Spin-orbit coupling induced spin dynamics in metals and semiconductors

Lénárd Szolnoki
Supervisor: Ferenc Simon
Budapest University of Technology and Economics

Budapest
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Chapter 1

Introduction and motivations

Understanding the basic transport phenomena during the first half of the 20th century was vital in the success of conventional electronics, which in turn revolutionized all aspects of humankind. It was recognized back in 1965 that the computing capacity growth is exponential, which is now known as the Moore’s law [1]. Although Moore’s law has been followed quite well throughout the past 5 decades, this unparalleled growth of computing and storage power may come to an end soon, which is dictated by the fundamental limits of quantum transport phenomena.

Conventional electronics operates essentially with a control over the number of electrons (or flow of electrons) in the devices for both information storage and manipulation. The idea to use the intrinsic angular-momentum of electrons (also known as spin angular momentum or shortly spin), for the same purposes arose first in the 1990s with the first published appearance in 2001 by Wolf et al. [2], soon followed by an excellent review paper by Žutić, Fabian and Das Sarma [3]. The field was coined spintronics which reminds us that it uses spin and is related to electronics. The basic idea behind spintronics is that the electron spin is much less affected by the environment than the electron momentum. The earlier property is affected only through relativistic interactions (the spin-orbit coupling in this case), whereas the electron momentum is altered due to the much stronger Coulomb interaction. When devised and controlled wisely, the spin direction of an electron ensemble (or of a few electrons) could in principle be used for information storage and computing.

It turns out that the fundamental theory of spin relaxation needs to be developed prior to any successful implementation of spintronics. The reason is that a spin ensemble loses the coherence of its spin direction which is characterized by various spin-dephasing, spin-relaxation, or decoherence times (their origin and notations are clarified later). The longevity of the spin-relaxation time indicates how much time is allowed for the spin information manipulation and read-out. A fundamental
property of spin relaxation is that it is a usually orders of magnitude slower process than the conventional momentum relaxation. As a result, spin transport is a diffusive process in most cases, i.e. the spin direction is retained over several momentum scattering events.

Two fundamental theories are known in the field which apply to the majority of cases: the so-called Elliott-Yafet theory [4, 5] is known to be valid in metals with inversion symmetry (which is the case for most elemental metals) and when momentum scattering is moderate, whereas the D’yakonov-Perel’ theory [6, 7] applies when the inversion symmetry is broken (like in GaAs) and the momentum scattering is significant. Several questions might arise immediately from this description. To what extent can these theories be validated experimentally? What happens when the above conditions are not satisfied, i.e. for a metal with inversion symmetry when the momentum scattering is large or for GaAs when the momentum scattering is moderate? Can one provide a unified mathematical basis and a unified physical description for these two relaxation mechanisms?

The present thesis was motivated by these open questions and herein I present my theoretical results which I made in this field along the above three questions. The results include an empirical verification of the Elliott-Yafet theory in metals with inversion symmetry [8], a Monte Carlo based approach which allows calculation of spin-relaxation time in materials without inversion symmetry for arbitrary value of the momentum scattering and spin-orbit interaction strength, it thus extends the D’yakonov-Perel’ theory [9], and also an intuitive approach to unify the seemingly disparate theories which apply for materials with and without inversion symmetry [10]. Although not discussed in this thesis, I contributed to an experimental study of spin relaxation in alkali atom doped graphite [11], a study of the anisotropic spin relaxation in graphite [12], and the extension of the Elliott-Yafet theory for large spin-orbit coupling [13].

This thesis is organized as follows: I introduce the basic theory of spin-orbit coupling and the theories of spin relaxation, which were known prior to this thesis. This includes the conventional Elliott-Yafet and D’yakonov-Perel’ theories and also the advances made to the field in the group which has hosted me [14], as the present results are a continuation of those efforts. I present my results in three sections and conclude with presenting my thesis points. An Appendix is provided with algorithmic details and supplementary calculations.
Chapter 2

Theoretical background

2.1 Phenomenology of spin relaxation

2.1.1 A historical introduction to spin relaxation

The origin of spin dynamics studies in metals and semiconductors can be dated back to the first observation of electron spin resonance (ESR) on itinerant electrons in Na in 1952 by Griswold, Kip, and Kittel [15]. This work was soon followed by a more detailed study on several elemental metals (Li, Na, K, and Be) by Fehér and Kip [16] including the study of temperature dependent ESR linewidth and the $g$-factor. It was recognized early on by Dyson [17] that the diffusion of electrons plays an important role for the phenomenon, which was termed CESR for conduction electron spin resonance. Dyson recognized that electrons retain their spin state during several momentum scattering events and the corresponding spin-diffusion length (the distance an electron ensemble covers diffusively while retaining half of its non-equilibrium magnetization) can be a macroscopic quantity, much larger than the mean-free path or even larger than the skin-depth in metals. One can therefore regard the Dyson theory as the first work which paved the way for spintronics.

It had been known prior to the Dyson theory that the description of magnetic resonance involves three different timescales: $T_1$, $T_2$, and $T_2^*$, which due to historical reasons are called spin-lattice, spin-spin, and spin-dephasing relaxation times, respectively. The first two relaxation times were introduced by the Bloch equations which describe magnetic resonance phenomena in general (including nuclear magnetic resonance, NMR, and ESR) [18]:

...
CHAPTER 2. THEORETICAL BACKGROUND

\[
\frac{dM_x(t)}{dt} = \gamma (M(t) \times B(t))_x - \frac{M_x(t)}{T_2},
\]

\[
\frac{dM_y(t)}{dt} = \gamma (M(t) \times B(t))_y - \frac{M_y(t)}{T_2},
\]

\[
\frac{dM_z(t)}{dt} = \gamma (M(t) \times B(t))_z - \frac{M_z(t) - M_0}{T_1},
\]

where \( \gamma \) is the gyromagnetic ratio (for electrons \( \gamma/2\pi = 28.0 \text{ GHz/T} \)). The first terms in the above equations reflect that the magnetization precesses around the external magnetic field with angular frequency \( \omega = \gamma B \). For a \( B_0 \) magnetic field along the \( z \) axis, this results in the so-called Larmor precession around \( z \) with \( \omega_0 = \gamma B_0 \) when \( M \) is tilted away from \( z \) by any perturbation.

\( M \) and \( M_0 \) are the time dependent and the equilibrium magnetization of the electron or nuclear spin ensemble in an applied magnetic field, respectively. An additional magnetic field, which rotates the magnetization away from the equilibrium \( z \) direction can be also present. The Bloch equations describe that without this additional exciting field, the magnetization is parallel to the external magnetic field and lies along the \( z \) axis. When it is tilted from this equilibrium direction, it returns to the equilibrium \( M_0 \) value with the \( T_1 \) relaxation time. In an external magnetic field, the processes which lead to \( T_1 \), involves energy transfer, which is the origin of the terminology: spin-lattice relaxation time, i.e. the lattice is thought to take up the energy during the relaxation process. The Bloch equations also describe the relaxation of a finite \( M_{x,y} \) to their equilibrium zero value with a timescale of \( T_2 \). For nuclei, \( T_2 \) is known to be dominated by the nuclear dipole-dipole interaction [19, 20]. This is the origin of the common name for \( T_2 \): spin-spin relaxation time.

Magnetic resonance experiments are commonly performed with a static \( B_0 \) magnetic field along the \( z \) axis (also called the DC magnetic field) and a circularly polarized magnetic field with amplitude \( B_1 \) (the so-called AC magnetic field), which is in the \( x,y \) plane and rotates in the direction of the Larmor precession with angular frequency \( \omega \), not necessarily matching \( \omega_0 \). This scenario is the so-called continuous wave NMR/ESR, or cw-NMR/ESR, spectroscopy. The result can be best represented in a coordinate system which rotates with \( \omega_0 \) around \( z \), where the rotating components of the magnetization are denoted as \( M_{x,\text{rot}}, M_{y,\text{rot}} \). The well-known steady-state solution of the Bloch equations read in this case[19, 20]:

\[
M_{x,\text{rot}} = \frac{B_1}{\mu_0} \chi'(\omega) = \frac{B_1}{\mu_0} \chi_0 T_2 \omega_0 \frac{T_2 \left( \omega_0 - \omega \right)}{1 + T_2^2 \left( \omega_0 - \omega \right)^2},
\]

\[
M_{y,\text{rot}} = \frac{B_1}{\mu_0} \chi''(\omega) = \frac{B_1}{\mu_0} \chi_0 T_2 \omega_0 \frac{1}{1 + T_2^2 \left( \omega_0 - \omega \right)^2},
\]
2.1. PHENOMENOLOGY OF SPIN RELAXATION

where \( \mu_0 \) is the permeability of the vacuum, \( \chi_0 \) is the static spin-susceptibility (e.g. the Curie susceptibility for non-interacting spins or the Pauli susceptibility for conduction electrons in metals). Here, the (complex) dynamic spin-susceptibility is introduced: \( \chi(\omega) = \chi'(\omega) + i\chi''(\omega) \), whose components are Lorentzian Kramers-Kronig pairs with half width at half maximum linewidths of \( \Delta \omega = 1/T_2 \). This clearly shows that cw-NMR and ESR experiments directly determine the \( T_2 \) relaxation time.

We note that the \( T_1 \) relaxation time is not immediately visible in most cw-NMR and ESR experiments. It only affects the result when the strength of the irradiating AC magnetic field is significant (i.e. saturation is present) when the solution of the Bloch equations read\[19, 20\]:

\[
M_{x,\text{rot}} = \frac{B_1}{\mu_0} \frac{\chi_0 T_2 \omega_0}{1 + T_2^2 (\omega_0 - \omega)^2 + \gamma^2 B_1^2 T_1 T_2},
\]

\[
M_{y,\text{rot}} = \frac{B_1}{\mu_0} \frac{\chi''_0 T_2 \omega_0}{1 + T_2^2 (\omega_0 - \omega)^2 + \gamma^2 B_1^2 T_1 T_2}.
\]

Much as the importance of \( T_2 \) is highlighted by the Bloch equations, the observed NMR linewidth is rarely given by \( T_2 \) as lattice defects in a solid or inhomogeneity of the magnetic field gives rise to an inhomogeneous broadening and a linewidth (in magnetic field units), which is given by \( 1/\gamma T_2^* \). \( T_2^* \) is often referred to as dephasing, as the above described inhomogeneities cause the electrons to precess with differing Larmor frequencies. This is analogous to the dephasing sound of several oscillators, which oscillate with different frequencies. In ESR, the most common origins of \( T_2^* \) are defects or sample inhomogeneities, which was first described by Portis in 1953 \[21\].

It is important to note for the later description of the Loschmidt echo in this thesis that \( T_2 \) is related to physical processes which result in a "memory loss" (in other words irreversible), whereas the processes during a \( T_2^* \) are reversible.

The usual hierarchy in NMR spectroscopy or for ESR on non-interacting paramagnetic ions is: \( T_1 \geq T_2 \gg T_2^* \). This corresponds to a magnetic resonance line, which is strongly inhomogeneously broadened and whose linewidth is independent of the temperature. It was described early in the seminal work of Fehér and Kip \[16\] that the CESR linewidth is strongly temperature dependent and that it is homogeneously broadened, i.e. the effect of \( T_2^* \) is not significant. It means that measurement of the CESR linewidth provides a convenient and direct way of determining \( T_2 = 1/\gamma \Delta B \), where \( \Delta B \) is the CESR linewidth. Another important observation was made by Yafet \[5\] that in isotropic and cubic solids \( T_1 = T_2 \) holds and due to the above arguments \( T_1 = T_2 = T_2^* \). The criterion for this equality to hold is \( \gamma B_0 \ll 1/\tau_m \), where \( \tau_m \) is the momentum relaxation time \[3\]. Given that typical values of the momentum relaxation time are \( \tau_m = 10^{-12} \ldots 10^{-14} \) s and that \( \gamma B_0 \approx 300 \) GHz for a magnetic field of 10 T, this relation is satisfied in most cases.
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The $T_1 = T_2$ equality also means that the CESR linewidth (which otherwise only senses $T_2$) provides a direct spectroscopic measure of $T_1$. The experimental observation in elemental metals [15, 16] and in phosphorous doped (i.e. n-type) silicon [22, 23] was that the CESR line-width increases with increasing temperature, i.e. $T_2$ shortens. This observation was at first astonishing and I quote from Ref. [22]: "The temperature dependence of the line-width is baffling". Several proposals, including dipolar origin of the line-width, hyperfine interaction with the $^{29}$Si, or a motional averaging of otherwise $g$-factor distributed CESR signal, failed to explain for the observed temperature dependence [22]. Dipolar interaction would give a small and temperature independent CESR line-width and electron motion should average out the hyperfine fields. The third proposal suggests that conduction electrons with different momentum, $k$, values have different $g(k)$-values. Were the conduction electrons static, one would observe the distribution of $g(k)$. However, the motion of electrons (i.e. scattering between the different $k$ values on the Fermi surface) would lead to a temperature dependence of the CESR linewidth. However, it would lead to the wrong temperature dependence as the larger the temperature (i.e. the larger the scattering), the smaller the CESR line-width would be due to motional narrowing, which is opposite to the observation.

The first successful description of $T_2$ in metals was provided in the seminal work of Elliott [4], titled: "Theory of the Effect of Spin-Orbit Coupling on Magnetic Resonance in Some Semiconductors". Although, it appears to be valid for semiconductors, it is today regarded as the first successful description of spin-relaxation in elemental metals. The Elliott theory was later amended by Yafet [5] to extend its validity towards low temperatures, which is now known as the Elliott-Yafet theory. This is described in details in this thesis.

2.1.2 Relevance of spin relaxation in spintronics

Much as the discussion of $T_1$, $T_2$, and $T_2^*$ is important to clarify the origin of the different types of spin relaxation, the discussion is greatly simplified when no external magnetic field is present: then the $T_1 - T_2$ distinction disappears. In addition, most experimentally relevant dephasing effects, which contribute to $T_2^*$, are proportional to the external magnetic field (e.g. magnetic field inhomogeneity or a Larmor frequency distribution in the sample due to defects), thus $T_2^*$ also becomes irrelevant.

In zero magnetic field conditions the literature often describe these commonly as "spin-relaxation time", or $\tau_s$. The reason for first introducing the $T_1$ and $T_2$ in this thesis, rather than their zero field counterpart, $\tau_s$ is purely historical: measurement of $\tau_s$ only became possible with the advent of precise micro- and nanotechnology in the past two decades which allow its measurement in spin transport studies[3]. It is the subject of the present doctoral thesis to study the origin and behavior of $\tau_s$. 
2.1. PHENOMENOLOGY OF SPIN RELAXATION

for an electron ensemble in metals and semiconductors.

Figure 2.1: Spin relaxation and diffusion in a spintronics device, after Ref. [3].

The spintronics literature [24, 25, 26] focuses on the behavior of $\tau_s$ as it determines how a non-equilibrium spin ensemble magnetization, which is for example injected into a device with a ferromagnetic lead, decays with time or decays spatially which results in an inhomogeneous magnetization. This is shown schematically in Fig. 2.1.

The corresponding diffusion equation reads:

$$\partial_t M = D \Delta M - \frac{M}{\tau_s},$$

(2.4)

which has an additional relaxation term as compared to the ordinary diffusion equation. In transport experiments often the one-dimensional form is sufficient with an additional source term, which describes the spin-injection [3, 27] by ferromagnetic contacts:

$$\partial_t M = D \partial_x^2 M - \frac{M}{\tau_s} + S(x,t).$$

(2.5)

For $S(x,t) = \delta(x-x_0)\delta(t-t_0)$, the solution reads:

$$M(t,x) = \frac{\Theta(t-t_0)}{\sqrt{4\pi D(t-t_0)}} e^{-\frac{t-t_0}{\tau_s}} e^{-\frac{(x-x_0)^2}{4D(t-t_0)}},$$

(2.6)
where $\Theta$ is the Heaviside step function.

For $S(x,t) = \delta(x - x_0)$, there exists a stationary solution:

$$M(x) = \frac{1}{2} \sqrt{\frac{\tau_s}{D}} e^{-\frac{|x|}{\sqrt{D\tau_s}}},$$  \hspace{1cm} (2.7)

where $\delta_s = \sqrt{D\tau_s}$ is the so-called spin diffusion length. In the mean-free path approximation, the diffusion coefficient, $D$ reads: $D = \frac{1}{d} \bar{v} \bar{l}$, where $\bar{v}$ is the average velocity of the particles and $\bar{l}$ is the mean-free path, and $d$ is the dimensionality of the system. For electrons in a metal, $D$ is well approximated as $\frac{1}{d} v_F^2 \tau$, where $v_F$ is the Fermi velocity and $\tau$ is the momentum scattering time. This gives the final result for the spin-diffusion length as $\delta_s = \frac{1}{\sqrt{d}} v_F \sqrt{\tau \tau_s}$.

Often, $T_1$ is referred to in the literature as generically spin-relaxation time. Although is not a fully proper notation, I adopt it and I shall also use it in this thesis.

### 2.2 The spin-orbit interaction

Spin-orbit coupling is a relativistic correction to the Schrödinger equation of an electron. A common intuitive interpretation of the spin-orbit coupling is the following: We assume an electron moving in a uniform electric field with no magnetic field present in the laboratory frame of reference. In the frame of reference of the electron, the surrounding electric field needs to be transformed according to the Lorentz transformation, thus the electron experiences a non-zero magnetic field in its rest frame of reference. This magnetic field has an associated Zeeman energy, which is the spin-orbit coupling term.

In the rest frame of reference of the electron, the Lorentz transformation of the electromagnetic tensor $F_{\mu\nu}$ is needed to be taken into account. In general, the magnetic field in the rest frame of reference is $B' = \gamma (B - \frac{v \times E}{c^2})$, where $\gamma = \frac{1}{\sqrt{1-v^2/c^2}}$ is the Lorentz-factor. It is $B' = -\gamma \frac{v \times E}{c^2}$, when there is zero magnetic field in the laboratory frame of reference (Fig. 2.2).

Besides this semiclassical consideration, spin-orbit coupling can be exactly derived from the Dirac equation in the low momentum limit. The Dirac equation in the presence of external electromagnetic fields reads:

$$(\gamma^\mu k_\mu - m_0 c I) |\Psi\rangle = 0.$$  \hspace{1cm} (2.8)

Taking the limit of $v/c \rightarrow 0$, multiple relativistic corrections emerge, one of them is the spin-orbit coupling. This gives the so-called Pauli-Schrödinger equation:
2.2. THE SPIN-ORBIT INTERACTION

\[ E \]

\[ s \]

\[ v \]

\[ \text{Lorentz transformation} \]

\[ E \]

\[ B \]

\[ \Omega \]

Figure 2.2: A classical interpretation of the spin orbit coupling. The electron experiences a magnetic field in its rest frame of reference even when there is no magnetic field in the laboratory frame of reference.

\[ H = H_{\text{PS}} + H_{\text{K}} + H_{\text{SOC}} + H_{\text{D}}, \]

\[ H_{\text{PS}} = \left( \frac{(p - qA)^2}{2m_0} - \frac{\hbar q}{2m_0} \sigma B + q\phi \right), \]  

\[ H_{\text{K}} = -\frac{1}{8m_0^3c^2} k^4, \]  

\[ H_{\text{SOC}} = -\frac{\hbar}{4m_0^2c^2} \sigma \left( qE \times k \right), \]  

\[ H_{\text{D}} = \frac{\hbar^2 q}{8m_0^2c^2} \cdot (\Delta \phi), \]

where \( H_{\text{PS}} \) contains the main kinetic energy term and the interaction with external electromagnetic field. \( H_{\text{K}} \) is the kinetic energy correction, \( H_{\text{SOC}} \) is the spin-orbit coupling term, and \( H_{\text{D}} \) is the so-called Darwin term. In a periodic potential of a solid, the kinetic and Darwin terms can be included in the \( \varepsilon_k \) kinetic energy, since neither of them depends on the spin state of the electron.

