey clear selection rules with respect to optical polarization and spin.

Such research with GaAs materials has for this reason already a rich history that goes back to the early days of advanced semiconductor processing about 50 years ago. Optical studies addressing spin were already carried out from the start, with very impressive results that were for example reviewed in the authoritative (and still widely cited) book *Optical Orientation* in 1984 [7]. Around 1995, the development [8] of the pulsed optical technique Time-Resolved Kerr Readout (TRKR, based on the magneto-optical Kerr effect) led to the discovery [9] that GaAs materials at the metal-insulator transition (due to a level of $n$-doping of about $2 \times 10^{16}$ cm$^{-3}$) could show electron spin dephasing times in excess of 100 ns (about 2 orders of magnitude longer than reported till then). Around the same time new schemes for spintronics and quantum information processing were discovered [10]. Together, this stimulated further research aimed at understanding how engineering and controlling the electronic states and electron motion in GaAs materials and device structures could be used for applying coherent electron spin dynamics with a long coherence time, or even spin manipulation via interactions...
with the material [10].

The work that is reported in this thesis aimed at answering questions and realizing proof-of-principle demonstrations at the forefront of this field. The mentioned TRKR technique is central in most of the experiments. In the course of this work, several experiments led to observations that pointed out that the fundamental understanding of the underlying physical mechanisms was—despite the large body of earlier work—in fact incomplete, or even to observations that were in conflict with the established descriptions. On these occasions the reported work aimed at characterizing and understanding these new observations. The remainder of this Section summarizes the questions that underly the work that is reported in this thesis. We used a variety of materials and device structures, with electron ensembles in the form of free bulk populations (3D) and localized donor electrons (0D). We also used materials with a two-dimensional electron gas (2DEG) in a quantum well, that were for some studies processed into quasi-1D wire structures.

![Figure 1.1](image.png)

**Figure 1.1:** Typical example of a Time-Resolved Kerr Rotation trace, with Kerr rotation angle $\theta_K$ (representing electron spin orientation) as a function of pump-probe delay $t$. The oscillatory character directly reflects spin precession about the applied magnetic field.

**Application of the TRKR technique**

The mentioned Time-Resolved Kerr Readout (TRKR) technique is a stroboscopic method that directly measures the electron spin orientation along a certain direction with a time-resolution (about 1 ps) that is smaller than the timescales that govern the electron spin dynamics in semiconductors. The relevant processes here include spin precession in a magnetic field, spin dephasing, and electron-hole recombination. It is an optical pump-probe method with picosecond laser pulses that are tuned near resonance with transitions across the bandgap. The pump
1.2 Motivation

A pulse induces electron spin orientation along a certain direction in a sample. After a controlled delay $t$ a probe pulse is reflected on this sample and the amount of polarization rotation (angle $\theta_K$) that occurs during this reflection event is a direct measure for the amount of spin orientation at that moment. Typical results of such a TRKR experiment are shown in Fig. 1.1, and results of $\theta_K$ as a function of delay $t \geq 0$ are typically fit with the equation

$$\theta_K = A \exp\left(-\frac{t}{\tau}\right) \cos\left(\frac{|g|\mu_B B}{\hbar} t + \phi\right).$$

In this expression $A$ is the initial amplitude, $|g|$ is the $g$ factor of the electrons, $\mu_B$ is the Bohr magneton, $B$ is the magnitude of the external magnetic field, $\hbar$ is the reduced Planck constant, and $\tau$ is the signal decay time. The cosine factor directly reflects spin precession about the external magnetic field at the angular Larmor frequency $|g|\mu_B B/\hbar$, with an initial phase $\phi$.

The format of TRKR signals as in Eq. 1.1 can be used to illustrate the potential of this method, while highlighting at the same time examples where fundamental understanding appeared less complete than we anticipated at the onset of this thesis work. There is in principle a wide range of knowledge available about the variation of $|g|$ and $\tau$ as a function of material composition, quantum confinement, kinetic energy, donor concentration, etc. This suggests that analysis of the observed values for $|g|$ or $\tau$ can be used to obtain time-resolved information about –for example– the location and displacement of electron ensembles in device structures made of layers with different $g$ factors, such as GaAs/AlGaAs heterostructures. Our attempts along these lines were successful [11, 12], but also pointed to gaps in our basic understanding.

A first example of a fundamental aspect that is not fully understood is the initial phase $\phi$ of the electron spin precession in Eq. 1.1. The conventional description of the TRKR method states that it prepares and detects spin orientation along the propagation direction of the laser pulses [9], which is typically identical for the pump and probe beams in a TRKR experiment. However, fitting of experimental results often requires using a non-zero value for the parameter $\phi$. This is widely applied, while it is often not clear why the amount of spin orientation does not show a maximum at the moment of preparation ($t = 0$). We could extend such initial observations of nonzero $\phi$ values to a study in which we could fully control arbitrary $\phi$ values, in work on ensembles of localized electrons in GaAs samples with Si doping at very low concentration (donor-bound electron ensembles, $n_{Si} < 10^{15}$ cm$^{-3}$).
A second example where the field did not yet provide a conclusive picture concerns the spin dephasing time for localized electrons in this type of material. The experimental reports differ from each other by as much as two orders of magnitude [13] and this indicates that it is for many experiments difficult to point out which electrons (free photo-induced electrons versus localized electrons) dominate the signal, and that results can depend on the probing method. With TRKR we looked into this with photon-energy and pump-intensity dependant measurements of the spin dephasing time.

A third aspect that turned out to be not fully established concerns the value of the g factor in Eq. 1.1. For example, for bulk GaAs, nearly all of the established literature and textbooks report $-0.44$ for the GaAs g factor, and the fact that it can obtain values closer to zero as a result of band filling, quantum confinement or excess kinetic energy. However, we observed with TRKR values as negative as $-0.49$ in very pure GaAs samples with Si doping at $n_{\text{Si}} = 2 \times 10^{16} \text{ cm}^{-3}$, and could provide new insights for spin dynamics in this material at the metal-insulator transition.

Despite the fact that our TRKR studies forced us to look into these various fundamental questions, our work also generated proof-of-principle results which are potentially valuable for the applied fields of spintronics, and the emerging science of quantum information processing. These studies are further introduced below here.

**Suppressed spin dephasing for mobile electrons in wire structures**

The first part of this thesis presents experiments and numerical simulations on suppressed spin dephasing for mobile electron spin ensembles in GaAs-based device structures. Spin dephasing for mobile electrons in GaAs is mainly due to spin-orbit (SO) interaction [14], which couples the spin precession of an electron to its motion. We used a GaAs/AlGaAs heterostructure, for which the strength of this spin-orbit interaction can be highly anisotropic with respect to the direction of electron motion. Interestingly, it can be engineered at a value close to zero for electrons that move along the [110] crystal direction. We investigated whether this could result in suppressed spin dephasing for wires that were nano-fabricated along this crystal direction (see Fig. 1.2). We considered the case of quasi-ballistically confined structures where the width of the wire is smaller than the electron’s mean free path and the spin precession length, but larger than the width that gives strong quantum confinement. Whether electron ensembles in this regime (with frequent random momentum scattering within the ensemble)
could show suppressed spin dephasing was an open question.

![GaAs/AlGaAs heterostructure](image.png)

**Figure 1.2:** (a) Scanning electron microscope image of a GaAs/AlGaAs heterostructure with parallel wires of about 1 \( \mu m \) width processed in its top layer. Such samples were used for investigating a suppression of spin dephasing for mobile electrons in wires structures.

Spin-orbit interaction occurs when an electron is moving in an electric field, which, due to Lorentz coordinate transformation, acts on the electron spin as an effective magnetic field. The nett effect for materials with a centrosymmetric crystal structure averages out, while the lack of such symmetry (typically the case for III-V semiconductors with a zincblende crystal structure) complicates the issue. In a material like GaAs it results in the presence of SO interaction which is determined by the crystallographic symmetries (Dresselhaus SO term [15]). If the band structure of the sample is modified such that its inversion symmetry is broken, an additional SO term will arise (Rashba SO term [16]). This occurs for example at the interface between to different layers, and is therefore relevant for quantum well systems. Together, these effects give an anisotropic SO interaction, which results in an enhanced dependency of spin precession on the motion of an electron.

Within this picture the random motion of electrons in an ensemble leads to rapid dephasing of the ensemble-averaged spin orientation. This is known as the D’yakonov-Perel’ (DP) spin dephasing mechanism [17]. Wires with strong quan-
tum confinement, which allows electrons to move only in one direction, would suppress this DP dephasing, but the technical realization of such wires is in practice of little use since a weak level of disorder leads to strong electron localization. Our experimental study successfully demonstrated that the suppressed spin dephasing persists for mobile electrons with quasi-ballistic confinement. In this case, there is no quantization of the $k$ vector in the transverse wire direction, but there is much more frequent scattering for electrons moving in this direction (on the system’s edges) than for electrons moving along the wire. We studied the limits of this effect and possible implications for mesoscopic spin transport. In addition, we studied the difference between quasi-ballistic confinement for 2DEG and bulk electron ensembles. We support our experimental studies with numerical simulations.