For an electric field, which stems from a central Coulomb potential (i.e. that of an atom, \( E = -\frac{\phi}{\sigma} e_r \)) the SOC term can be written as:

\[ H_{\text{SOC}} = \frac{\hbar}{4m_0^2c^2} \frac{\partial \phi}{\partial \sigma} \left( e_r \times k \right), \]

\[ = \frac{\hbar}{4m_0^2c^2} \frac{\partial \phi}{\partial r} \sigma L. \]
In this formalism, the direct coupling between the spin and orbital momentum is apparent, hence the name “spin-orbit coupling.” Strictly speaking, the name SOC is inappropriate for cases when the orbital quantum number is not a good quantum number, still the name stuck to describe similar, relativistic phenomena, which stem from an electric field. The atomic potential induced SOC is commonly called “intrinsic” SOC. In this thesis I also consider SOC in bulk conductors and semiconductors. The nature of the SOC significantly differs between inversion symmetric and inversion symmetry breaking materials. The SOC in inversion symmetric materials is described by the Elliott-Yafet mechanism. The SOC in inversion breaking materials is described by the D’yakonov-Perel’ mechanism. An inversion breaking electric field can emerge from internal electric potential or an external electric field (the so called Bychkov-Rashba effect).

2.3 Kramers theorem and the spin-orbit coupling

The Kramers theorem states that when the Hamiltonian has an antiunitary symmetry $T$ such that $T^2 = -I$, then its energy states are pairwise degenerate. Generally, in an inversion symmetry breaking material, the SOC induced by the internal electric field splits the spin-degenerate bands. In a conductor the SOC splitting in the conduction band dominates the SOC induced behavior.

However, in inversion symmetric materials the SOC does not split the kinetic bands. The product of inversion and time reversal forms an antiunitary symmetry. This symmetry also does not change the wave vector of a given state. There is a degenerate orthogonal state with the same wave vector for every energy state with wave vector $k$ due to Kramers theorem.

To prove the Kramers theorem, antiunitary operators have to be rigorously defined. $A : H_1 \rightarrow H_2$ is an antilinear operator if for every $|\phi\rangle, |\psi\rangle \in H_1$ and $\alpha, \beta \in \mathbb{C}$:

$$A(\alpha |\phi\rangle + \beta |\psi\rangle) = \alpha^* A |\phi\rangle + \beta^* A |\psi\rangle.$$  

(2.11)

$A$ is also antiunitary if for every $|\phi\rangle, |\psi\rangle \in H_1$:

$$\langle \phi |\psi\rangle = \langle A\psi | A\phi \rangle.$$  

(2.12)

Kramers theorem is usually demonstrated on time-reversal symmetry, however the theorem generally applies to any antiunitary symmetry as well. We consider the time-reversal symmetry $T$, which is an antiunitary symmetry. For a single electron, $T$ is commonly represented as:

$$T = e^{-i\pi\mathbf{S}_y/\hbar} K,$$  

(2.13)
where $K$ is conjugation. This yields $T^2 = -1$.

Kramers theorem only requires an antiunitary symmetry $A$ and $A^2 = -1$. Note that for any unitary symmetry $U$, $UT$ is an antiunitary symmetry. Consider an energy eigenstate $|\phi\rangle$ with energy $\varepsilon$. Since $A$ is a symmetry operation, $|A\phi\rangle$ is also an energy eigenstate with the same energy. To prove that it is orthogonal to the original state I use Eq. (2.12) above with $|\psi\rangle = |A\phi\rangle$:

$$\langle \phi | A\phi \rangle = \langle A^2\phi | A\phi \rangle = -\langle \phi | A\phi \rangle .$$

(2.14)

### 2.4 The Elliott-Yafet theory

The Elliott-Yafet theory assumes a metal with inversion symmetry. As mentioned above, the spin states remain degenerate even in the presence of a finite SOC interaction. Elliott showed that the simplest model to describe spin relaxation meaningfully requires two additional bands, i.e. altogether a four band model. Therefore the theory takes into account the conduction band and a nearby band. The interpretation of the EY theory is depicted in Fig. 2.3. The spin state of an electron may flip at momentum scattering events with a low probability and between scattering events, the spin state remains unchanged.

```latex
\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{elliot-yafet.png}
\caption{The interpretation of the Elliott-Yafet theory.}
\end{figure}
```

Since the spin-flip probability and the frequency of scattering events is proportional, the EY theory predicts that the spin-relaxation time is proportional to the momentum relaxation time:
\[
\frac{1}{\tau_s} = \alpha \left( \frac{L}{\Delta E} \right)^2 \frac{1}{\tau_m}, \tag{2.15}
\]

where \( L \) is a specific matrix element of the spin-orbit coupling, \( \Delta E \) is the gap between the two bands and \( \tau_{s,m} \) are the spin- and momentum-relaxation times, respectively. \( \alpha \) is a dimensionless parameter around unity that depends on the crystal structure.

Elliott further showed that the magnetic energy of the admixed states is different from that of the pure spin-states, i.e. there is a shift in the electron \( g \)-factor:

\[
\Delta g = g - g_0 = \alpha_2 \frac{L}{\Delta}, \tag{2.16}
\]

where \( g_0 \approx 2.0023 \) is the free electron \( g \)-factor, \( \alpha_2 \) is another band structure dependent constant near unity. Eqs. (2.15) and (2.16) gives the so-called Elliott relation:

\[
\Gamma_s = \frac{\alpha_1}{\alpha_2^2} \Delta g^2 \Gamma, \tag{2.17}
\]

Where we introduced the spin- and momentum scattering rates: \( \Gamma_s = \hbar / \tau_s \) and \( \Gamma = \hbar / \tau_m \).

As Eq. 2.15 shows, the microscopic origin of spin relaxation in this model is the spin-orbit coupling.

The model Hamiltonian for a single electron in the framework of the Elliot-Yafet theory is:

\[
H = \frac{p^2}{2m} + V + \frac{\hbar}{4mc^2} (\nabla V \times p) \cdot \sigma, \tag{2.18}
\]

where \( V \) is the periodic potential, \( \sigma \) is the vector of the Pauli-matrices. The last term is the spin-orbit coupling (SOC) Hamiltonian. The theory also takes four bands into account (two kinetic bands with their spin states). The spin-orbit coupling does not break the degeneracy of the kinetic bands due to Kramers theorem: The product of time reversal and inversion is an antiunitary symmetry of the Hamiltonian. This symmetry also leaves the wave vector of an electron Bloch-state in place, so for every energy eigenstate with a given wave vector there is a degenerate, orthogonal state with the same energy and wave vector (Fig. 2.4).

The SOC term can be better described using Wannier-functions as follows. The wave function is expanded conventionally as:

\[
\Phi_k^s(r) = \frac{1}{\sqrt{N}} \sum_d w_k^s(r - d) \exp(i \mathbf{k} \cdot \mathbf{d}). \tag{2.19}
\]

where \( w_k^s \) are the Wannier functions.
2.4. THE ELLIOTT-YAFET THEORY

Figure 2.4: Schematics of the band model of the EY theory. The spin-orbit coupling does not break up the degenerate spin states. $\Gamma = \hbar/\tau_m$ is the quasi-particle broadening.

Superposition of the electric fields allows the periodic potential to be written as a sum of the atomic potentials:

$$V = \sum_d V(r - d),$$  \hspace{1cm} (2.20)

where $V(r - d)$ is non-zero in the Wigner-Seitz cell. Substituting this potential into the Hamiltonian yields for the SOC term:

$$H_{SOC} = \frac{\hbar}{4mc^2} \sum_d \left( \nabla V(r - d) \times p \right) \cdot \sigma.$$  \hspace{1cm} (2.21)

An isotropic atomic potential in each Wigner-Seitz cell allows the gradient in the SOC term to be expressed using spherical coordinates:

$$\frac{\hbar}{4mc^2} \frac{1}{r} \frac{\partial V}{\partial r} l \cdot \sigma = \xi(r) l \cdot s.$$  \hspace{1cm} (2.22)

Since only two kinetic bands are taken into account, only an effective SOC Hamiltonian is needed that gives the correct matrix elements for only these bands.
CHAPTER 2. THEORETICAL BACKGROUND

\[ \langle \phi_s(k) | H_{\text{SOC}} | \phi_t(k') \rangle = \delta_{kk'} \langle w^s_k(r) | \xi(r) | l \cdot s | w^{t}_{k'}(r) \rangle = \delta_{kk'} \lambda \langle w^s_k(r) | l \cdot s | w^{t}_{k'}(r) \rangle, \]  

(2.23)

where \( \lambda \) is assumed to be independent of \( k \) and \( k' \) for the fixed bands, \( s \) and \( t \). This approximation yields a reduced effective Hamiltonian for calculating this matrix element:

\[ H_{\text{SOC}} = \lambda \cdot s. \]  

(2.24)

In the EY model, the SOC term is treated as first order perturbation. Even though the SOC term does not split the kinetic bands, it still mixes the spin states with the neighboring kinetic band. Without SOC, the spatial wave functions can be written using Wannier-functions:

\[ \Psi^s_{k, \pm}(r) = \Phi^s_k(r) \chi^\pm = \frac{1}{\sqrt{N}} \sum_d w^s_k(r - d) \exp(i k \cdot d) \chi^\pm, \]  

(2.25)

where \( \chi^\pm \) is the spin wave function. The corresponding SOC matrix-elements read:

\[ \langle \Psi^s_{k, \sigma} | H_{\text{SOC}} | \Psi^t_{k', \sigma'} \rangle = \frac{1}{N} \sum_{d, d'} \langle w^s_k(r - d), \sigma | H_{\text{SOC}} | w^t_{k'}(r - d'), \sigma' \rangle \exp(i k' \cdot d' - k d). \]  

(2.26)

Assuming that the \( d \neq d' \) terms are negligible and using Eq. 2.24 within a cell around \( d \), we obtain:

\[ \langle \Psi^s_{k, \sigma} | H_{\text{SOC}} | \Psi^t_{k', \sigma'} \rangle = \frac{\lambda}{N} \sum_d \langle w^s_k, \sigma | l_z s_z + l_x s_x + l_y s_y | w^t_{k'}, \sigma' \rangle \exp(i k \cdot d - k' \cdot d). \]  

(2.27)

The sum is zero for \( k \neq k' \):

\[ \langle \Psi^s_{k, \sigma} | H_{\text{SOC}} | \Psi^t_{k', \sigma'} \rangle = \delta_{k,k'} \lambda \langle w^s_k, \sigma | l_z s_z + l_x s_x + l_y s_y | w^t_{k'}, \sigma' \rangle. \]  

(2.28)

When in the perturbed wave functions only the \( s \) and \( t \) bands are taken into account:

\[ | \tilde{\Psi}^s_{k, \sigma} \rangle = | \Psi^s_{k, \sigma} \rangle + \sum_{k', \sigma'} \frac{\langle \Psi^t_{k', \sigma'} | H_{\text{SOC}} | \Psi^s_{k, \sigma} \rangle}{\Delta} | \Psi^t_{k', \sigma'} \rangle = | \Psi^s_{k, \sigma} \rangle + \frac{\lambda}{\Delta} \sum_{\sigma'} \langle w^t_{k}, \sigma' | l_z s_z + l_x s_x + l_y s_y | w^s_k, \sigma \rangle | \Psi^t_{k', \sigma'} \rangle, \]  

(2.29)
where $\Delta$ is the energy separation between the kinetic bands. For a specific spin direction, i.e. $\sigma = +$:

$$|\tilde{\Psi}_{k,+}^s\rangle = |\Psi_{k,+}^s\rangle + \frac{\lambda}{2\Delta} \left( \langle w_k^t|l_z|w_k^s\rangle |\Psi_{k,+}^t\rangle + \langle w_k^t|l_x + il_y|w_k^s\rangle |\Psi_{k,-}^t\rangle \right). \quad (2.30)$$

Finally, the perturbed Wannier-functions read:

$$|\tilde{w}_{k,+}^s\rangle = |w_{k,+}^s\rangle + \frac{\lambda}{2\Delta} \left( \langle w_k^t|l_z|w_k^s\rangle |w_{k,+}^t\rangle + \langle w_k^t|l_x + il_y|w_k^s\rangle |w_{k,-}^t\rangle \right). \quad (2.31)$$

The momentum and spin relaxations are induced by a spin-independent Hamiltonian term $(H_{\text{int}})$, which describes the interaction of electrons with phonons and non-magnetic impurities. This term is treated as a time dependent perturbation. Even without the exact form of $H_{\text{int}}$, a relationship between the momentum and spin relaxation times can be deduced. The terms which are relevant for the momentum relaxation time are:

$$\frac{1}{\tau_m} \propto \left| \langle \tilde{\Psi}_{k',+}|H_{\text{int}}|\tilde{\Psi}_{k,+}\rangle \right|^2 + \left| \langle \tilde{\Psi}_{k',-}|H_{\text{int}}|\tilde{\Psi}_{k,+}\rangle \right|^2, \quad (2.32)$$

where the second term is negligible compared to the first term. In turn, the spin relaxation time is related to the following terms:

$$\frac{1}{T_1} \propto \left| \langle \tilde{\Psi}_{k',-}|H_{\text{int}}|\tilde{\Psi}_{k,+}\rangle \right|^2 \quad (2.33)$$

Eq. 2.30 can be rewritten with simplified coefficients for the perturbed wave functions:

$$\tilde{\Psi}_{k,+}(r) = a_k(r)\chi_+ + b_k(r)\chi_-, \quad (2.34)$$

where the $a_k$ and $b_k$ coefficients read:

$$a_k(r) = \Phi_k^s(r) + \frac{\lambda}{2\Delta} \langle w_k^t|l_z|w_k^s\rangle \Phi_k^t(r) \quad (2.35)$$

$$b_k(r) = \frac{\lambda}{2\Delta} \langle w_k^t|l_x + il_y|w_k^s\rangle \Phi_k^t(r).$$

Application of the spatial and time inversion operators yields for the perturbed wave function with opposite spin:

$$\tilde{\Psi}_{k,-}(r) = a_k^*(r)\chi_- - b_k^*(r)\chi_+. \quad (2.36)$$

The matrix elements for the momentum scattering and spin-flip processes read:
\[ \langle \tilde{\Psi}_{k',+} | H_{\text{int}} | \tilde{\Psi}_{k,+} \rangle = \langle a_k | H_{\text{int}} | a_k \rangle + \langle b_k | H_{\text{int}} | b_k \rangle \]
\[ \langle \tilde{\Psi}_{k,-} | H_{\text{int}} | \tilde{\Psi}_{k,+} \rangle = \langle a_{-k} | H_{\text{int}} | b_k \rangle + \langle b_{-k} | H_{\text{int}} | a_k \rangle \]  
(2.37)

In the framework of first order time dependent perturbation calculation, the ratio of the two relaxation times is:

\[ \frac{T_1}{\tau_m} \propto \frac{\left| \langle \tilde{\Psi}_{k',-} | H_{\text{int}} | \tilde{\Psi}_{k,+} \rangle \right|^2}{\left| \langle \tilde{\Psi}_{k',+} | H_{\text{int}} | \tilde{\Psi}_{k,+} \rangle \right|^2} \propto \left( \frac{\lambda}{\Delta} \right)^2 \]  
(2.38)

2.5 The D’yakonov-Perel’ theory

2.5.1 Introduction

The D’yakonov-Perel’ mechanism describes spin-relaxation in metals with broken inversion symmetry. This inversion breaking can originate from the internal electric fields (bulk SOC) of the material, or from an applied external electric field (Bychkov-Rashba SOC).

In the framework of the D’yakonov-Perel’ model, it is sufficient to consider a single kinetic band to obtain the leading terms to spin relaxation. The degeneracy of the otherwise spin-degenerate states is lifted by spin-orbit coupling in this case. A common interpretation of this degeneracy lifting is to describe it with an effective Zeeman splitting due to a built-in effective field that is shown in Fig. 2.5.

The model Hamiltonian for a single electron in the framework of the D’yakonov-Perel’ theory is the following:

\[ H = \sum_{k,\sigma} \varepsilon(k) a_{k,\sigma}^+ a_{k,\sigma} + \sum_{k,\sigma,\sigma'} a_{k,\sigma}^+ H_{\text{SOC},\sigma',\sigma}(k) a_{k,\sigma}, \]
\[ H_{\text{SOC}}(k) = g_s \mu_B S B(k) = \frac{\hbar}{2} \sigma \Omega(k), \]  
(2.39)

where the first term is the kinetic term, the second term is due to the spin-orbit coupling. The second term can be written in the form of a Zeeman Hamiltonian with a built-in, \( k \)-dependent effective magnetic field.

The DP description considers that an electron experiences this intrinsic field which causes a Larmor-precession of the spin state between scattering events. Scattering events leave the spin unchanged, however change the direction and magnitude of the Larmor precession frequency. As a result, the spin follows a random walk, which is semantically depicted in Fig. 2.6.
2.5. THE D’YAKONOV-PEREL’ THEORY

Figure 2.5: The spin-orbit coupling breaks up the spin-degenerate states in materials without inversion symmetry. $\Gamma = \hbar/\tau_m$ is the quasi-particle broadening.

Figure 2.6: Schematics of the conventional D’yakonov-Perel’ spin relaxation mechanism. Note that the electron spin precesses around the internal effective magnetic fields, whose direction and magnitude changes after each scattering event.

2.5.2 Bychkov-Rashba SOC

A simplified description of the Bychkov-Rashba SOC is that it originates from a homogeneous external electric field. The exact description is more involved, screening and boundary effects are needed to be taken into account [3]. The Bychkov-Rashba SOC Hamiltonian can be directly derived from Eq. 2.9 assuming
a homogeneous electric field, as follows:

\[ H_{\text{BR}} = -\frac{\hbar}{2}\sigma\Omega_{\text{BR}}(k), \]
\[ \Omega_{\text{BR}}(k) = \frac{e}{2m_e^2c^2}E \times k. \]  

(2.40)

The Bychkov-Rashba SOC is commonly encountered in field effect transistor devices, which constitute a two-dimensional electron gases with a perpendicular electric field. In this case, the Bychkov-Rashba SOC can be written:

\[ \Omega_{\text{BR}}(k) = \alpha[-k_y, k_x, 0], \]
\[ \alpha = \frac{eE}{2m_e^2c^2}. \]  

(2.41)

Fig. 2.7 depicts such a configuration.

Figure 2.7: A two-dimensional electron gas with gate-tuned perpendicular electric field. The electric field strength determines the SOC strength in the electron gas, this is partly the working principle of the so-called Datta-Das spin transistor [28]. However the exact principle of this tailoring has been controversial because of the screening and boundary effects mentioned before [3].

### 2.5.3 Bulk SOC

Time-reversal symmetry restricts the internal field so that \( \Omega(k) = -\Omega(-k) \). Other than this, the bulk SOC is only restricted by the symmetries of the material. One such example is the bulk SOC in the conduction band of GaAs (and more generally in zincblende crystal structures) [29, 30]. This is the so called Dresselhaus spin-orbit Hamiltonian, which reads:
\[ \Omega(k) = \frac{\mathcal{L}}{\hbar k_F^3} \left[ k_x (k_y^2 - k_z^2), k_y (k_z^2 - k_x^2), k_z (k_x^2 - k_y^2) \right]. \] (2.42)

The vector components of \( \Omega(k) \) are shown in Fig. 2.8 while \( k \) traces a spherical Fermi-surface.