These results can be extended to any other material with lack of inversion symmetry in the crystal structure, and are thus valuable for a wider scope of research in semiconductor spintronics.

Spin preparation and detection for localized electron spins

The second topic addressed in this thesis is related to obtaining fully quantum coherent behavior for the interaction between electron spins and optical preparation and detection pulses. A roadmap for the field that studies quantum computation [6] discusses several possible implementations, and systems that have a natural way for coupling optical photons to stationary quantum bits (qubits) have an advantage if one aims to combine quantum computation with quantum communication. Besides the typical examples for this from the field of atomic physics, there are also several solid state systems that support a direct link between the quantum states of an electron spin and the polarization state of an absorbed or emitted photon. We focused on a few of the basic requirements in a research line that aims to realize quantum information processing with optical photons as mediators of information and electron spins as quantum registers. As for classical computation, these basic requirements concern the ability to prepare, manipulate, and detect the state of a computational bit fast and reliably [18].

In our study the stationary qubit was formed by the spin state of a donor-bound electron, localized at a Si donor site in low-doped $n$-GaAs ($D^0$ system). These were addressed as a homogeneous ensemble. Earlier studies already demonstrated a long spin coherence time ($> 20 \mu$s) for this system, in combination with the ability for ultrafast all-optical spin manipulation [19, 20, 21, 22, 23]. However, the spin preparation and spin detection were implemented via optical pumping,
photo-luminescence or spectroscopic techniques which are relatively slow. Preparation and detection with picosecond laser pulses was believed to be very difficult, given the complexity of the optically excited state that is addressed for such optical interactions. This optically excited state has several sub-levels with energy splittings that are smaller than the spectral width of short laser pulses, and this could hamper coherent spin preparation. However, our attempts to characterize the spin dephasing times for $D^0$ systems with TRKR showed robust spin preparation and detection, and this initiated more detailed studies in this direction.

Figure 1.3: (Color) Bijective mapping of optical polarization states onto spin states. (a) Bloch-sphere representation for the electron spin. (b) Poincaré-sphere representation for optical polarization states. The common symbols in (a) and (b) illustrate the bijective mapping that was operated during the preparation of $D^0$ spin states with picosecond laser pulses.

In this research, we focused on finding the right conditions for robust picosecond spin preparation and detection, with a bijective mapping of the quantum states of optical polarization onto $D^0$ spin states and vice versa (see Fig. 1.3). Earlier work by Kosaka et al. had already explored this concept for non-equilibrium photo-electrons, where the coherence time for the electron spin was limited by electron hole recombination [24, 25]. We explored how this concept could be extended to spins of localized donor electrons in GaAs, which can show much longer coherence times. In particular, earlier work on $D^0$ systems already demonstrated a ratio between the coherence time [22] and manipulation time [21] in excess of $10^7$. Our work added picosecond preparation and detection to the available operations for $D^0$ spins. We thus expand a set of tools with a ratio between coherence time and operation time in excess of $10^7$. Achieving such a high ratio is one of
the essential steps on the roadmap for quantum computation research [6], since
this is essential for investigating quantum error correction schemes. While our
work thus brings us further on this road, we could not yet confirm that our new
preparation and detection scheme is equivalent to a fast projective measurement,
as is assumed in much of the theory for quantum error correction [18].

**Time-resolved study of the Landé g factor**

The third and final part of this thesis concerns a detailed study of the earlier
mentioned observation of g factor values in bulk GaAs as negative as \(-0.49\),
while most of the established literature assumes \(g \geq -0.44\). The value \(-0.44\) was
reported for low temperature (4.2 Kelvin) and low electron density by Weisbuch
et al. (Ref. [26]) from 1977, and most references to the GaAs g factor value build
on this seminal work.

The g factor governs the frequency of spin precession (angular Larmor fre-
quency \(\omega_L\)) in an external magnetic field \(B\). For a free electron the g factor value
is very close to 2 (in fact 2.0023193043622 with an uncertainty of \(1.5 \cdot 10^{-12}\)). For
electrons in a material it is altered by spin-orbit interaction. In the case of GaAs
conduction electrons this is a strong effect, which even changes sign of the g fac-
tor (leading to the opposite direction of spin precession). A closer look reveals
that the g factor does not have a fixed single value, but that it can be anisotropic
[27, 28] and that it varies as a function of parameters like temperature and photo
excitation density [29]. With the development of new time-resolved techniques it
became possible to study the g factor in more detail [30], but the field is still not
showing concensus on the values of this material parameter.