Figure 2.8: The distribution of Dresselhaus \( \Omega(k) \) assuming a spherical Fermi-surface.

This form of the intrinsic Larmor-frequency vector can be deduced from the \( T_d \) (tetrahedral) symmetry group of the zincblende structure (Fig. 2.9).

To obtain this result, only the leading terms in \( k \) are considered and the kinetic band of interest is assumed to be non-degenerate at the \( \Gamma \)-point. The latter requirement does not hold for the valence band in GaAs. Due to Kramers-theorem, \( \Omega(k) = -\Omega(-k) \), so in the series expansion of \( \Omega(k) \) only odd-degree terms are present. We obtain from Eq. 2.9 and Eq. 2.39:
Figure 2.9: The zincblende structure has $T_d$ (tetrachedral) symmetry.

$$\Omega(k) = \alpha \langle E \rangle_k \times k,$$

$$\alpha = \frac{\hbar e}{4m^2_0 c^2}. \quad (2.43)$$

In the series expansion of $\langle E \rangle_k$ only even-degree terms are present, which are denoted as follows:

$$\langle E_i \rangle_k = E_{0,i} + A_{i,jl} k_j k_l + \ldots, \quad (2.44)$$

where $A$ is a 3-index tensor. Due to the $T_d$ symmetry, $E_0 = 0$. $A$ transforms as vector in each of its indices. Vector transformations are represented by the $T_2$ irreducible representation of $T_d$. Therefore transformations of $A$ are represented by $T_2 \times T_2 \times T_2$. Since $A$ is invariant to symmetry transformations, it must lie in the subspace of $A_1$. In the tensorial product representation, this subspace is one
2.5. THE D’YAKONOV-PEREL’ THEORY

dimensional. Therefore \( A \) and \( \langle E \rangle_k \) have a single degree of freedom and thus a single possible form which reads:

\[
\langle E \rangle_k = \lambda [k_y h_z, k_x h_z, k_x k_y],
\]

\[
\Omega(k) = \alpha \langle E \rangle_k \times k = -\alpha \lambda \left[ k_x (k_y^2 - k_z^2), k_y (k_z^2 - k_x^2), k_z (k_x^2 - k_y^2) \right].
\] (2.45)

The effective Dresselhaus SOC Hamiltonian is used in this thesis to calculate spin-relaxation time in semiconductors with the zincblende structure.

2.5.4 Toy model of spin kinetic equation

The D’yakonov-Perel’ mechanism describes the regime of \( \Omega \ll 1/\tau_m \), where \( \Omega \) is the typical (or average) magnitude of the SOC related Larmor-precession frequencies and \( \tau_m \) is the momentum-scattering time. In this regime, the angle that the spin rotates between scattering events is of the order of \( \Omega \tau_m \ll 1 \), i.e. it is a small angle.

An ensemble of the spin vectors diffuse on the surface of the Bloch sphere, with a diffusion coefficient that is proportional to the square of the traveled distance between scatterings \( (\Omega^2 \tau_m^2) \) and in addition it is inversely proportional to the time between scattering events \( (\tau_m) \). The spin relaxation time is inversely proportional to this diffusion coefficient, which yields the main result of the D’yakonov-Perel’ theory:

\[
\frac{1}{\tau_s} \propto \Omega^2 \tau_m.
\] (2.46)

Although this intuitive approach gives a quantitatively correct description of the spin-relaxation under the above condition, one can arrive at a more appropriate result following analytic calculations.

2.5.5 Analytical calculation of the spin-relaxation

For the analytical calculations, I follow the work of Pikus and Titkov[7]. Their approach is to track the density matrix \( \rho(k) \) of a single electron, with components \( \rho_{ij}(k) \), \( i \) and \( j \) being the indices of spin states.

\[
\frac{\rho(k)}{\tau} + \frac{i}{\hbar} [H_{SOC}(k), \rho(k)] + \sum_{k'} W_{k',k} (\rho(k) - \rho(k')) = G,
\] (2.47)

where \( G \) is the so-called generation matrix, \( \tau \) is the lifetime of any given electron state, \( W_{k',k} \) is the transition probability (supposedly independent of spin), and \( H_{SOC}(k) \) is the SOC Hamiltonian. Note that \( \tau \) is neither the spin or momentum relaxation time, it is the lifetime of any electron state relaxation in any given \( k \).
wave vector. This is a variant of the linearized Boltzmann-equation, where the coupling between different $k$ states are disregarded but within a $k$ kinetic state, the full density matrix is taken into account.

To give a somewhat different view on the calculations of Pikus and Titkov, I change the representation from the electron density matrix to the following:

$$s_i(k) = \text{Tr} [\rho(k) s_i], \quad (2.48)$$

where $s_i = \sigma_i/2$, and $\sigma_i$ are the Pauli matrices. I also consider the following form of the SOC Hamiltonian:

$$H_{SOC}(k) = \frac{\hbar}{2} \sigma \Omega(k), \quad (2.49)$$

where $\Omega(k)$ is the Larmor precession of a given electron at $k$. After taking the trace of Eq. 2.47:

$$s(k) \frac{\tau}{\tau} + \Omega(k) \times s(k) + \sum_{k'} W_{k',k} (s(k) - s(k')) = \text{Tr} [G s]. \quad (2.50)$$

Herein, $s(k)$ can be written in the form $s(k) = \overline{s} + s_1(k)$, where $\overline{s}$ is the average of $s(k)$ over all directions of $k$. After averaging 2.50 for all $k$ directions:

$$\frac{\overline{s}}{\tau} + \overline{\Omega}(k) \times \overline{s(k)} = \text{Tr} [\overline{G} s], \quad (2.51)$$

where the overline denotes averaging over all directions of $k$. The equation for $s_1(k)$ can also be found from Eq. 2.50:

$$\frac{s_1(k)}{\tau} + \Omega(k) \times s_1(k) + \overline{\Omega}(k) \times \overline{s}(k) + \sum_{k'} W_{k',k} (s_1(k) - s_1(k')) + \text{Tr} [\overline{G} - G(k)] s = 0. \quad (2.52)$$

For the D'yakonov-Perel' regime, $s_1(k) \ll \overline{s}$ is assumed and therefore the second term in the latter equation can be omitted. At the same time, for $\tau_m \ll \tau_s$ the first and last terms can also be omitted. In the further treatment, it is assumed that the electron scattering is elastic, the electron energy spectrum is isotropic, and the scattering cross section $\sigma(k',k)$ depends only on the scattering angle $\theta$. Then, we obtain for $s_1(k)$ the following result:

$$\Omega(k) \times \overline{s} + \int \frac{d\Omega}{2\pi} \sigma(\theta) (s_1(k) - s_1(k')) = 0. \quad (2.53)$$

We consider the following for a particular solution of Eq. 2.53:
2.5. THE D’YAKONOV-PEREL’ THEORY

\[ s_1(k) = -\tau^* \Omega(k) \times \mathbf{s}, \]  
(2.54)

where \( \tau^* \) is an arbitrary parameter which is to be determined later from Eq. 2.53. In order to find the solution, \( \Omega(k) \) needs to be expanded over the spherical functions:

\[ \Omega_i(k) = \sum_m C_{i,l,m} Y^l_m(\theta, \varphi). \]  
(2.55)

Here, it is taken into account that the power index \( l \) is the same for all terms. Now, Eq. 2.54 can be substituted into Eq. 2.53, making use of a known relation for spherical functions [7]:

\[ \int Y^l_m(\vartheta', \varphi') \sigma(\theta') \frac{d\Omega'}{2\pi} = Y^l_m(\vartheta, \varphi) \int_0^\pi \sigma(\theta) P^l(\cos \theta) \sin \theta d\theta. \]  
(2.56)

Here \( \vartheta \) and \( \varphi \) are the angular coordinates of \( k \), \( \vartheta' \) and \( \varphi' \) are the analogous coordinates for \( k' \), and \( P^l(\cos \theta) \) is the Legendre polynomial. Then, we obtain from Eq. 2.53:

\[ \frac{1}{\tau^*} = \int_{-1}^1 \sigma(\mu)(1 - P^l(\mu)) d\mu, \quad \text{where} \ \mu = \cos \theta. \]  
(2.57)

The momentum scattering time is given by a similar equation:

\[ \frac{1}{\tau_m} = \int_{-1}^1 \sigma(\mu)(1 - P^l(\mu)) d\mu. \]  
(2.58)

The expression for \( 1/\tau^* \) can be rewritten in the form:

\[ \frac{1}{\tau^*} = \gamma_i \frac{1}{\tau_m}, \quad \text{where} \ \gamma_i = \frac{\int_{-1}^1 \sigma(\mu)(1 - P^l(\mu)) d\mu}{\int_{-1}^1 \sigma(\mu)(1 - P^l(\mu)) d\mu}. \]  
(2.59)

Substituting Eq. 2.54 into Eq. 2.51, the following relation for \( \mathbf{s} \) is obtained:

\[ \frac{s}{\tau} - \tau^* \Omega(k) \times (\Omega(k) \times \mathbf{s}) = \text{Tr} \left[ \mathbf{G} \mathbf{s} \right]. \]  
(2.60)

The second term describes the spin relaxation due to the DP mechanism,

\[ \left( \frac{\partial \mathbf{s}}{\partial t} \right)_{\text{sp. rel}} = \tau^* \Omega(k) \times (\Omega(k) \times \mathbf{s}). \]  
(2.61)

Expressing it for a single component component gives:

\[ \left( \frac{\partial s_z}{\partial t} \right)_{\text{sp. rel}} = -\tau^* \left( s_z \left( \frac{\Omega^2_x}{\Omega_z} \right) - s_x \left( \frac{\Omega_x \Omega_z}{\Omega_z} \right) - s_y \left( \frac{\Omega_y \Omega_z}{\Omega_z} \right) \right), \]  
(2.62)
where the overline denotes average on the Fermi-surface.

As a result, the spin relaxation time is generally expressible in tensorial form, with components which read:

\[
\frac{1}{\tau_s} \sigma_{ij} = \gamma_l \tau_m \left( \Omega^2 \delta_{ij} - \Omega_i \Omega_j \right).
\] (2.63)

It is the same result as presented in Ref. [3] and it also returns the prediction of the toy model (Eq. (2.46)).

2.6 Unified theory of the Elliott-Yafet and D’yakonov-Perel’ spin-relaxation mechanisms

As discussed above, conventionally spin relaxation is explained by the Elliott-Yafet (EY) and the D’yakonov-Perel’ (DP) mechanisms. It was discussed that these are valid depending on whether the inversion symmetry in the material is broken or retained. It was also presented that the mathematical foundations and formalisms of the two descriptions are quite different: first order time dependent perturbation calculation versus a random-walk based motional narrowing approach for the EY and DP theories, respectively.

This different methodology is quite intriguing as in the end the very same physical phenomenon, spin-relaxation, is studied. Although the interplay between these mechanisms has been studied in semiconductors [27, 7, 31, 32], no attempts have been made to unify their descriptions. In addition, it was found experimentally that strongly correlated novel metals (MgB$_2$ and K$_3$C$_60$), i.e. when the quasiparticle broadening is large due to a large electron-phonon interaction, display spin-relaxation whose phenomenology resembles both the EY and DP type mechanisms[33, 34]. This lead the authors of Ref. [14] to considered whether the Elliott-Yafet and D’yakonov-Perel’ relaxation mechanism results could be obtained simultaneously by imposing a four band Hamiltonian which contains spin-orbit coupling terms which correspond to atomic (intrinsic) SOC and to those which are related to the inversion symmetry breaking terms. This means that a common mathematical basis could be provided for the two seemingly disparate mechanisms. This section is presented mainly after Ref. [14].

The minimal model of spin-relaxation in a four-state (two bands with spin) model Hamiltonian for a two-dimensional electron gas (2DEG) in a magnetic field
was considered, which reads:

\[ H = H_0 + H_Z + H_{\text{scatt}} + H_{\text{SO}} \]  \hspace{1cm} (2.64a)
\[ H_0 = \sum_{k,\alpha,s} \epsilon_{k,\alpha} c_{k,\alpha,s}^\dagger c_{k,\alpha,s} \]  \hspace{1cm} (2.64b)
\[ H_Z = \Delta_Z \sum_{k,\alpha,s} s c_{k,\alpha,s}^\dagger c_{k,\alpha,s} \]  \hspace{1cm} (2.64c)
\[ H_{\text{SO}} = \sum_{k,\alpha,\alpha',s,s'} \mathcal{L}_{\alpha,\alpha',s,s'}(k) c_{k,\alpha,s}^\dagger c_{k,\alpha',s'} \]  \hspace{1cm} (2.64d)

where \( \alpha = 1 \) (nearby), \( 2 \) (conduction) is the band index with \( s = (\uparrow), (\downarrow) \) spin, \( \epsilon_{k,\alpha} = \hbar^2 k^2 / 2m^*_\alpha - \delta_{\alpha,1}\Delta \) is the single-particle dispersion with \( m^*_\alpha = (-1)^\alpha m^* \) effective mass and \( \Delta \) band gap, \( \Delta_Z = g\mu_B B_z \) is the Zeeman energy. \( H_{\text{scatt}} \) is responsible for the finite quasi-particle lifetime due to impurity and electron-phonon scattering and \( \mathcal{L}_{\alpha,\alpha',s,s'}(k) \) is the SOC.

![Band structure](image)

Figure 2.10: The band structure of a 2DEG in a magnetic field after Ref. [14]. The effects of the SOC are not shown. Vertical arrows indicate the energy separations between relevant bands with the same \( k \) value.

The relevant band structure is shown in Fig. 2.10. The energies and eigenstates
It was found that the most general expression of the SOC for the above levels reads:

\[
L_{\alpha,\alpha',s,s'}(k) = \begin{pmatrix}
L_{\uparrow\uparrow} & L_{\uparrow\downarrow} & L_{\downarrow\uparrow} & L_{\downarrow\downarrow} \\
L_{\downarrow\uparrow} & L_{\downarrow\downarrow} & L_{\downarrow\uparrow} & L_{\downarrow\downarrow} \\
L_{\uparrow\uparrow} & L_{\downarrow\uparrow} & L_{\downarrow\uparrow} & L_{\downarrow\downarrow} \\
L_{\downarrow\uparrow} & L_{\downarrow\downarrow} & L_{\downarrow\uparrow} & L_{\downarrow\downarrow}
\end{pmatrix},
\]

(2.66)

where \( L_{ss'}(k), L_{ss'}(k) \) are the wavevector dependent intra- and inter-band terms, respectively, which are phenomenological, i.e. not related to a microscopic model. The terms, which mix the same spin direction can be ignored as these commute with the \( S_z \) operator and do not lead to spin-relaxation. The SOC terms, which contribute to spin-relaxation are

\[
L_{\alpha,\alpha',s,s'}(k) = \begin{pmatrix}
0 & \mathcal{L} & 0 & L \\
\mathcal{L}^\dagger & 0 & L^\dagger & 0 \\
0 & L & 0 & \mathcal{L} \\
L^\dagger & 0 & \mathcal{L}^\dagger & 0
\end{pmatrix}.
\]

(2.67)

Table 2.1: The effect of the presence or absence of the inversion symmetry on the intra- (\( \mathcal{L} \)) and inter-band (\( L \)) SOC terms and on the energy splitting of spin-states for the same band, \( \varepsilon_{k,\uparrow} - \varepsilon_{k,\downarrow} \).

Table 2.1 summarizes the role of the inversion symmetry on the spin-orbit coupling parameters. For a material which has inversion symmetry, the Kramers theorem dictates (in the absence of a magnetic field) that \( \epsilon_{\uparrow}(k) = \epsilon_{\downarrow}(k) \) and thus
2.6. UNIFIED THEORY OF THE EY AND DP MECHANISMS

\( \mathcal{L} = 0 \). This term would otherwise split the spin degeneracy for the same band. When the inversion symmetry is broken, like in GaAs, \( \mathcal{L} \) is finite and the previous degeneracy is reduced to a weaker condition: \( \epsilon_\uparrow (k) = \epsilon_\downarrow (-k) \) as the time-reversal symmetry is retained in the absence of a magnetic field.

The authors considered the SOC to be the smallest energy scale in the model \( (\mathcal{L} (k_F), L (k_F)) \), while a competition of the other energy scales, namely \( \Delta_Z, \Gamma \) and \( \Delta \) was allowed. These can be of the same order of magnitude, as opposed to the conventional EY or DP case, where \( \Gamma \) is limited to small and large values with respect to the other energy scales, respectively. The quasi-particle scattering rate was treated to infinite order thus large values of \( \Gamma \) were possible.

The energy spectrum of the spins (or the ESR line-width) was calculated from the Mori-Kawasaki formula \([35, 36]\) using steps whose presentation is beyond the scope of the present thesis. As the main result, the authors found for the spin-relaxation rate the following expression:

\[
\Gamma_s = \frac{4 \Gamma |\mathcal{L} (k_F)|^2}{4 \Gamma^2 + \Delta_Z^2} + \frac{4 \Gamma |L (k_F)|^2}{4 \Gamma^2 + \Delta^2 (k_F)}.
\]  \hspace{1cm} (2.68)

According to Eq. (2.68), the contributions from intra- (\( \mathcal{L} (k_F) \)) and inter-band (\( L (k_F) \)) processes are additive to lowest order in the SOC and had a surprisingly similar form. A competition was observed between lifetime induced broadening (due to \( \Gamma \)) and the energy separation between states (\( \Delta (k_F) \) or \( \Delta_Z \)). The situation, together with schematics of the corresponding band-structures, is shown in Fig. 2.11. When the broadening is much smaller than the energy separation, the relaxation is EY-like, \( \Gamma_s \propto \Gamma \), even when the intra-band SOC dominates, i.e. for a material with inversion symmetry breaking. This situation was also studied in Ref. [37] and it may be realized in III-V semiconductors in high magnetic fields. For metals with inversion symmetry, this is the canonical EY regime.

When the states are broadened beyond distinguishability (i.e. \( \Gamma \gg \Delta (k_F) \) or \( \Delta_Z \)), spin-relaxation is caused by two quasi-degenerate states and the relaxation is of DP-type, \( \Gamma_s \propto 1/\Gamma \), even for a metal with inversion symmetry, \( \mathcal{L} = 0 \). For usual metals, the \( \Gamma \gg \Delta (k_F) \) criterion implies a breakdown of the quasi-particle picture as therein \( \Delta (k_F) \) is comparable to the bandwidth, thus this criterion means strong-localization. In contrast, metals with nearly degenerate bands remain metallic as e.g. MgB\(_2\) (Ref. [33]) and the alkali fullerenes (K\(_3\)C\(_{60}\) and Rb\(_3\)C\(_{60}\)) (Ref. [38]), which are strongly correlated metals with large \( \Gamma \). When the intra-band SOC dominates, i.e. for a strong inversion symmetry breaking, this is the canonical DP regime.

The real merit of Eq. (2.68) is that it provided the first hint that the EY and DP mechanisms could be treated in a common way and that these relaxation
mechanisms could lead to remarkably similar phenomenological results. As shown in the Results chapter, the same result can be obtained from an intuitive consideration which recognizes the similarity between the structure of the EY and DP model
Hamiltonians.
Chapter 3

Results

In this Chapter, the new results are presented which were obtained during my PhD work. I discuss the new results on spin-relaxation obtained for materials with inversion symmetry (3.1), without inversion symmetry (3.2), a unified description which is valid for both types of structures (3.3), and finally the effects of dephasing and the Loschmidt echo (3.4).

3.1 Spin relaxation in materials with inversion symmetry


3.1.1 Introduction and formulation of the problem

As mentioned in the Introduction, the Elliott relation, i.e. Eq. 2.15 became an important tool to validate whether spin relaxation in a given material follows the description of the Elliott-Yafet theory. To empirically verify Eq. 2.15, i.e. the Elliott-relation, knowledge of the spin-orbit matrix element between bands, the momentum relaxation time and the dimensionless $\alpha$ parameter for the given material is required. The most straightforward of the three parameters is the momentum relaxation time, since it is readily available for most metals as the function of temperature from resistivity measurements. The $\alpha$ parameter is less obvious, but it should be very similar for metals with similar crystal structure and chemistry (e.g. alkali atoms are expected to behave alike). Determination of the spin-orbit coupling matrix element is the most complicated, since theoretical calculations and simulations of SOC have proven to be difficult.
In order to perform the empirical verification, Monod and Beuneu gathered ESR linewidth data for many different metals [39], which is directly connected to the spin-relaxation times. For the SOC parameter they used SOC induced atomic splitting. For the band structure dependent parameter they used 1 as it had been predicted to be around unity. They scaled the data so that they could compare it to a universal Grüneisen function, which describes the momentum relaxation for all the metals they examined. Much of their work became a standard for our understanding of the spin-relaxation in elemental metals, however it has some shortcomings and widespread misinterpretations in the literature which motivated my work in Ref. [8].

First, the transport momentum-relaxation time scales with the transport electron-phonon coupling and the Debye temperature, which was neglected in the rescaling of Eq. 2.15. Second, their use of the SOC induced atomic energy splittings was only an estimation for the SOC matrix element in the Elliott-relation. It is not clear if the two are the same, even though one expects similar orders of magnitude (Fig. 3.1). Indeed, Fabian and Sarma showed that band-structure effects play an important role in aluminium and in other polyvalent metals[40, 41].

Herein, I show that in Ref. [39] the variation of the transport electron coupling constant and $T_D$ among the different metals were neglected, which affects the transport momentum scattering rate. I present an analysis which provides the empirical spin-orbit admixture coefficients. I also show that, even though the atomic spin-orbit coupling induced energy splittings have the same order of magnitude as the relevant SOC matrix element, the two quantities differ. This underlines the conceptual difference between atomic energy splittings and the matrix elements in a solid.

### 3.1.2 Momentum relaxation and the Elliott-Yafet theory

In Ref. [39] Monod and Beuneu investigated the scaling of the normalized ESR line-width with the transport momentum-relaxation time, $\tau_{tr}$, and found that the normalized ESR linewidth data falls on a universal Grüneisen function:

$$\Delta B \cdot \left(\frac{\Delta E}{L}\right)^2 = \text{const} \cdot \frac{T}{T_D} \cdot G_{MB} \left(\frac{T_D}{T}\right),$$

where $G_{MB}(x) = 4x^{-4} \left[ 5 \int_0^x \frac{z^4}{e^z - 1} - \frac{x^5}{e^x - 1} \right]$, where the constant was considered to be metal independent, and $T_D$ is the Debye temperature.

The $L/\Delta E$ spin-orbit admixture coefficient (SOAC) data were based on atomic spectra and were taken from Ref. [5]. The Grüneisen function, $G_{MB}$, used by
Monod and Beuneu was taken from Ref. [42]. The original paper, Ref. [39], did not explicitly mention the normalization with $T_D$. However since a single, "universal" Grüneisen function was argued to represent well the data [39], this presentation implies the $T_D^{-1}$ factor. This – as it is shown below – makes the value of the SOAC uncertain. The role of the spin-orbit coupling admixture is discussed further below and here first the focus is on the parameters of the transport momentum-scattering theory.

The contemporary description of the transport momentum-relaxation for alkali metals within the Debye-model assuming zero residual scattering reads [43]:

![Figure 3.1: The level scheme of the EY model demonstrating the relevant matrix elements of the Hamiltonian. Note that a small magnetic field induced splitting, with magnitude $2h$ was introduced in order to better show the otherwise degenerate bands.](image)
Figure 3.2: Measured ESR linewidth, normalized by the spin-orbit admixture parameter, for various materials as a function of the reduced temperature ($T/T_D$). This figure is from Ref. [39].

\[ \frac{1}{\tau_{\text{tr}}} = \frac{2\pi k_B}{\hbar} \lambda_\nu T \cdot G \left( \frac{T}{T_D} \right), \]

where \[ G(x) = \int_0^1 du \frac{u^5}{x^2 \sinh^2 (u/(2x))}, \] (3.2)
3.1. SPIN RELAXATION WITH INVERSION SYMMETRY

where $k_B$ and $\hbar$ are the Boltzmann and Planck constants, respectively and $\lambda_{tr}$ is the transport electron-phonon coupling constant. The two forms of the Grüneisen function, $G(x)$ and $G_{MB}(1/x)$, in Eqs. (3.1) and (3.2) are equivalent.

Eq. (3.2) when substituted into Eq. (2.15) reads for the normalized ESR line-width:

$$\Delta B \cdot \left( \frac{\Delta E}{L} \right)^2 = \alpha_1 \frac{2\pi k_B}{\gamma \hbar} \lambda_{tr} T \cdot G \left( \frac{T}{T_D} \right). \tag{3.3}$$

Here, $\gamma$ is the electron gyromagnetic ratio.

Clearly, an uncertainty remains due to the parameter $\alpha_1$, which is however supposed to be around unity and the same for all alkali metals [4]. Eq. (3.3) allows to introduce a universal function:

$$F(x) = \frac{2\pi k_B}{\gamma \hbar} x G(x), \tag{3.4}$$

which yields the final result of

$$\Delta B \cdot \left( \frac{\Delta E}{L} \right)^2 = \alpha_1 T_D \lambda_{tr} F \left( \frac{T}{T_D} \right). \tag{3.5}$$

This shows that the linewidth, that is normalized by the spin-orbit coupling coefficient, depends on $\lambda_{tr}$ and $T_D$ besides a universal function. The variation of the earlier two parameters among the alkali metals was not considered by Monod and Beuneu and it is discussed in the next section quantitatively.

3.1.3 Empirical verification of the Elliott-Yafet theory

As mentioned above, the left-hand side of Eq. (3.5) is proportional to $\alpha_1$, $T_D$ and $\lambda_{tr}$. However, Monod and Beuneu plotted the measured ESR line-widths while neglecting the variation of $T_D \cdot \lambda_{tr}$ among the alkali metals, even though it can amount to a factor 4.

In Table 3.1., the values of $\lambda_{tr}$ and $T_D$ are given for the four alkali metals. The SOAC values are also given as used by Monod and Beuneu for the scaling. I proceed with the analysis of the available data by using the values of $\lambda_{tr}$ and $T_D$ given in Table 3.1. The $\Delta B \cdot \left( \frac{\Delta E}{L} \right)^2$ data is taken from Ref. [39].

In Fig. 3.3., $\Delta B \cdot \left( \frac{\Delta E}{L} \right)^2 / T_D \lambda_{tr}$ is shown versus $T/T_D$. The universal $F(x)$ function from Eq. (3.4) is also shown. Clearly, the normalized line-width data do
Table 3.1: The electron-phonon coupling constants from Ref. [44] and Debye-temperatures from Ref. [45] of alkali elements. The \((L/\Delta E)^2\) values are given from Ref. [39] (in the original notation \((\lambda/\Delta E)^2\) ). The fitted values of \((L/\Delta E)^2\) are determined herein.

<table>
<thead>
<tr>
<th>Alkali element</th>
<th>(\lambda_{tr})</th>
<th>(T_D) [K]</th>
<th>atomic ((L/\Delta E)^2)</th>
<th>fitted ((L/\Delta E)^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na</td>
<td>0.14</td>
<td>158</td>
<td>2.73 \cdot 10^{-5}</td>
<td>3.81 \cdot 10^{-6}</td>
</tr>
<tr>
<td>K</td>
<td>0.11</td>
<td>91</td>
<td>2.06 \cdot 10^{-4}</td>
<td>8.99 \cdot 10^{-5}</td>
</tr>
<tr>
<td>Rb</td>
<td>0.15</td>
<td>56</td>
<td>3.16 \cdot 10^{-3}</td>
<td>2.96 \cdot 10^{-3}</td>
</tr>
<tr>
<td>Cs</td>
<td>0.16</td>
<td>38</td>
<td>1.91 \cdot 10^{-2}</td>
<td>3.08 \cdot 10^{-2}</td>
</tr>
</tbody>
</table>

Figure 3.3: The experimental \(\Delta B \cdot (\Delta E/L)^2 / \lambda_{tr} T_D\) plotted against \(T/T_D\). It is important to note that the atomic values of \((\Delta E/L)^2\) are used herein for the scaling (such as it was done by Monod and Beuneu). Solid curve shows the universal \(F(x)\) function after Eq. (3.4). Note that the line-width data do not fall on the same universal curve. The deviation being the largest for Na and smallest for Rb.
not fall on the same curve when the variation of $\lambda_{tr}$ and $T_D$ among the four alkali metals is taken into account. This means that the atomic SOC induced energy splitting per the energy difference between the adjacent states do not approximate well the real SOAC values. Accidentally, the data for Rb lies well on the plot indicating that then the proper SOAC value is well approximated by the atomic one.

![Figure 3.4: Comparison of the herein determined spin-orbit admixture coefficients and the values used by Monod and Beuneu in Ref. [39]. The values for Na are multiplied by 10 for better visibility. Note the agreement for Rb between the present values and those determined previously.](image)

Once the relevance of $T_D$ and $\lambda_{tr}$ is recognized, the experimental data is used to determine the experimental SOAC. In Fig. 3.4., the SOAC values which are determined herein and those considered by Monod and Beuneu in Ref. [39] are shown. One can observe a non-negligible difference between the values used previously and those which are obtained considering the role of $\lambda_{tr}$ and $T_D$. The difference is as large as a factor 3 for Na. Nevertheless, no independent experimental method is known besides ESR which could yield the SOAC parameters.

The present empirical values could be used as input for improved first principles calculations, which consider the band structure of these elements including spin-orbit coupling. Naturally, such calculations were unavailable at the time of Ref. [39], therefore our refinement of the values do not detract from the merit of the original work which highlighted the role of the atomic spin-orbit coupling.
CHAPTER 3. RESULTS

3.2 Spin relaxation in inversion symmetry breaking materials


3.2.1 Introduction and formulation of the problem

The D'yakonov-Perel’ model is well understood and it describes the $\Gamma \gg L$ regime of spin relaxation ($\Gamma$ is the quasiparticle broadening or the momentum scattering rate in energy units and $L$ is an average value of the SOC induced splitting of the conduction band). The result of the DP model, i.e. $\Gamma_s = L^2/\Gamma$ is used widely in the literature to benchmark the spin-relaxation properties of zincblende type semiconductors. However the opposite limit, i.e. when $\Gamma \ll L$ is less well known. Experiments on ultra-pure GaAs demonstrate[46, 47] the physical relevance of this regime and some theoretical attempts were made to describe its consequences in Ref. [48].

The $\Gamma \ll L$ limit is also briefly and qualitatively discussed in Ref. [3], which I discuss herein. It was argued that the observable spin relaxation is caused by a rapid dephasing of the spin ensemble due to the distribution of the SOC fields ($\Delta L$). The so-called reversible dephasing in NMR spectroscopy is analogous to this mechanism with the timescale known as $T_2^*$ (Ref. [20]). Actual memory loss is caused by the momentum scattering which can be at a much larger timescale. The observable spin-decay time in this case roughly equals the spin-dephasing time. The spin-relaxation rate (using our notation) was suggested to read in this limit as[3]:

$$\Gamma_s = \Delta L,$$  \hspace{1cm} (3.6)

where $\Delta L = \hbar \Delta \Omega$ is the width of the distribution of SOC splittings.

While this qualitative picture is probably correct under some circumstances, a more quantitative description is required. I show herein that the simple description breaks down depending on the relative magnitude of $\Gamma$ and $\Delta L$. Another interesting question is, whether the motional narrowing description of the D’yakonov Perel’ mechanism could be continued to the large SOC regime. This regime would be relevant for ultra-clean semiconductors.

These unresolved questions motivated me to study the full phase space of $(\Gamma, \langle L \rangle, \Delta L)$ with the case of large SOC emphasized, i.e. when $\Gamma \ll L$. Two distinct approaches are presented and compared: a semi-classical stochastic (Monte Carlo) method which directly follows the time evolution of individual spins and a
3.2. SPIN RELAXATION WITHOUT INVERSION SYMMETRY

\[ \Gamma \ll \hbar \langle \Omega \rangle \]

\[ \otimes \frac{2 \Gamma}{\hbar} \Delta \Omega \]

\[ A_r \]

\[ \Im \chi(\omega)/\omega \]

\[ \langle \Omega \rangle \]

\[ \langle \Omega \rangle \]

\[ \Gamma \gg \hbar \langle \Omega \rangle \]

\[ 2 \hbar \langle \Omega^2 \rangle / \Gamma \]

\[ \omega \] (in units of \[ \langle \Omega \rangle \])

Figure 3.5: Schematics of the D’yakonov-Perel’ model. The SOC related built-in magnetic fields give rise to a distribution of Larmor frequencies in the material (top left panel). The quasiparticle broadening function is also shown (bottom left panel). Under the action of the motional narrowing effect, the static distribution of the Larmor frequencies collapses into a single Lorentzian function.

diagrammatic technique, taking both SOC and impurity scattering into account. I studied the Bychkov-Rashba Hamiltonian for a two-dimensional electron gas with both methods. The two approaches yield quantitatively identical spin-relaxation times without any adjustable parameters. One can observe a multi-exponential spin decay with both methods in certain cases. This validates the Monte Carlo approach and presents a method which allows the calculation of \[ \tau_s \] (or \[ \Gamma_s \]) for an arbitrary SOC model, including e.g. the Dresselhaus SOC. I identify a yet unknown regime: when \[ \Gamma \gg \Delta L \], then \[ \Gamma_s \approx \Gamma \] is realized. For the case of \[ \Gamma \lesssim (\langle L \rangle, \Delta L) \] a strongly non-exponential spin decay is observed. All the results can be elegantly visualized by considering the evolution of the dynamic spin-susceptibility from the clean to the dirty limit (Fig. 3.5). The motional narrowing analogy remains valid in the large SOC regime.

3.2.2 A Monte Carlo simulation of spin-relaxation

Herein, I provide the details of the Monte Carlo calculations which I implemented, in order to obtain the spin relaxation properties of electron ensembles in semiconductors without inversion symmetry. The time evolution of the electron spins is studied with a stochastic approach, which models Boltzmann’s equation of the spin ensemble. Assuming a quasiparticle picture to be valid, an initially polarized spin ensemble travels in a solid with differing \( k \) momenta and where the intrinsic SOC induced magnetic fields (\( B(k) \)) are present. The spins precess freely around the
SOC fields with the angular frequency $\Omega(k)$ between two scattering events, where $k$ is a wave vector on the Fermi surface. This time evolution is demonstrated in Fig. 3.6. The time distribution of the scattering events are modeled as a Poisson-process with an expectation value of $1/\tau$. Each scattering event induces a uniformly random $k$ on the Fermi surface thus precession starts around a new axis. It is also assumed that the scattering does not affect the spin direction, i.e. the Elliott-Yafet type spin-flip mechanisms are not considered.

![Figure 3.6: The time evolution of a single spin on the Bloch sphere under the action of the SOC related magnetic fields. The spin freely precesses between scattering events and the vector of precession changes at the scattering events.](image)

The time difference between successive scattering events follow an exponential distribution with the Poisson $1/\tau$ parameter and in the simulations, the time is measured in $\tau$ units. This approach allows to keep $\tau = 1$ in all simulations and the SOC strength between simulations is varied.

The points of the scattering events are randomly generated using this distribution and the time evolution of individual spins are calculated exactly between scattering events. The average spin component for the spin ensemble is then sampled in uniform time intervals. The spin is measured in units of $\hbar/2$ for simplicity. The calculation starts with all spins fully polarized along the $z$ axis at $t = 0$.

The Fourier transform of the time dependent $s_z(t)$ is denoted by $S(\omega)$ and as I show below, it is closely related to the dynamic spin-susceptibility.

$$S(\omega) = \int_{-\infty}^{\infty} s(t)e^{i\omega t} dt.$$  

(3.7)
3.2. SPIN RELAXATION WITHOUT INVERSION SYMMETRY

3.2.3 Spin dynamics and the spin auto-correlation function

In the following, I prove that the above described Monte Carlo approach, which uses a fully spin polarized starting ensemble, is equivalent to calculating the spin autocorrelation function.

I consider a spin ensemble with \( s(t = 0) = [0, 0, 1] \) initial spin (measured in \( \hbar/2 \) units). Let \( f_1(t) = \langle s_z(t) \rangle_p \), where the \( \langle .. \rangle_p \) brackets denote average for a spin ensemble which is started from a polarized state.

Alternatively, one may choose a spin ensemble where \( s(t = 0) \) is uniformly chosen from the Bloch sphere. Such a spin density is a stationary solution of the Boltzmann-equation used by Pikus and Titkov [7]. Let \( f_2(t) = \langle s_z(0)s_z(t) \rangle_u \), where the \( \langle .. \rangle_u \) brackets denote the ensemble average from this initial (unpolarized spin states) condition. In what follows, I prove that \( f_2(t) = f_1(t)/3 \) regardless of the underlying Hamiltonian.

Let \( t_1, t_2, \ldots \) denote the time coordinates of the scattering events and \( \Omega_0, \Omega_1, \ldots \) the Larmor angular frequency vectors after each event (\( \Omega_0 \) is the initial Larmor frequency). The ensemble in our simulation can be described by a probability measure over these time coordinates and frequencies (denoted by \( \mu(\Omega_i, t_j) \)) and the probability measure over the initial spins (denoted by \( \nu(s_0) \)). This \( \mu \) measure fully captures the behavior of the Hamiltonian and momentum scattering in this stochastic model. \( \nu \) captures the initial conditions for the spins, therefore there are separate \( \nu_p \) and \( \nu_u \) measures for the ensembles introduced above. Ensemble averages are integrals over these measures:

\[
\langle s_z(t) \rangle = \iint s_z(t)d\mu(\Omega_i, t_j)d\nu(s_0),
\]

\[
\langle s_z(0)s_z(t) \rangle = \iint s_z(0)s_z(t)d\mu(\Omega_i, t_j)d\nu(s_0).
\]

The time evolution of single spin can be described by the action of a time dependent unitary matrix:

\[
s(t) = U(\Omega_i, t_j) s_0. \tag{3.9}
\]

Since the time evolution is a series of Larmor precession around different Larmor frequencies, \( U \) always describes a rotation, i.e. \( \det(U) = 1 \).

For \( f_1(t) \), all initial spins are the same. In the language of measures it reads:

\[
d\nu(s_0) = \delta(s_0 - [0, 0, 1])d^3s_0. \]

Substituting this and Eq. (3.9) into Eq. (3.8) yields:

\[
f_1(t) = \langle s_z(t) \rangle_p = \iint [U(\Omega_i, t_j)[0, 0, 1]]_z d\mu(\Omega_i, t_j). \tag{3.10}
\]

Since \( U \) describes a rotation, it can be parametrized by a rotation vector \( \alpha \), where \( \alpha = |\alpha| \) is the angle and \( \alpha/\alpha \) is the axis of the rotation. The rotation matrix
using this parametrization:

\[ U_{ij} = \frac{1}{\alpha^2} \left( \delta_{i,j} \alpha^2 \cos(\alpha) - \varepsilon_{i,j,k} \alpha_k \alpha \sin(\alpha) + \alpha_i \alpha_j (1 - \cos(\alpha)) \right), \]  

(3.11)

where \( \alpha \) has the same arguments as \( U \), \( \alpha = \alpha(\Omega_i, t_j, t) \). For the following equations these arguments are left out for brevity.