We used TRKR for a *time-resolved* analysis of the g factor values, as its value
may change during kinetic energy relaxation of an electron. We also observe a
connection between the background doping concentration and g factor values, and
compared studies on \(n\)-GaAs samples where the donor states form a band with
ultra-pure GaAs sample with extremely low levels of intentional \(n\)-doping. For
all these materials we investigated the influence of the experimental conditions
(power of optical pumping, laser wavelength etc.) on the apparent value of the
g factor. This work showed that the g factors can indeed have a drifting value
during transient TRKR signals, and that the values \(-0.49 < g < -0.44\) only
occur for low-energy electrons in samples where the donor states form a band.
1.3 Outline of this thesis

The presentation of this research is organized as follows. Chapter 2 gives a summary of the physical concepts that play a role, such as the optical selection rules for GaAs. This chapter also presents the experimental methods and the setup for TRKR measurements that was used in all of the experiments.

Chapters 3 till 5 are devoted to the experimental and numerical studies of suppressed spin dephasing for mobile electrons in 2DEG and bulk GaAs wire structures. Chapter 3 reports the experimental studies. The work compares the spin dephasing time for electrons in wires oriented along different crystal directions and for unconfined electrons in the same material, in external magnetic fields that range from 0 to 7 Tesla. The results show a suppressed spin dephasing for wires along the [110] crystal direction at low fields. Notably, the effect was observed both for electrons in a 2DEG layer and for photo-excited electrons in a bulk layer.

Chapter 4 describes results from a numerical study of the suppressed spin dephasing for 2DEG electrons in quasi-1D wires. The model assumes 2DEG electrons which are laterally confined in an elongated strip of 1 $\mu$m width and 200 $\mu$m length (similar dimensions as the wires that were studied in the experiment). The electrons freely move inside the strip, and reflect on the walls. Inside the strip elastic scattering occurs with a probability that is determined by the mobility of the 2DEG. Between the scattering events the spin of each individual electron precesses about a field that is the sum of the external magnetic field and the spin-orbit field, which depends on the direction of electron motion. Thus, we can study how the ensemble-averaged spin orientation evolves and dephases as a function of time. To explore the limits of dephasing anisotropy and suppression of spin dephasing in such wire systems we vary the strength of the spin-orbit fields and the degree and orientation of the confinement.

In Chapter 5 we present a numerical study that explores the possibility of observing a similar suppression of spin dephasing for electrons that are ballistically confined in a wire of bulk GaAs material. The approach is similar as for the 2DEG case, but now the electron ensemble is contained in a box of 1 $\mu$m by 1 $\mu$m and 200 $\mu$m length. This modeling aims at providing an explanation for the observation of suppressed spin dephasing for electrons in a bulk GaAs layer in our experiments with wire structures, which was an unexpected result. We extend this study to exploring the difference between ballistic confinement of 2DEG electrons and photo electrons in a bulk layers. We find that wires of bulk
GaAs material can indeed also show suppressed spin dephasing, and this extends the prospects for using thin bulk layers for realizing spin transport in spintronic device structures.

Chapter 6 reports the experimental results on the preparation and detection of spin states of localized electrons ($D^0$ systems) with picosecond laser pulses. By addressing resonant transitions of the Zeeman-split $D^0$ spin states to a donor-bound exciton state ($D_0X$) with the TRKR technique we could prepare the $D^0$ spins in a state that is purely determined by the polarization of the preparation pulse: The mapping between the Poincaré-sphere for optical polarization state and the Bloch-sphere for the electron spin state is bijective (Fig. 1.3). In combination with optically controlled spin echo [22] our results give prospects for achieving a $10^7$ ratio between coherence and operation time for qubits that are based on $D^0$ spins.

The final chapter, Chapter 7, presents our experimental results on characterization of the apparent g factor values which can be observed with TRKR measurements. We apply it to $n$-doped GaAs samples in a wide range of Si donor concentrations, from very low ($3 \times 10^{13} \text{ cm}^{-3}$) to metal-to-insulator transition concentrations ($2 \times 10^{16} \text{ cm}^{-3}$). We show that the g factor dependence on pump power has a very different character for samples where the donor concentration is sufficiently high for forming a donor band, as compared to samples with lower donor concentrations. For obtaining time-resolved information about the g factor we apply a wavelet analysis to the experimental TRKR data. This analysis was used for demonstrating how relaxation of the electron kinetic energy influences the apparent g factor.

References


Introduction