Substituting this parametrization into Eq. (3.10) yields:

\[ f_1(t) = \int \frac{\cos(\alpha)(\alpha_x^2 + \alpha_y^2) + \alpha_z^2}{\alpha^2} d\mu(\Omega_i, t_j). \]  

(3.12)

For the other ensemble, an initial condition with uniform spin distribution on the Bloch sphere were assumed. Using spherical coordinates the corresponding measure reads:

\[ d\nu_u(s_0)(\theta, \phi) = \frac{1}{4\pi} \sin(\theta)d\theta d\phi. \]  

(3.13)

Substituting this and Eq. (3.9) into Eq. (3.8) yields:

\[ f_2(t) = \frac{1}{4\pi} \int \int s_{0,z}[U(\Omega_i, t_j, t)s_0]_z \sin(\theta)d\theta d\phi d\mu(\Omega_i, t_j). \]  

(3.14)

Applying Eq. (3.11) and substituting the spherical parametrization of \( s_0 \) the inner integral can be evaluated:

\[ f_2(t) = \int \frac{\cos(\alpha)(\alpha_x^2 + \alpha_y^2) + \alpha_z^2}{3\alpha^2} d\mu(\Omega_i, t_j) = \frac{1}{3} f_1(t). \]  

(3.15)

\[ \langle s_z(0)s_z(t) \rangle_u = \frac{1}{3} \langle s_z(t) \rangle. \]  

(3.16)

This proves the equivalence of the two quantities as it was claimed above. I denote the Fourier-transform of \( \langle s_z(t) \rangle \) as \( S(\omega) \).

The dynamic (i.e. frequency dependent) spin-susceptibility, \( \chi(\omega) \), can be derived from the autocorrelation function, which allows a comparison to the dynamic spin-susceptibilities of different Hamiltonians, as it is discussed below. The spin-susceptibility is defined as the linear response to an external magnetic field:

\[ s(t) = \int_{-\infty}^{\infty} \chi(t') B(t - t') dt' \]  

\[ S(\omega) = \chi(\omega) B(\omega). \]  

(3.17)
3.2. SPIN RELAXATION WITHOUT INVERSION SYMMETRY

According to one of the forms of the fluctuation-dissipation theorem:

\[-\chi(\omega) = i\omega\beta \int_0^\infty e^{-i\omega t} A(t) dt - \beta A(0), \quad (3.18)\]

where \( \beta = 1/(k_B T) \) and \( A(t) \) is the physical spin-autocorrelation.

\[A(t) = \langle S(t)S(0) \rangle, \quad (3.19)\]

where \( S(t) \) is the magnetization of the whole electron-system. It is compelling to assume that \( f_2(t) \) (and according to the above result, \( f_1(t) \)) is proportional to the physical auto-correlation \( A(t) \), because the calculated spin-susceptibility from \( f_2(t) \) is proportional to the physical susceptibility in specific models as it is shows in the following section. If \( f_2(t) \propto A(t) \) holds, the following relationship holds:

\[\omega \text{Re}S(\omega) \propto \text{Im}\chi(\omega). \quad (3.20)\]

This relation provides a convenient way to obtain the dynamics spin-susceptibility, which in turn captures the essentials of the spin-dynamics: it width gives the relaxation rate and its position can unveil if any oscillatory behavior is present. Although I cannot provide a rigorous proof, I show its validity by a quantitative agreement for two specific cases: in the clean limit (i.e. when \( \Gamma \) is low) and in the dirty limit. For both cases, I take calculations which are available in the literature and I compare the simulation results with those.

3.2.4 Validation of the Monte Carlo simulation in the clean limit

The previous sections discussed that the time decay of a spin-polarized ensemble (described by \( s_z(t) \)) is calculated with a stochastic approach for both the clean and dirty cases. The real part of its Fourier transform, \( \text{Re}S(\omega) \) can be conveniently displayed in order to demonstrate the spin-relaxation properties. I also motivated (although I did not prove rigorously) that a relationship between \( \text{Re}S(\omega) \) and the dynamic spin-susceptibility, \( \chi(\omega) \) holds:

\[\omega \text{Re}S(\omega) \propto \text{Im}\chi(\omega). \quad (3.21)\]

The above relationship is now proven by a quantitative agreement which was numerically obtained by comparing the Monte Carlo results on \( S(\omega) \) in the \( x \) direction with analytic calculations of \( \chi_{xx}(\omega) \) for a particular Hamiltonian which is available in the literature (Ref. [49]). The result is shown in Fig. 3.7 with the calculation details as follows.
Erlingsson et. al. (Ref. [49]) calculated $\chi_{xx}(\omega)$ for a two-dimensional electron gas for a Hamiltonian containing both Bychkov-Rashba and Dresselhaus type SOC terms as follows:

$$H_0 = \frac{\hbar^2 k^2}{2m} + \frac{\mathcal{L}_R}{k_F} (s_x k_y - s_y k_x) + \frac{\mathcal{L}_D}{k_F} (s_y k_y - s_x k_x).$$

(3.22)

where $\mathcal{L}_R$ and $\mathcal{L}_D$ are the strengths of the Bychkov-Rashba and the Dresselhaus type spin-orbit couplings, respectively.

The dynamic spin-susceptibility was calculated in the absence of momentum relaxation and when $\mathcal{L}_R \sim \mathcal{L}_D \ll E_F$. Eq. 15 in Ref. [49] gives the dynamic spin-susceptibility for the $x$ direction as:

$$\chi_{xx}(\omega) = \lim_{\eta \to 0^+} \frac{m}{2\pi \hbar^2} \left(1+ \frac{(\omega + i\eta)^2}{\sqrt{(\frac{\mathcal{L}_R + \mathcal{L}_D}{\hbar})^2 - (\omega + i\eta)^2} \sqrt{(\frac{\mathcal{L}_R - \mathcal{L}_D}{\hbar})^2 - (\omega + i\eta)^2}}\right).$$

(3.23)
Fig. 3.7 demonstrates a good agreement between the two kinds of data which supports the connection between $S$ and $\chi$. This proves that the Monte Carlo method properly describes the spin dynamics in the clean limit, i.e. when scattering rate (or $\Gamma$) is much smaller than the relevant spin-orbit coupling terms.

### 3.2.5 Validation of the Monte Carlo simulation in the dirty limit

The spin-relaxation properties of a two-dimensional electron gas for a Bychkov-Rashba type SOC were calculated by Burkov and Balents\[37\] for an arbitrary magnitude of $\Gamma$, the SOC energy, and even including a magnetic field. However, for the present calculations, no external magnetic field is considered.

Burkov and Balents considered the following single electron Hamiltonian of the two-dimensional electron gas:

$$H = \frac{\hbar^2 k^2}{2m} + \frac{\mathcal{L}}{k_F} (s_x k_y - s_y k_x),$$  \hspace{1cm} (3.24)

where the first term is the kinetic energy, $k_F$ is the Fermi wavenumber, $s_{x,y}$ and $k_{x,y}$ are the spin and momentum components, respectively. The corresponding SOC related Larmor frequency reads:

$$\Omega(k) = \frac{\mathcal{L}}{\hbar k_F} [k_y, -k_x, 0].$$  \hspace{1cm} (3.25)

The calculation of Burkov and Balents is valid in a wide range of the parameters in the Hamiltonian. Although they applied it in the D’yakonov-Perel’ regime (i.e. for weak SOC), their result can also be used for an arbitrary strength of the SOC. They assumed impurity scattering and applied self-consistent Born approximation to calculate the so-called spin-diffusion propagator, $D(\omega)$. The spin-diffusion propagator is the vertex-correction in the presence of impurity scattering. The spin-susceptibility can be directly calculated from this propagator and it inherits its poles. In the $z$ direction (i.e. perpendicular to the plane of the 2DEG) it reads:

$$D_{zz}(\omega) = \frac{(-i\Gamma + \mathcal{L} - \hbar \omega)(i\Gamma + \mathcal{L} + \hbar \omega)}{-\hbar^2 \omega^2 - i\hbar \omega + \mathcal{L}^2}.$$  \hspace{1cm} (3.26)

The real and imaginary parts of the two poles ($\omega_{1,2}$) of Eq. (3.26) correspond to the oscillation frequency and the damping of the average spin time evolution, respectively. The poles read:

$$\hbar \omega_{1,2} = -i\Gamma \pm \sqrt{4\mathcal{L}^2 - \Gamma^2}.$$  \hspace{1cm} (3.27)

The spin relaxation time is obtained from the poles as:
\[
\frac{1}{\tau_s} = -\text{Im}\omega_{1,2}.
\] 

(3.28)

Similar results can be obtained for the diffusion propagator which is perpendicular to the quantization axis \(D_{xy}\) and the poles are the roots of a third order polynomial:

\[
(h\omega)^3 + 2i\Gamma(h\omega)^2 - (\Gamma^2 + L^2)h\omega - \frac{L^2\Gamma}{2} = 0.
\] 

(3.29)

I used these results for a comparison with the Monte Carlo simulations on the same Bychkov-Rashba SOC model system. Two typical time evolutions of spin-ensemble polarization are shown in Fig. 3.8 for two different regimes: when the quasiparticle broadening is comparable to the SOC and when the quasiparticle broadening dominates. The latter is the usual D'yakonov-Perel' regime. An exponential decay of the ensemble spin polarization is observed with and without an oscillating component for both cases.

The real part of the Fourier transform of the time dependent \(s_z(t)\) data, \(\text{Re}[S(\omega)]\) is also shown in Fig. 3.8, which demonstrate better the presence of the oscillation (peaks at \(\omega \neq 0\)) and a single decay (a nearly Lorentzian peak at \(\omega = 0\)). Two Lorentzian curves are fitted to \(\text{Re}[S(\omega)]\), whose position and width give the frequency and relaxation time of the damped oscillation, respectively. These parameters are compared to the analytic calculations for the same Bychkov-Rashba Hamiltonian performed in Ref. [37]. The clean limit (\(\Gamma = 0\)) would yield a \(\text{Re}[S(\omega)]\) with two Dirac delta peaks at \(\omega = \pm\Omega\). The evolution of these two Dirac delta functions with increasing \(\Gamma\) is discussed further below but is recaptured briefly herein: for small \(\Gamma\) values, the Dirac delta functions start to broaden and form two Lorentzian functions and eventually they collapse to common line on the origin, but the latter consists of two Lorentzian components as our calculations and also the analytical calculations have shown.

I validate the above described Monte Carlo approach by comparing the numerical results with the diagrammatic results from Ref. [37]. The comparison is shown in Fig. 3.9. The real and imaginary part of the poles of the spin-diffusion propagator from (3.27) and (3.29) are shown in Fig. 3.9 with solid curves. The real part represents the frequency of the oscillation component, whereas the imaginary part describes the damping and it is the spin-relaxation rate in angular frequency units. A multi-exponential fit (allowing for oscillatory results) to the numerically obtained time evolution of the magnetization yield the same frequency and damping quantities. This result is shown in Fig. 3.9 with symbols. It is clear from the figure that the two types of data agree surprisingly well without any adjustable parameters. This agreement is quite reassuring from the point of view of the Monte Carlo method and in our opinion validates that the approach is well founded to
Figure 3.8: Simulation of the time dependent average spin component for an ensemble of electrons under the Bychkov-Rashba SOC Hamiltonian for \(\hbar\Omega = \Gamma\) (upper panel) and \(\hbar\Omega = 0.4\Gamma\) (lower panel). The thick solid curve in the upper left graph is the averaged \(s_z\), the thin solid curves are the spin components of a few individual electrons. The dashed curve show the oscillating spin component in the absence of momentum scattering. The real part of the Fourier transform of the simulated \(s_z(t)\), \(\text{Re}\mathcal{S}(\omega)\) (square symbols) and Lorentzian fits (solid curves) as explained in the text, are also shown.
Figure 3.9: Spin-relaxation parameters as obtained from the diagrammatic technique (solid lines) and the Monte Carlo simulations (symbols) for the two orientations of the initial spin polarization. The real part is the angular frequency of the damped oscillation and the imaginary part is the spin-relaxation rate in angular frequency units. Vertical blue arrows show the two distinct regimes which were reported in the previous figure. The two kinds of data fit well without introducing any scaling parameter. Horizontal arrow indicates the conventional DP regime. The scattering and large error of some parameter values is the effect of a less reliable fit due to a vanishing spectral weight of the corresponding poles.
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a two-dimensional electron gas and the same approximation (namely neglecting the SOC splitting of the band structure) but the methods are quite different.

The conventional DP regime is marked in Fig. 3.9 and both the diagrammatic and Monte Carlo methods give $\Gamma_s = L^2/\Gamma$ (i.e. we obtain $\alpha = 1$ for the band-structure dependent constant) for the relaxation in the $z$ direction. However, the figure also indicates the presence of two additional, previously unknown spin-relaxation regimes: on the far right, when $\Gamma \lesssim L$, two oscillating components are present with a broadening of $\Gamma_s = \alpha \Gamma$ (where $\alpha = 0.5$). Another spin-relaxation regime occurs right below the “bifurcation point” ($L = 0.5\Gamma$): therein two relaxation components with different damping are present, i.e. it represents a non-single-exponential relaxation. This regime crosses over smoothly to the conventional DP regime where a single exponential damping is observed: the weight of the component with the faster relaxation gradually disappears. The scattering and error of the fitting in this regime is caused by the diminishing weight of the faster relaxing component.

3.2.6 Analogy to conventional motional narrowing

The conventional DP mechanism is often described as a motional narrowing effect, which yields the $\Gamma_s \propto \Gamma^{-1}$ behavior. Motional narrowing is the extremal case of a theory known as “the effect of motion on the spectral lines”, which is well developed in e.g. NMR spectroscopy[19]. I show herein that the so-called two-site NMR motional narrowing problem (after Chapter X. in Ref. [19]) gives numerically identical results (when the corresponding parameters are considered) to the above described spin-relaxation problem with a Bychkov-Rashba SOC for the full phase space, including the SOC strength and $\Gamma$. The two situations, i.e. momentum scattering of an electron between different $\Omega(k)$ and motion of a nucleus between different sites with different Larmor frequencies leads to the same result. An interesting consequence is that the analogy is not restricted to the conventional DP regime but it applies to the whole domain of interest.

Abragam[19] considered the two-site NMR motional narrowing problem: a nucleus is allowed to jump with the transition rate $\Gamma_c = 1/\tau_c$ between two sites with different Larmor frequencies: $\pm \Omega$ around a central Larmor frequency (defined as zero in this case). The resulting NMR lineshape is shown in Fig. 3.10 for a fixed $\Omega = 1$ and different values of the jumping frequency, $\Gamma_c$. The analogy between the spin relaxation and the motional narrowing is clear: the $\pm \Omega$ local Larmor frequencies in NMR correspond to the built-in Zeeman field distribution of the spin-relaxation problem (that is two Dirac delta functions for the Bychkov-Rashba SOC) and the jumping frequency ($\Gamma_c$) of the motional narrowing problem corresponds to the $\Gamma$ momentum relaxation rate (besides a factor of 2 which is discussed below). The analogy can be quantified for the simplest case as follows.
Abragam derived that the spectral shape, \( I(\omega) \) for the two-site NMR motional narrowing problem reads:

\[
I(\omega) = \text{Re} \left( \frac{2i\omega + 4\Gamma_c}{(\Omega^2 - \omega^2) + 2i\omega\Gamma_c} \right), \tag{3.30}
\]

The denominator of Eq. (3.30) has poles at \( \omega_{1,2} = i \left( \Gamma_c \pm \sqrt{\Gamma_c^2 - \Omega^2} \right) \), i.e. Eq. (3.30) can be rewritten as:

\[
I(\omega) = \text{Re} \left( \frac{A}{\omega - \omega_1} + \frac{B}{\omega - \omega_2} \right), \tag{3.31}
\]

where \( A = -i\frac{\Gamma_c + \Delta}{\Delta} \) and \( B = i\frac{\Gamma_c - \Delta}{\Delta} \), where \( \Delta = \sqrt{\Gamma_c^2 - \Omega^2} \). Evaluation of Eqs. (3.30) and (3.31) yields the curves shown in Fig. 3.10.

\( \Gamma \), as defined as the momentum relaxation rate, differs from \( \Gamma_c \) in the work of Abragam by a factor of 2 as discussed herein. \( \Gamma \) corresponds to the relaxation rate
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of a non-equilibrium population of the different $k$ states. $\Gamma_c$ corresponds to the transition rate between two states. The rate equations for the populations of the two state, $n_{1,2}$ read:

$$\dot{n}_1 = -\frac{1}{\tau_c} (n_1 - n_2),$$
$$\dot{n}_2 = \frac{1}{\tau_c} (n_1 - n_2),$$
$$\partial_t(n_1 - n_2) = -\frac{2}{\tau_c} (n_1 - n_2).$$

(3.32)

The analogous momentum relaxation rate in this case is $1/\tau_m = 2/\tau_c$, so $\Gamma = 2\Gamma_c$.

![Figure 3.11: Real and imaginary parts of the $\omega_{1,2}$ roots as defined above from the NMR motional narrowing problem.](image)

With this substitution, the real and imaginary values of the above defined $\omega_{1,2}$ are shown in Fig. 3.11. The real part of the roots describe the position of the two peaks and the imaginary parts describe the linewidths in agreement with Fig. 3.11. Remarkably, this figure is identical to the spin-relaxation problem for the 2D Rashba model as presented above with a straightforward identification of the correspondence of the two parameters.

3.2.7 Non-exponential spin polarization decay

The agreement between the spin-relaxation parameters as obtained from the Monte Carlo and from the analytic calculations validates the use of the numerical method for the two-dimensional electron gas with the Bychkov-Rashba SOC. Although it represents no formal proof, I believe that it justifies the use of the Monte Carlo
method to obtain spin-relaxation parameters and eventually ReS(ω) for more complicated distributions of the SOC fields, where analytic calculations are not available.

Figure 3.12: The spin dynamics in the presence of the Dresselhaus spin-orbit coupling (from Eq. (3.33) with \( \mathcal{L} = 1 \)) for different momentum scattering rates (Γ). The horizontal arrow indicates the \( \Delta \mathcal{L} \) distribution width of the SOC. Note the presence of a Dirac delta function at \( \omega = 0 \) in ReS(ω) in the clean limit and the gradual broadening with increasing Γ. The beating pattern in the time domain for Γ = 0 is not noise and is related to the details of the Dresselhaus SOC. The D'yakonov-Perel’ limit manifests itself as an exponential time decay in \( s_z \) and a single Lorentzian at \( \omega = 0 \) for the ReS(ω). The novel regime is identified in the middle graph: an initial rapid decay is followed by a longer exponential decay.

The choice of the 2D electron gas with the Bychkov-Rashba SOC is somewhat exceptional as its ReS(ω) in the clean limit consists of two Dirac delta functions (when quantization is along the z axis), i.e. this model has a zero width of the SOC field distribution. Generally, the width of the SOC field distribution (\( \Delta \mathcal{L} = \hbar \Delta \Omega \)) could be sizeable, i.e. comparable to the average SOC field (\( \langle \mathcal{L} \rangle = \hbar \langle \Omega \rangle \)). In fact, the Dresselhaus SOC represents such a case. Its Larmor frequency distribution
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reads:

$$\Omega(k) = \frac{L}{\hbar k_F^3} \left[ k_x \left( k_y^2 - k_z^2 \right), k_y \left( k_z^2 - k_x^2 \right), k_z \left( k_x^2 - k_y^2 \right) \right]. \quad (3.33)$$

The corresponding Re$\mathcal{S}(\omega)$ in the clean limit is shown in Fig. 3.12 obtained with $\mathcal{L} = 1$. The SOC field distribution is sizeable and $\Delta\mathcal{L}$ and $\langle \mathcal{L} \rangle$ have the same order of magnitude.

Two features are observed for this type of SOC in the clean limit: a Dirac delta function in Re$\mathcal{S}(\omega = 0)$ and beating-like pattern in $s_z(t)$. While similar in its form, the Re$\mathcal{S}(\omega)$ function is not identical to the histogram of the $|\Omega(k)|$ Larmor frequency distribution, which is evident by the presence of the Dirac delta function in the earlier. This is due to some geometric factors which appear in the calculation of Re$\mathcal{S}(\omega)$ and is discussed in the Appendix (A.3). The beats in the time domain data are the consequence of coherent spin oscillations and its details are specific for the angular distribution of the SOC field.

Fig. 3.12. also presents the time dependence of $s_z$ and Re$\mathcal{S}(\omega)$ for finite $\Gamma$. The D'yakonov-Perel' limit is recovered when $\Gamma$ is much larger than $\langle \mathcal{L} \rangle$ and $\Delta\mathcal{L}$. However, a novel regime is identified when $\Gamma \lesssim (\langle \mathcal{L} \rangle, \Delta\mathcal{L})$. Then, an initial rapid dephasing due to the distribution of SOC fields is present in agreement with Ref. [3], however the dephasing is not complete (i.e. $s_z \neq 0$) and the remaining ensemble $s_z$ decays on the timescale of $\tau = \hbar / \Gamma$ only. This feature is due to the presence of the Dirac delta function in addition to the SOC fields at $\omega \neq 0$. This observation mimics the situation encountered in pulsed NMR spectroscopy[20]: therein a rapid dephasing (on a timescale denoted as $T_2^*$) is caused by local magnetic field inhomogeneities which is not accompanied by a true information loss. It is followed by a true relaxation (a timescale denoted as $T_2$ or in the absence of an external field $T_1 = T_2$) where the information is inevitably lost. This phenomenon leads to the presence of NMR spin-echo, i.e. the spins can be restored in-phase on a timescale within $T_2$ with a suitable external excitation. Our observation predicts that a similar scheme may lead to the observation of spin-echo in semiconductors under the circumstances which correspond to the situation shown in Fig. 3.12 and is discussed in depth in Section 3.4.

I believe that the well-known Dresselhaus Hamiltonian is general enough to properly capture the essential features of spin-relaxation for the entire $(\Gamma, \langle \mathcal{L} \rangle, \Delta\mathcal{L})$ phase space. It is also the most important Hamiltonian which is relevant for most III-V semiconductors where bulk spatial inversion symmetry breaking occurs. I summarize our qualitative findings in a compact form in Table 3.2., which is briefly repeated herein: i) the conventional DP regime occurs when $\Gamma$ is much larger than the SOC, ii) the spin decay is oscillatory and spin-relaxation time has the same order of magnitude as momentum relaxation time when the SOC is significant but its distribution is sharp: $\Delta\mathcal{L} \ll \Gamma \lesssim \langle \mathcal{L} \rangle$, iii) an NMR-like rapid spin dephasing followed by a true spin-relaxation occurs when $\Gamma \lesssim (\langle \mathcal{L} \rangle, \Delta\mathcal{L})$. 
Table 3.2: Summary of the relaxation regimes which are encountered for different values of the momentum scattering rate $\Gamma$, average SOC energy $\langle \mathcal{L} \rangle$ and its spread $\Delta \mathcal{L}$.

<table>
<thead>
<tr>
<th>Condition</th>
<th>Relaxation type</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Gamma \gg (\langle \mathcal{L} \rangle, \Delta \mathcal{L})$</td>
<td>exponential (DP regime), $\Gamma_s = \alpha \mathcal{L}^2 / \Gamma$</td>
</tr>
<tr>
<td>$\Delta \mathcal{L} \ll \Gamma \lesssim \langle \mathcal{L} \rangle$</td>
<td>oscillatory + exponential decay, $\Gamma_s = \alpha \Gamma$</td>
</tr>
<tr>
<td>$\Gamma \lesssim (\langle \mathcal{L} \rangle, \Delta \mathcal{L})$</td>
<td>non-exponential, $T_2 = h / \Delta \mathcal{L}, T_{1,2} = h / \Gamma$</td>
</tr>
</tbody>
</table>

I finally comment on the most important approximation of my approach, i.e. that the SOC is smaller than the kinetic energy. This approximation is valid for most technically relevant semiconductors but the opposite is true for heavy elements such as e.g. Bi, where the SOC becomes the leading energy term. Those materials, however, are characterized by very unconventional spin-dynamical properties whose study was attempted in e.g. Ref. [50] and this discussion is beyond the scope of the present thesis.

### 3.2.8 Anisotropic spin-relaxation

For an isotropic Hamiltonian, the time evolution of each spin-component is identical and is independent of each other. For an exponential relaxation, like for the DP regime, it can be described with a single $\tau_s$ spin-relaxation parameter.

$$\dot{s}_i = -\frac{1}{\tau_s} s_i, \quad (3.34)$$

where $i$ denotes the coordinate component.

The linear response to an external magnetic field can be generally described with a single, scalar dynamic spin-susceptibility, $\chi(\omega)$.

$$s_i(\omega) = \chi(\omega) B_i(\omega). \quad (3.35)$$

As we have shown previously, this can be used to describe the whole phase space of spin relaxation, except that it cannot account for spin-dephasing effects, which are discussed in Section 3.4.

For an anisotropic Hamiltonian, non-independent spin-relaxation terms are expected, which can be described using cross-susceptibilities between magnetic and spin components and their most general form reads:

$$s_i(\omega) = \chi_{i,j}(\omega) B_j(\omega). \quad (3.36)$$
\[ -\chi_{i,j}(\omega) = i\omega \beta \int_0^\infty e^{-i\omega t} A_{i,j}(t) dt - \beta A_{i,j}(0), \]  
(3.37)

where \( A_{i,j}(t) \) is the spin auto-correlation between spin components \( i \) and \( j \).

However, generalization of Eq. (3.16) is straightforward for cross correlations. Namely, if a simulation is started from a spin polarized state in the direction \( i \) and the average spin of the ensemble is measured in the direction \( j \) in the function of time, then the result is proportional to the spin auto-correlation between the spin components \( i \) and \( j \).

3.3 Intuitive approach to the unified theory of spin relaxation


3.3.1 Introduction and formulation of the problem

The spin-relaxation time, \( \tau_s \), depends on the momentum relaxation time, \( \tau \), however with the opposite dependence: \( \tau_s \propto \tau \) in the EY theory whereas \( \tau_s \propto 1/\tau \) in the DP theory. The different phenomenology has become a benchmark for the classification of different spin-relaxation mechanisms in a novel material (e.g. graphene [24, 25, 26, 51, 52, 53, 54, 55]) into one or the other mechanism.

The different foundations and different phenomenology was the reason why no apparent links between the two mechanisms were discovered. This is despite the fact that Yafet realized in his seminal paper [56] that e.g. near band degeneracies the EY theory should inevitably break down and additional discussion is required but this was not further pursued\(^1\).

The first hint toward a possible unification came from electron spin relaxation studies of materials with spatial inversion symmetry and a short momentum relaxation time, e.g. MgB\(_2\) (Ref. [57]), a material which displays both phenomenology: \( \tau_s \propto \tau \) for temperatures below 150 K and \( \tau_s \propto 1/\tau \) for \( T > 400 \) K and an intermediate range in between. Another material with a short \( \tau \) and retained inversion symmetry is Rb\(_3\)C\(_{60}\) [58] which displays \( \tau_s \propto 1/\tau \) in the whole temperature range.

---

\(^1\)Yafet mentions (p. 81 in Ref. [56]): "In the degenerate valence bands, the relaxation time is expected to be much shorter since the matrix element for scattering with spin reversal from one band to the other is not restricted, as the intraband matrix element is, by time reversal or space inversion."
The experiments in both materials were successfully explained by the so-called generalized EY theory (GEY) in Refs. [33, 34]. It was shown in Ref. [14] that the spin relaxation expressions for inversion breaking and inversion retaining materials are analogous in a wide regime of the quasi-particle broadening (Eq. (2.68)).

![Diagram of band structure schematics and quasiparticle broadening for the D'yakonov-Perel' (a) and Elliott-Yafet (b) models, i.e. for inversion breaking (a) and inversion retaining (b) materials, respectively. $\mathcal{L}$ is the SOC matrix element, $\Delta_Z$ is the Zeeman-splitting, $\Delta$ is the band gap and $\Gamma$ is the quasi-particle broadening.](image)

Figure 3.13: Band structure schematics and quasiparticle broadening for the D’yakonov-Perel’ (a) and Elliott-Yafet (b) models, i.e. for inversion breaking (a) and inversion retaining (b) materials, respectively. $\mathcal{L}$ is the SOC matrix element, $\Delta_Z$ is the Zeeman-splitting, $\Delta$ is the band gap and $\Gamma$ is the quasi-particle broadening.

The analogy is even more compelling by visualizing the analogous physical quantities on electron level scheme, as shown in Fig. 3.13. The DP description is also known to result in an EY-like phenomenology in e.g. inversion symmetry breaking semiconductors with a very long $\tau$ or in high magnetic fields [3, 7, 37].

In this section, I intend to shed light on this link and I show that formally the DP and EY Hamiltonians can be transformed to each other. This allows to derive the results of both mechanisms in the framework of the other. This is explicitly shown for the EY and GEY mechanisms: the results of spin-relaxation for these cases are derived from a DP-like approach using the quasiparticle picture combined with internal magnetic fields.

This not only provides a unified treatment of spin-relaxation but also provides a readily programmable tool to study spin-dynamics in metals and semiconductors.

### 3.3.2 Equivalence of the EY and DP Hamiltonians

The Elliott-Yafet model considers 4 electron bands, two of which remain pairwise degenerate even in the presence of an intrinsic (i.e. atomic) SOC. This is the direct consequence of the Kramers theorem in the presence of inversion symmetry. An external magnetic field however breaks the time-reversal symmetry and splits the degenerate bands. An external electric field could also break the inversion
3.3. INTUITIVE APPROACH TO THE UNIFIED THEORY

symmetry and also split the bands, as it was seen for the Bychkov-Rashba type SOC. The most general description of a 4-band Hamiltonian in the presence of SOC but retained inversion symmetry reads:

$$H_{\text{EY}} = \begin{pmatrix} 1^{\uparrow} & 1^{\downarrow} & 2^{\uparrow} & 2^{\downarrow} \\ 1^{\uparrow} & 0 & 0 & L_{2,k} \\ 1^{\downarrow} & 0 & 0 & L_{1,k} \\ 2^{\uparrow} & L_{2,k} & L_{1,k} & \Delta_k \\ 2^{\downarrow} & -L_{1,k} & -L_{2,k} & 0 \end{pmatrix}, \quad (3.38)$$

where 1 and 2 label the kinetic states, $L_{i,k}$ are the SOC matrix elements of which $L_{2,k}$ is real.

The D’yakonov-Perel’ Hamiltonian considers a $k$ dependent splitting between the two spin sublevels of the conduction band, with splitting $\mathcal{L}_k$ and it reads:

$$H_{\text{DP}} = \begin{pmatrix} \uparrow & \downarrow \\ \uparrow & 0 \\ \downarrow & \mathcal{L}_k \\ \downarrow & 0 \end{pmatrix}, \quad (3.39)$$

Clearly, the DP Hamiltonian is based on spin eigenstates. The connection between the effective magnetic field and the spin-Hamiltonian are: $\hbar |\Omega_k| = 2\mathcal{L}_k$.

The main idea is to rename electron states in a way that transforms the Hamiltonian in Eq. 3.38 to two independent, DP-like $2 \times 2$ Hamiltonians. In this way, one can describe the same system in either the EY or DP pictures (Fig. 3.14).

The states in Eq. (3.38) are renamed in the following way:

$$|A^{\uparrow}\rangle = |1^{\uparrow}\rangle$$
$$|A^{\downarrow}\rangle = |2^{\downarrow}\rangle$$
$$|B^{\uparrow}\rangle = |2^{\uparrow}\rangle$$
$$|B^{\downarrow}\rangle = |1^{\downarrow}\rangle.$$ \hspace{1cm} (3.40)

After the renaming, the Hamiltonian reads:

$$H_{\text{EY,renamed}} = \begin{pmatrix} A^{\uparrow} & A^{\downarrow} & B^{\uparrow} & B^{\downarrow} \\ A^{\uparrow} & 0 & L_{1,k} & L_{2,k} \\ A^{\downarrow} & L_{1,k} & 0 & -L_{2,k} \\ B^{\uparrow} & L_{2,k} & 0 & \Delta_k \\ B^{\downarrow} & -L_{2,k} & L_{1,k} & 0 \end{pmatrix}, \quad (3.41)$$

We note that in the absence of the $L_{2,k}$ terms, this would be already a block-diagonal Hamiltonian. This intuitive renaming is equivalent to a unitary transformation described by the following matrix:
Figure 3.14: A demonstrative picture to visualize the present approach: the conventional EY approach considers mixed (i.e. perturbed), but degenerate spin states. The novel approach suggests to transform the Hamiltonian instead, which yields two pseudo-spin subspaces which are not connected. The two pseudospin subspaces are represented by the direct sum of two DP Hamiltonians.

\[ H_{\text{EY, renamed}} = P^\dagger H_{\text{EY}} P, \]
\[ P = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \end{bmatrix}. \]  

(3.42)

The \( \pm L_{2,k} \) matrix elements can be eliminated by a two dimensional rotation between the new kinetic states.

\[ |A'\rangle = a |A\rangle - b |B\rangle, \]
\[ |B'\rangle = b |A\rangle + a |B\rangle, \]  
\[ a^2 + b^2 = 1, \quad a, b \in \mathbb{R}. \]  

(3.43)

This rotation can also be written in the form of a unitary matrix:
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\[ H_{\text{EY,transformed}} = R^\dagger H_{\text{EY,renamed}} R, \]

\[ R = \begin{bmatrix} a & 0 & b & 0 \\ 0 & a & 0 & b \\ -b & 0 & a & 0 \\ 0 & -b & 0 & a \end{bmatrix}. \] \quad (3.44)

The two subsequent transformations can be combined into a single unitary transformation:

\[ H_{\text{EY,transformed}} = U^\dagger H_{\text{EY}} U, \]

\[ U = PR = \begin{bmatrix} a & 0 & b & 0 \\ 0 & -b & 0 & a \\ -b & 0 & a & 0 \\ 0 & a & 0 & b \end{bmatrix}. \] \quad (3.45)

With carefully chosen \( a \) and \( b \) values the \( 2 \times 2 \) off-diagonal blocks of (3.41) can be eliminated.

\[ \Delta_k' = \sqrt{\Delta_k^2 + 4L_{2,k}^2}, \]

\[ a_k = \sqrt{\Delta_k' + \Delta_k} / 2\Delta_k', \]

\[ b_k = \text{sign}(L_{2,k}) \sqrt{\Delta_k' - \Delta_k} / 2\Delta_k'. \] \quad (3.46)

Applying the transformation yields the following Hamiltonian:

\[ H_{\text{EY,transformed}} = \begin{bmatrix} \begin{bmatrix} A'_{\uparrow} & A'_{\downarrow} & B'_{\uparrow} & B'_{\downarrow} \\ A'_{\uparrow} & 1/2(\Delta_k - \Delta_k') & L_{1,k} & 0 \\ A'_{\downarrow} & L_{1,k}^* & 1/2(\Delta_k + \Delta_k') & 0 \\ B'_{\uparrow} & 0 & 1/2(\Delta_k - \Delta_k') & L_{1,k} \\ B'_{\downarrow} & 0 & 0 & 1/2(\Delta_k + \Delta_k') \end{bmatrix} \\ \begin{bmatrix} H_A & 0 \\ 0 & H_B \end{bmatrix} \end{bmatrix}. \] \quad (3.47)

The EY Hamiltonian is block diagonalized in a way, that does not mix spin-up and spin-down states.

The \( 2 \times 2 \) blocks can be written as following:
\[ H_{A,B} = \frac{\Delta_k}{2} + \frac{\hbar}{2} \tilde{\sigma} \Omega_{A,B}(k), \]
\[ \Omega_A(k) = \frac{2}{\hbar} \begin{pmatrix} -\text{Re}[L_{1,k}] \\ \text{Im}[L_{1,k}] \\ \Delta_k/2 \end{pmatrix}, \]
\[ \Omega_B(k) = \frac{2}{\hbar} \begin{pmatrix} -\text{Re}[L_{1,k}] \\ \text{Im}[L_{1,k}] \\ -\Delta_k'/2 \end{pmatrix}, \]

which is similar to the DP Hamiltonian in (2.39), except for the presence of a spin independent offset energy, which does not alter the spin dynamics in a given \( k \). This allows to formally treat the EY spin-relaxation mechanism with the usual DP-like approach, i.e. by the evolution of the pseudo-spin ensemble upon internal built-in magnetic fields.

Note that the \( \tilde{\sigma} \) operator no longer acts on the physical spin but herein it is a pseudo-spin operator and is not identical to \( \sigma \) which is a vector composed of the Pauli matrices in the original basis. The \( \tilde{\sigma}_x \) and \( \tilde{\sigma}_y \) operators in this picture mix spin states of different kinetic bands from the original EY basis. However the \( \tilde{\sigma}_z \) operator coincides with the \( \sigma_z \) operator defined in the original EY basis, so the derived time dependence of the \( \tilde{\sigma}_z \) in this DP like basis remains physical.

In the next step, I demonstrate that the DP Hamiltonian, Eq. (3.39), can be transformed into an Elliott-Yafet like Hamiltonian. Clearly, this two band Hamiltonian requires two additional states and also another factor which acts like a kinetic energy gap. This is achieved by introducing an external magnetic field, with Zeeman energy \( \Delta_Z \), and amend the system with a pair of virtual electron spin states, which have the same SOC but interact with the external magnetic field with opposite sign as compared to the physical electron states. This gives:

\[ H'_{DP} = \begin{bmatrix} 0 & L_k & 0 & 0 \\ L_k^* & \Delta_{Z,k} & 0 & 0 \\ 0 & 0 & L_k & \Delta_{Z,k} \\ 0 & 0 & \Delta_{Z,k} & L_k^* \end{bmatrix}. \]

The zero of the energy is also shifted as the Zeeman term would normally introduce a \( \pm \Delta_Z/2 \) shift of the levels. We retained the \( k \) dependence of the Zeeman term as the electron \( g \)-factor can be in general \( k \)-dependent. Clearly, a similar transformation of the states, identical to that presented above, yields the EY Hamiltonian for the DP problem. We emphasize that the \( L_k \) terms originate from an SOC due to inversion symmetry breaking, still it can be rewritten as if it was coming from a system with retained inversion.
I note that the additional virtual electron states are not connected with the true physical electrons, it is thus plausible that the usual treatment of spin-relaxation in inversionally symmetric materials, i.e. first order perturbation theory for the conventional EY and many-body approach for the GEY, can be performed for these states, too.

### 3.3.3 Fitting simulation results to the spin relaxation of MgB$_2$

The above presented transformation between the EY-GEY and DP pictures not only represents an intuitive description but it also allows for a practical and implementable calculation for spin-relaxation times. In the original EY theory and in particular in the GEY regime, it is difficult to implement the direct calculation of the spin-relaxation time. In contrast, the DP treatment is more intuitive: the time evolution of the spin ensemble is caused by effective intrinsic SOC fields. I demonstrate this for MgB$_2$: in this material a crossover between the EY and GEY regimes was observed experimentally using electron spin resonance measurements [57, 33].

Before presenting the result, I briefly summarize the experimental situation about the spin-relaxation properties of MgB$_2$. The first electron spin resonance measurements in 2001 by Simon et al. (Ref. [57]) indicated that the ESR linewidth, $\Delta B$, is anomalous: it is proportional to the resistivity, $\rho$, in the 40-200 K range, thus $\Gamma_s \propto \Gamma$. This is in accordance with the Elliott-Yafet theory. However above this temperature it deviates from this behavior, the linewidth has a maximum around 400 K, above which it decreases with increasing temperature. This highly anomalous temperature dependence could not be explained for a relatively long time. The ESR linewidth measures the inverse spin-spin relaxation time, $T_2$, through $T_2 = 1/\gamma \Delta B$. In 2008, Simon et al. (Ref. [33]) showed that the anomalous temperature dependence of $\Delta B$ could be explained by the so-called generalized Elliott-Yafet theory with a somewhat involved methodology (which was developed by Balázs Dóra), which is briefly discussed in the Chapter "Theoretical Backgound". It was shown that the quasiparticle broadening, $\Gamma$, becomes comparable to the band-band separation of boron $\sigma$ bands in the vicinity of the Fermi surface. The large $\Gamma$ is the effect of a large electron-phonon coupling, which is at the same time the cause of the superconducting transition at 40 K. The nearly degenerate boron $\sigma$ bands on the Fermi surface are due to the two boron basis atoms in the elementary cell.

It is in fact the purpose of the present section to show that the intuitive unification of the Elliott-Yafet and the D’yakonov-Perel’ theories allows to calculate the relaxation times in a transparent and elegant manner. In order to do so, we
need to justify the applicability of the present zero field calculations for a finite field study such as that in the ESR studies (Refs. [57, 33]). First, I note that the 0.3 T (approximately 0.4 K) magnetic field is the smallest parameter compared to the other energy scales (temperature and $\Gamma \approx 10 - 100$ meV). Second, little magnetic field dependence of the ESR linewidth was found in Refs. [59, 60] and it was experimentally verified that $T_1 = T_2$ in the temperature range where $T_1$ could be studied by direct means. This means that our zero field calculation of the spin-relaxation time is applicable to study the spin-spin relaxation time in ESR.

To calculate the spin-relaxation rate, the EY Hamiltonian of Eq. (3.38) with an isotropic SOC is considered. Here, isotropic means $SO(3)$ symmetry of the Hamiltonian. It is transformed to a DP-like Hamiltonian according to the procedure described above. The rest of the calculation proceeds according to the Monte Carlo simulation procedure of a DP problem, which is described in Section 3.2.2.

In this isotropic case, it is sufficient to specify the Hamiltonian for a single $k_0$ point on the spherical Fermi-surface. The matrix elements are obtained for an arbitrary point by transforming this Hamiltonian by means of rotations. For convenience I specify this given $k_0$ wave vector as the “north pole” on the Fermi-sphere at $k_0 = (0, 0, k_F)$. We are interested in the matrix elements of the Hamiltonian such as:

$$H_{\alpha,\sigma;\alpha',\sigma'}(k) = \langle k, \alpha, \sigma | H | k, \alpha', \sigma' \rangle,$$

where $\alpha$ and $\sigma$ are the band and spin indices, respectively.

The matrix elements between wave functions with different $k$ and $k'$ are 0. Treating the $\alpha, \sigma$ pair as a single index, the Hamiltonian can be represented as a $k$ dependent $4 \times 4$ matrix. For the case of a retained inversion symmetry, the most general form of this matrix reads:

$$H_{\alpha,\sigma;\alpha',\sigma'}(k) = \langle k, \alpha, \sigma | H | k, \alpha', \sigma' \rangle,$$

where $\alpha$ and $\sigma$ are the band and spin indices, respectively.

The matrix elements between wave functions with different $k$ and $k'$ are 0. Treating the $\alpha, \sigma$ pair as a single index, the Hamiltonian can be represented as a $k$ dependent $4 \times 4$ matrix. For the case of a retained inversion symmetry, the most general form of this matrix reads:

$$H = \begin{bmatrix}
1^\uparrow & 1^\downarrow & 2^\uparrow & 2^\downarrow \\
0 & 0 & L_{2,k} & L_{1,k} \\
0 & 0 & L_{1,k}^* & -L_{2,k} \\
L_{2,k} & L_{1,k} & \Delta_k & 0 \\
-L_{1,k} & -L_{2,k} & 0 & \Delta_k
\end{bmatrix}.$$

Rotations around the $z$ axis leave the $k_0$ vector invariant. The generator of rotations around the $z$ is $J_z = L_z + S_z$.

$$[H, J_z] = 0,$$

$$\langle k_0, \alpha, \sigma | [H, J_z] | k_0, \alpha', \sigma' \rangle = 0.$$

Since there are two non-degenerate kinetic bands, one can assume that the angular momentum is quenched. It yields:
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\begin{equation}
\langle k_0, \alpha, \sigma \mid [H, S_z] \mid k_0, \alpha', \sigma' \rangle = 0,
\end{equation}

\begin{equation}
[H, S_z]_{\alpha, \sigma; \alpha', \sigma'}(k_0) = \begin{bmatrix}
0 & 0 & 0 & -L_{1,k_0}^* \\
0 & 0 & L_{1,k_0} & 0 \\
0 & -L_{1,k_0} & 0 & 0 \\
L_{1,k_0}^* & 0 & 0 & 0
\end{bmatrix},
\end{equation}

\begin{equation}
L_{1,k_0} = 0.
\end{equation}

This means that for the $k_0$ point, the SOC matrix elements are described by a single real parameter: $L = L_{2,k_0}$.

A general rotation transformation is described by the following operation:

\begin{equation}
R = e^{-iJa\varphi/h} = e^{-iLa\varphi/h} e^{-iSa\varphi/h},
\end{equation}

where $a$ and $\varphi$ denote the axis and angle of the rotation, respectively. Clearly, the rotation operation acts separately on the kinetic and spin parts of the wave function, i.e.:

\begin{equation}
R \mid k, \alpha, \sigma \rangle = (e^{-iLa\varphi/h} \mid k, \alpha \rangle) \otimes (e^{-iSa\varphi/h} \mid \sigma \rangle).
\end{equation}

For any $k \neq k_0$, a single operation that rotates $k_0$ to $k$ can be defined.

\begin{equation}
\alpha_{k_0,k} = \frac{k_0 \times k}{|k_0 \times k|},
\end{equation}

\begin{equation}
\varphi_{k_0,k} = \sin^{-1}(|k_0 \times k|),
\end{equation}

\begin{equation}
R_{k_0,k} = e^{-iJa_{k_0,k} \varphi_{k_0,k} / h}.
\end{equation}

The spatial rotations are symmetries of the kinetic part of the Hamiltonian thus the rotation does not mix wave functions from different bands, i.e.:

\begin{equation}
|k, \alpha \rangle = R_{k_0,k} \mid k_0, \alpha \rangle.
\end{equation}

Our Hamiltonian is symmetric to these rotations, therefore:

\begin{equation}
[H, R_{k_0,k}] = 0,
\end{equation}

\begin{equation}
H = R_{k_0,k} HR_{k_0,k}^{-1}.
\end{equation}

The only non-zero matrix elements in the $k$ vector read:

\begin{equation}
\begin{aligned}
\langle k, \alpha, \sigma \mid H \mid k, \alpha', \sigma' \rangle &= \langle k, \alpha, \sigma \mid R_{k_0,k} HR_{k_0,k}^{-1} \mid k, \alpha', \sigma' \rangle \\
\langle k, \alpha, \sigma \mid H \mid k, \alpha', \sigma' \rangle &= \langle k_0, \alpha, \sigma \mid e^{-iSa_{k_0,k} \varphi_{k_0,k} / h} H e^{iSa_{k_0,k} \varphi_{k_0,k} / h} \mid k_0, \alpha', \sigma' \rangle, \\
H_{\alpha, \sigma; \alpha', \sigma'}(k) &= \sum_{\sigma_1, \sigma_2} [e^{-iSa_{k} / h}]_{\sigma, \sigma_1} H_{\alpha, \sigma_1; \alpha', \sigma_2}(k_0) [e^{iSa_{k} / h}]_{\sigma_2, \sigma'}.
\end{aligned}
\end{equation}
The spin rotation can be calculated using matrix exponential of $2 \times 2$ matrices. This allows us to arrive at the final SOC values at the $k = (k_x, k_y, k_z)$ point:

$$L_{1,k} = -\frac{Lk_x - iLk_y}{k_F},$$
$$L_{2,k} = \frac{Lk_z}{k_F}. \quad (3.60)$$

This remarkably simple and symmetric result allows the calculation of the SOC Hamiltonian for any $k$ points. This, together with the above transformation of the EY Hamiltonian to the DP problem and the corresponding Monte Carlo method described in the previous chapters, allows the calculation of spin-relaxation times for this isotropic system.

![Figure 3.15: Spin-relaxation rate, $\Gamma_s$, calculated for an isotropic SOC with varying momentum relaxation rate, $\Gamma$. Although the system under study follows the EY model, it was transformed to a DP like Hamiltonian and the spin relaxation was calculated with the Monte Carlo method. The schematics of the band structure is also shown and the EY and GEY regimes are indicated. Note that the crossover between the two regimes is rather smooth.](image)

**Fig. 3.15.** shows the universal curve which is obtained for the spin-relaxation of a metal with inversion symmetry and isotropic SOC. The curve is well fitted by

$$\Gamma_s(\Gamma) = \frac{2}{3} \left( \frac{L^2}{\Delta} \right) \frac{\Gamma/\Delta}{1 + (\Gamma/\Delta)^2}. \quad (3.61)$$
This could be rewritten as \( \frac{2}{3} \frac{\Gamma L^2}{\Delta^2 + \Gamma^2} \) but I kept the above form to better demonstrate the dependence on \( \Gamma/\Delta \).

The 2/3 prefactor is geometrical, it depends on how the SOC Hamiltonian is normalized. The original Elliott result allows for the presence of similar factors (usually denoted by \( \alpha \)).

It is important that the SOC is the smallest energy scale, I used herein \( L = 0.1\Delta \), however the same curve fits the whole \( L \ll \Delta \) regime well. This is exactly the generalized EY result of Ref. [14] which was first deduced in Ref. [33] and is obtained herein numerically.

I also highlight an interesting analogy between my numerical data and analytic calculations, which are available in the literature. The spin-relaxation in inversion breaking materials under the action of built-in SOC related fields and an additional magnetic field along the \( z \) axis was worked out analytically in Ref. [29] (Eq. IV. 36):

\[
\Gamma_s = \left( \Omega_x^2 + \Omega_y^2 \right) \frac{\tau_c}{\Omega_0^2 \tau_c^2 + 1},
\]

where \( \Omega_x \) and \( \Omega_y \) denote the Larmor (angular) frequencies along the \( x \) and \( y \) axes due to the SOC, respectively. In our notation they are identified as \( h\Omega_x = -2\text{Re}[L_k] \) and \( h\Omega_y = 2\text{Im}[L_k] \). \( \tau_c \) is the electron scattering correlation time, which is identified as the momentum scattering. \( \Omega_0 \) is the Larmor angular frequency due to the magnetic field along the \( z \) axis. In our model, \( \Delta_k \) plays a similar role as the Zeeman splitting, thus the assignment is \( h\Omega_0 = \Delta \).

Substituting these assignments into Eq. (3.62) yields:

\[
\Gamma_s = \frac{4L^2}{\Delta^2 + \Gamma^2},
\]

which is the same result as the fitted curve to the simulations (Eq. (3.61)), besides the prefactor.

The experimental and calculated spin-relaxation rates (\( \Gamma_s \)) for MgB\(_2\) are shown in Fig. 3.16. The experimental data are from Ref. [33] and is obtained after subtracting the spin-relaxation rate contribution from the boron \( \pi \) bands. It is described in Ref. [33] that the latter obeys the conventional EY mechanism as the boron \( \pi \) bands are separated from each other with a large gap (2 eV). In contrast, the separation of the boron \( \sigma \) bands is as low as \( \sim 0.2 \) eV. The final result is obtained by presenting the data as a function of temperature using the \( T(\Gamma) \) relation from the Bloch-Grüneisen function with a Debye temperature of 535 K and also by scaling the data with the already known \( \Delta = 194 \) meV (Ref. [33]) and \( L = 1.7 \) meV.

An excellent agreement between the experiment and the spin-relaxation rate, which is obtained from the Monte Carlo simulation, can be observed. This also
Figure 3.16: Experimental (symbols) and simulation based (solid curve) spin-relaxation rate, $\Gamma_s$, in MgB$_2$. Note that the experimental data contains the contribution from the boron $\sigma$ bands only; as described in Ref. [33], the contribution from the $\pi$ bands remains explicable by the conventional EY theory and it is thus subtracted. The level scheme and the identification of the EY and GEY regimes is also depicted. Note that the two regimes cross over smoothly as a function of temperature and the vertical dashed line is intended as a guide only.

means that the spin-relaxation rate in MgB$_2$ can be appropriately described by a single band-band separation value and also by an isotropic SOC model. I emphasize that with this demonstration, a spin-relaxation problem in an inversion symmetric, strongly correlated metal is traced back to the methodology which was developed for an inversion symmetry breaking material.
3.4 The Loschmidt echo and spin relaxation

3.4.1 Introduction and formulation of the problem

This section discusses an interesting proposal which enables the separation of dephasing and spin-relaxation processes with the introduction of the concept of Loschmidt echo for the problem of spin dynamics in semiconductors.

Figure 3.17: Decay of the magnetization, $S_z$ for the Dresselhaus SOC when $\Gamma = 0.01\mathcal{L}$. Note the presence of a non-exponential spin-decay for short time values, which are followed by a more exponential looking decay. The corresponding Fourier transform $\text{Re} S(\omega)$ is also shown. This figure was calculated using the methodology as described in Section 3.2.2 and closely resembles Fig. 3.12 but is calculated for differing parameters.

Fig. 3.17 shows a situation where the loss in the $S_z$ ensemble magnetization decays strongly non-exponentially when $\Gamma \ll \Delta\mathcal{L}$. Then, the magnetization decay occurs due to dephasing, i.e. that the electron spins precess around the built-in SOC magnetic fields. Given that the latter has a distribution for varying $k$ vector directions, the precession gives rise to a reduced spin ensemble magnetization. In the absence of momentum relaxation, $\Gamma = 0$, the magnetization would show wiggles around an average value even for long times.

The characteristic timescale of the dephasing, i.e. when most of the ensemble magnetization occurs, is $\frac{\hbar}{\tau_{\text{dephasing}}} \approx \Delta\mathcal{L}$. This is followed by a more exponential-like drop in the ensemble magnetization for longer times. This unusual dependence of the magnetization is also captured in the corresponding $\text{Re} S(\omega)$: the $\omega = 0$ Dirac delta function is broadened as it corresponds to the exponential decay for long
times. The apparent dephasing is due to the non-Lorentzian side lobes, centered around $L$, which is due to the built-in SOC distribution of the Dresselhaus SOC Hamiltonian. The presence of dephasing for such a situation was discussed in Ref. [3], however, the details have not been worked out.

The non-exponential magnetization decay poses a challenge to describe the system with a spin-relaxation time. There are possible candidates for this identification: it could be e.g. the width of the Lorentzian at $\Omega = 0$ or one could imagine a deconvolution of the dirty (i.e. finite $\Gamma$) $\text{Re}\mathcal{S}(\omega)$ by the clean (i.e. $\Gamma = 0$), which would yield the broadening function. However, it is not immediately clear physically why this method would allow the separation of the true spin-relaxation from dephasing. Although the spin-relaxation is quite probably exponential, a clearer physical method is required which allow distinction of the two types of processes. I believe that the essential difference between spin-relaxation and dephasing is that spin-relaxation is a truly memory-loss process (and it thus irreversible), whereas dephasing does not correspond to a loss of memory (and it thus reversible). The latter two statements is supported by the fact, that were the time reversible, in the absence of momentum scattering, the precession around the built-in SOC fields would be reversed. Therefore dephasing retains the system memory. In contrast, this memory is lost after a momentum scattering event thus even if the time were reversed, the precession could not be inverted.

3.4.2 The concept of Loschmidt echo

The concept of the Loschmidt echo (1876) stemmed from a famous Gedankenexperiment (also known as the Loschmidt paradox) by Joseph Loschmidt when he attempted to object the statistical interpretation of the second law of thermodynamics by Ludwig Boltzmann. Loschmidt argued that the time-reversal invariance of classical mechanics leads to evolution processes in which the entropy decreases. These processes could be reached by inverting the velocity vectors of all molecules of the system. This inversion appears to be violating the second law of thermodynamics. Using arguments, which are beyond the scope of this thesis, Boltzmann successfully defended the statistical interpretation and showed that the "arrow of time" is indeed a valid concept for macroscopic systems. However, the Loschmidt proposal can be adapted to quantum systems with a limited number of degrees of freedom and it turns out that Loschmidt echo experiments can be performed. The best known practical realization of a Loschmidt echo scheme is the so-called spin-echo (or Hahn echo) discovered in 1950 by Erwin Hahn [61], which is discussed below.

I also give a brief introduction into the Loschmidt echo principle. The Loschmidt echo studies the amount of revival which occurs when an imperfect time-reversal procedure is enforced to a physical system. It considers an initial quantum state $|\psi_0\rangle$
which evolves during a time $t$ under a Hamiltonian $H_1$ to a state an reaching the state $|\psi(t)\rangle$. The recovery of the initial state $|\psi_0\rangle$ is attempted by the application of another Hamiltonian $-H_2$, which is applied between $t$ and $2t$. A perfect recovery of $|\psi_0\rangle$ could be attained by taking $H_2$ to be equal to $H_1$. However, it is usually impossible to achieve in realistic cases as there is always a difference between $H_2$ and $H_1$, which leads to a non-perfect recovery of the initial state. The evolution of the system between $t$ and $2t$ under the Hamiltonian $-H_2$ is equivalent to a time-reversed evolution from $t$ to $0$ under $H_2$, which clarifies why the Loschmidt echo is regarded as if it was studying the case of time-reversal.

### 3.4.3 Loschmidt echo in magnetic resonance

In fact, the dephasing problem is encountered in magnetic resonance and it is also tackled with a modified version of the Loschmidt echo. It is a common challenge in magnetic resonance that dephasing and spin relaxation processes are simultaneously present. The so-called method of spin-echo (which is technically a special case of a Loschmidt echo) is employed to tackle this problem. In order to explain the analogy and the solution that it provides, I briefly summarize the fundamentals of dephasing and spin relaxation as seen in magnetic resonance.

Most generally, one encounters three different time scales in magnetic resonance: $T_2^*$, $T_2$, and $T_1$. Of these, $T_{1,2}$ are irreversible relaxation processes and $T_2^*$ is related to the reversible dephasing processes [19, 20]. The distinction between $T_1$ and $T_2$ stems from the fact that a magnetic field is applied in this method, which inevitably leads to a distinction between relaxation processes which are parallel (the $T_1$ processes, also known as longitudinal relaxation time) and perpendicular (the $T_2$ processes, also known as transversal relaxation time). In zero magnetic field, this distinction vanishes.

I previously introduced the Bloch equations which describe the motion of spins in a DC magnetic field along the $z$ axis, which is accompanied by an AC magnetic field whose polarization rotates around $z$. In equilibrium, the magnetization of the spin ensemble, $\mathbf{M}$ is stationary along the $z$ axis with a value of $M_0$. When the AC magnetic field is applied in a pulsed manner, the magnetization is rotated away from the $z$ axis and starts to precess around $z$ with the Larmor frequency $\omega_L = \gamma B$ ($B$ is the magnetic field and $\gamma$ is the so-called gyromagnetic ratio of the studied spin system, e.g. $\gamma \approx 2\pi42.6\text{MHz/T}$ for protons and $\gamma \approx 2\pi28.0\text{GHz/T}$ for electrons).

In a typical experiment, an AC irradiation is applied with an angular-frequency matching $\omega_L$ and a pulse duration which is sufficient to rotate $\mathbf{M}$ into the $(x,y)$ plane. This is known as a $\pi/2$ pulse, as the magnetization is rotated perpendicular to $z$. Then the $(x,y)$ and $z$ components of the non-equilibrium spin magnetization decay to the respective equilibrium values (0 and $M_0$) with $T_2$ and $T_1$ relaxation
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times. However, in most cases the (x, y) components vanish much earlier than $T_2$ due to dephasing: local magnetic field inhomogeneities are present which lead to a distribution in $\omega_L$. The inhomogeneities are caused by either defects or impurities (these are the leading cause in solid state NMR) or by the inevitable inhomogeneity of the magnet (this is the leading cause in high resolution or liquid NMR) [19].

This process is usually described in a frame of reference which rotates with the mean value of the Larmor (angular frequency), $\overline{\omega_L}$ around the z axis and is schematically shown in Fig. 3.18. The coordinate axes of the rotating frame of reference are denoted by $x'$, $y'$, and $z'$ ($z'$ is identical to the z axis). The originally $M_0$ magnetization lies in the $(x', y')$ plane after a $\pi/2$ pulse where dephasing starts.

In the rotating frame of reference, some spins have angular frequencies which are larger (the blue arrows in the figure) or smaller (the red arrows in the figure) than $\omega_L$. The resulting net magnetization vanishes on a timescale of $T_2^* \approx 1/\Delta \omega_L$, where the latter is the spread in the Larmor frequencies. Another pulse is applied after a so-called "evolution time", $\tau_{\text{echo}}$, which rotates the spins by $\pi$. The figure depicts the location of the spins which precess faster (blue arrows) and slower (red arrows) than the average after the $\pi$ pulse. Clearly, after a waiting time of another $\tau_{\text{echo}}$ the spins are aligned again coherently in the $x', y'$ plane with a direction opposite to their original coherent direction.

Fig. 3.18. also shows the corresponding NMR signal: the initially decaying signal is partially recovered, i.e. an echo is observed at $2\tau_{\text{echo}}$ when the $\pi$ pulse is applied at $\tau_{\text{echo}}$. The reason why the NMR spin echo is observed, is that the dephasing is not accompanied by a memory loss, thus each spin "remembers" the magnitude of its Larmor frequency. However in reality, memory loss is also present on the spin-relaxation timescale, $T_2$, where typically $T_2^* \gg T_2$. In NMR, the physical origin of $T_2$ can be dipole-dipole interaction (this is the leading mechanism in solid state NMR) or molecular diffusion (this is the leading mechanism in high resolution or liquid NMR) [19].

Fig. 3.19. shows the schematics of the $T_2$ measurement: spin-echo experiments are performed consecutively (in different pulse sequence runs, each starting from the equilibrium $M_0||z'$) with varying $\tau_{\text{echo}}$. The envelope of the observed echoes follow $\exp^{-\tau_{\text{echo}}/T_2}$, which allow for the determination of $T_2$, which is a true, irreversible relaxation time, clearly distinguishable from dephasing. After Fourier transformation, the NMR signal has a large linewidth of $1/T_2^*$ (in frequency units) which consists of so-called spin-packets[21], whose linewidth is $1/T_2$.

3.4.4 Loschmidt echo in the Monte Carlo simulations

Our proposal to separate the dephasing and relaxation processes in the above described problem of spin relaxation, essentially mimics the magnetic resonance approach. In this approach, the Loschmidt echo can be induced by i) letting the
Rotating frame of reference

Figure 3.18: Schematics of the dephasing process in NMR experiments and the method of spin-echo. The figure assumes a right handed precession direction with $\omega_L$. The spin magnetization lies in the $(x', y')$ plane after a $\pi/2$ pulse when dephasing due to a spread in the Larmor frequencies starts: spins in blue and red precess faster or slower than $\omega_L$, respectively. After an evolution time of $\tau_{\text{echo}}$, a $\pi$ pulse is applied which rotates the spins around an axis perpendicular to $z'$. Clearly, the blue and red spins are now behind or before the average spin direction and as a result these will be aligned coherently after another $\tau_{\text{echo}}$ time, when the spin echo occurs. The lower panel depicts the corresponding NMR signal.
Figure 3.19: Schematics of the $T_2$ measurement. NMR spin echo experiments are performed with varying time delay between the $\pi/2$ and $\pi$ pulses. The individual echoes have a linewidth of $2T^*_2$ but the resulting spin echo envelope follows $e^{-t/T_2}$. The corresponding FT NMR signal reflects this behavior: it contains a broad signal whose width is $1/T_2^*$ and it consists of individual spin-packets whose width is $1/T_2$. 
3.4. THE LOSCHMIDT ECHO

Figure 3.20: Example of Loschmidt echo simulations with the method explained in the text. \( t_{\text{flip}} \) indicates the time point of SOC reversal. All simulations were run with \( \Gamma = 0.01\mathcal{L} \). The Fourier transformed spin decay is also shown (lower panel). Note that the sidelobes, which are present in the original dephasing problem (black curve), are absent for the echo envelope (red curve).
system evolve under the simultaneous action of dephasing and spin relaxation for a given time, $t$, ii) inverting the built-in SOC fields: $\Omega(k) \rightarrow -\Omega(k)$ and iii) detecting the amplitude of Loschmidt echo maximum at $2t$. The spin-relaxation time can be determined from the envelope of the consecutive Loschmidt echoes. The result is shown in Fig. 3.20. Technical details of a highly optimized algorithm are given in A.2. Both the original spin decay and the echo envelope can be Fourier transformed and the result is shown in Fig. 3.20. The Fourier transformed spin decay signal contains the sidelobes beside a Lorentzian centered at $\omega = 0$. The central Lorentzian describes the spin relaxation for longer times, whereas the sidelobes describe the rapid dephasing. Interestingly, the sidelobes are missing for the Fourier transformed Loschmidt echo envelope and only a central line, which carries all spectral weight, is preserved. This is a clear indication that the Loschmidt echo concept can successfully separate spin relaxation from dephasing.

I highlight a conceptual difference between the NMR spin echo and our numerical approach: in NMR, the spins lie in a plane and the spin direction was reversed by mirroring, which yielded essentially that the consecutive action of the same Hamiltonian let the spins evolve toward the echo. Our approach is more similar to the original concept of the Loschmidt echo, i.e. it is the SOC Hamiltonian which is reversed rather than the spin direction.

I finally note that this reversal of the internal SOC field could be realized experimentally in a system which is dominated by a Bychkov-Rashba type SOC due to an external electric field. This is well approximated in graphene, where the dominance of the Bychkov-Rashba type SOC over the intrinsic contribution was predicted [62]. For such a system, it is expected that the electric field and the resulting SOC fields have a distribution in the device. Then, a sudden reversal of the external electric field would lead to a Loschmidt echo. Certainly, this scenario could be challenged by several experimental factors including e.g. finite electric field switching field.
Chapter 4

Summary

This thesis focused on the description of spin relaxation in metals and semiconductors. Prior to the thesis there had been several open questions including how well the Elliott-Yafet theory can be validated in metals experimentally, whether the complete phase diagram of spin-relaxation can be constructed in zincblende semiconductors, how one can separate the effects of spin dephasing from spin-relaxation, and whether the two major theories of spin-relaxation, Elliott-Yafet and D’yakonov-Perel’, could be unified and brought to a common formalism. I managed to find the answers to these open questions and namely, I presented the accurate method for the experimental verification of the Elliott-Yafet theory in elemental metals. I developed a novel, Monte Carlo simulation based approach, which allows to study spin-relaxation in semiconductors without inversion symmetry with an arbitrary distribution of the spin-orbit coupling and for arbitrary values of the momentum relaxation. I also presented an intuitive way to unify the above two relaxation theories, which also allowed for the application of the Monte Carlo approach in materials with inversion symmetry, i.e. on a range of materials for which the original method was not thought to be applicable. A method, based on the concept of the Loschmidt echo was presented, which allows the separation of the spin dephasing from spin relaxation.
Chapter 5

Thesis points

1. I pointed out two shortcomings of the phenomenological Beuneu-Monod relation, which was developed to explain the spin-relaxation time in elemental metals by correlating the experimental electron spin resonance line-width with the so-called spin-orbit admixture coefficients and the momentum-relaxation theory. Namely that i) the momentum-relaxation involves the Debye temperature and the electron-phonon coupling whose variation among the elemental metals was neglected, ii) the Elliott-Yafet theory involves matrix elements of the spin-orbit coupling (SOC), which are however not identical to the SOC induced energy splitting of the atomic levels, even though the two have similar magnitudes. I obtained refined values for the empirical spin-orbit admixture parameters for the alkali metals by considering the proper description of the momentum relaxation theory. [T1]

2. I developed a stochastic model for the calculation of the dynamic spin-susceptibility for materials without inversion symmetry for an arbitrary distribution of the spin-orbit coupling and for an arbitrary value of the quasi-particle scattering. The calculation yields numerically the spin-relaxation time. I validated the model by comparing its predictions to analytic calculations in the clean (no momentum scattering) and dirty limits (large momentum scattering) [T2].

3. Using the stochastic model, I studied the full phase space of spin relaxation as a function of SOC strength, its distribution, and the magnitude of the momentum relaxation rate. This allowed the identification of two novel spin-relaxation regimes; where spin relaxation is strongly non-exponential and when the spin relaxation equals the momentum relaxation. I also found a compelling analogy between the spin-relaxation theory and the NMR motional narrowing. Using the stochastic model, I calculated the dynamic spin-susceptibility for a variety of SOC distributions [T2].
CHAPTER 5. THESIS POINTS

4. I developed an intuitive model which allowed the unification of the Elliott-Yafet and the D’yakonov-Perel’ spin-relaxation mechanisms. I showed that the respective Hamiltonians of the two theories can be transformed to each other. I showed that the so-called generalized Elliott-Yafet theory, which was developed for the case of large momentum scattering, can be straightforwardly obtained using the D’yakonov-Perel’ approach [T3].

5. I showed that the intuitive unification of the EY and DP theories not only provides an insight to the intimate relationship between the two theories but allows to numerically obtain spin-relaxation times using the stochastic approach, which was developed for the DP case. I presented that this can be successfully applied to calculate the experimentally determined spin-relaxation time in MgB$_2$, which shows a significant momentum scattering rate, it thus cannot be handled with the conventional Elliott-Yafet model [T3].

6. I developed an intuitive numerical tool which allows the separation of dephasing and spin-relaxation processes. The method essentially mimics the concept of the Loschmidt echo, i.e. it introduces a time reversal for the built-in magnetic fields and keeps track of the resulting ensemble magnetization decay. I showed that the envelope of the Loschmidt echoes recovers the true spin-relaxation processes, which are otherwise unobservable due to strong dephasing [T4].

The publications related to the thesis points are as follows:


Other published works which are related to the thesis but are not included in the thesis points:


Bibliography


Appendix A

Appendix

A.1 The user interface of the simulation software

The source code of the simulation software was published as the electronic Supplementary Materials for Ref. [9] and is also available at Github\(^1\). There is also a more recent, cleaned up but incomplete software\(^2\) that also includes an advanced spin-echo measurement implementation. Its user interface differs from the first implementation.

The software was developed on Debian\(^3\), but it should be possible to compile on any POSIX compliant operating system (on Windows it should compile under Cygwin or WSL). The software depends on the Armadillo\(^4\) and Boost\(^5\) external libraries.

The software has a command line interface which allows scripting large number of simulations with varying simulation parameters. The \texttt{--help} option produces all the other available command line options.

\begin{verbatim}
$ bin/main --help
Allowed options:
  -h [ --help ]        produce help message
  -v [ --version ]     print version number
  --autocorr                        set autocorr measurement
  --spins arg (=10)     set number of spins
  --duration arg (=300) set simulation duration
  --timestep arg (=1)   set timestep
  --omega arg (=0.20000000000000001) set the absolute value of Larmor
\end{verbatim}

\(^1\)https://github.com/leni536/DP_random_walk
\(^2\)https://github.com/leni536/DP_random_walk2
\(^3\)https://www.debian.org/
\(^4\)http://arma.sourceforge.net/
\(^5\)https://www.boost.org/
--delta_omega arg (=0)       precession  set the width of omega distribution
--seed arg (=rand)           set the seed for the random generator
-o [ --output ] arg (=)     output file path
-m [ --model ] arg (=naiv)   name of the model
--meas arg (=prep)           name of measurement method
-b [ --B_meas ] arg (=0)     measurement field
--tmin arg (=0)              starting time, B_meas turns in at t=0

Since all simulation parameters have a default value, when run without arguments, the program outputs the result of a sample simulation to the standard output. The output contains a header which describes all simulation parameters so the simulation can be reproduced. After the header section it outputs the spin component of interest in the function of time. The time interval and sampling interval is the same as specified in the command line.

$ bin/main | head -20
# Djakonov-Perel simulation
# t=0 Sz=1, no magnetic field
# version: commit_17cc1d6e5275608347fa311f7f7e64735470c311
# spins: 10
# duration: 300
# timestep: 1
# omega: 0.2
# seed: 2494990189
# model: naiv
# meas: prep
# B_meas: 0
# tmin: 0
# autocorr: false
# t, Sz
0, 1
1, 0.990826
2, 0.97055
3, 0.946542
4, 0.925218
5, 0.910056
A.2 Implementation details

A.2.1 Spin ensemble simulation

The core logic of the simulation is written in `src/Measurement.cpp`.

The initial condition of a single electron is taken according to a `InitialCondition::Base` base class. There are two initial conditions implemented: i) `InitialCondition::Polarized3D` for polarized spin but uniform distribution of the wave vector on a spherical Fermi surface. ii) `InitialCondition::Isotropic3D` for uniform distribution of the spin on the Bloch-sphere as well. The latter is useful for auto-correlation measurements.

The simulation for a single electron is event based, the times of scattering events are modeled as a Poisson-process. In a Poisson-process, the time duration between subsequent events are independent and follow the exponential distribution. The $\lambda$ parameter of both the process and the distribution is determined by the momentum-scattering time: $\lambda = 1/\tau_m$. At each scattering event, the electron scatters to a new wave vector $\mathbf{k}$ on the Fermi surface. Both the distribution of durations between scatterings and transition probabilities are encapsulated in the `ScatteringModel::Base` base class.

The time-evolution of a single spin is determined by both `SOCModel::Base` and `MagneticField::Base`. The former describes the bulk single-spin Hamiltonian within the kinetic band (therefore $\Omega(k)$) and the latter describes the external, time-dependent magnetic field.

After the simulation for a single electron completes, the spin vector is sampled at uniform time intervals (Fig. A.1. This is necessary for averaging separate single electron simulations at the same time points.

At the very end, the ensemble averaged data $s(t)$ is written out to a CSV file. Further analysis of the data was done mainly in Mathematica and Python.

A.2.2 Loschmidt-echo simulation

The simulation component above allows for Loschmidt-echo simulations by exploiting the MagneticField class. Since the time dependent external magnetic field can change in between scattering events, the MagneticField class has the authority to calculate time evolution between the events getting the SOC model as a parameter. (Perhaps MagneticField is a misleading name for this class and TimeEvolution would indicate its purpose better.) This allows one to create a time evolution where to a certain time point the spins are advanced with the original SOCModel, but after this time point, they are advanced by the inverse of the original Larmor precession. Example runs of such simulations are shown in Fig. A.2.

---

6 [github.com/leni536/DP_random_walk2/blob/6499581.../src/Measurement.cpp#L64](https://github.com/leni536/DP_random_walk2/blob/6499581.../src/Measurement.cpp#L64)
Figure A.1: Demonstration that the spin components are sampled at uniform time intervals after a single electron simulation.

Figure A.2: Several Loschmidt echo simulations and the envelope of the echo responses.

However, when we are interested in the echo amplitude of each run only, then we can significantly improve the code. Instead of advancing an individual spin, one can advance the time evolution operator itself. By advancing the time evolution operator of both the original SOC model and the inverse (where we invert the Larmor precession), the time evolution for a specific echo simulation can be welded
A.3. GEOMETRIC FACTORS IN $\text{ReS}_{\text{clean}}(\omega)$

I give an analytic description of the time evolution of the magnetization of an electron ensemble subjected to internal SOC fields to verify further the Monte Carlo method applied in the thesis since this description is equivalent to the numerical method in the clean limit. Consider the SOC in the form $H_0 = \frac{i}{2} \Omega(k) \cdot \sigma$, where $\sigma$ is a vector composed by the Pauli matrices, and $\Omega(k)$ is the $k$-dependent internal SOC field.

The time evolution of the state of an electron under the SOC is determined by the time evolution operator $U(t) = \exp(-iH_0t/\hbar)$. Supposing that the electron is

together from these time evolution operators (Fig. A.3). The envelope on Fig. A.2 was calculated using this method.

Figure A.3: The two spin evolution operators are advanced at the same time. When a spin-echo simulation is needed with a specific $t_{\text{echo}}$, the calculated operators are welded together.
initially in spin-up state, i.e. its spin is polarized along the $z$ direction, 

$$|\psi(0)\rangle = |\uparrow\rangle = \frac{v_2(-)|+\rangle - v_2(+)\rangle}{v_2(-)v_1^+(+) + v_2(+)v_1(-)}, \quad (A.1)$$

its state ket at time $t$ is obtained as

$$|\psi(t)\rangle = \frac{v_2(-)e^{-iE_+t/\hbar}|+\rangle - v_2(+)e^{-iE_-t/\hbar}|\rangle}{v_2(-)v_1^+(+) + v_2(+)v_1(-)}, \quad (A.2)$$

by applying the time evolution operator. Here, $|\pm\rangle = [v_1^{(\pm)}, v_2^{(\pm)}]$ and $E_{\pm}$ are the eigenkets and eigenenergies of the Hamiltonian $H_0$. Finally, the time development of the $z$ component of the electron spin is obtained as

$$S_z(t, \mathbf{k}) = \langle \psi(t)|\hat{S}_z|\psi(t)\rangle = \frac{\Omega_x(k)^2 + \Omega_y(k)^2}{\Omega(k)^2} \cos(\Omega(k)t) + \frac{\Omega_z(k)^2}{\Omega(k)^2}, \quad (A.3)$$

where $\Omega = \sqrt{\Omega_x^2 + \Omega_y^2 + \Omega_z^2}$. The quantity $S_z(t)$, i.e. the $z$ component of a spin ensemble, calculated by Monte Carlo method is obtained as

$$S_z(t) = \int_{\text{FS}} d\mathbf{k}S_z(t, \mathbf{k}) \quad (A.4)$$

within this approach, i.e. by integration over $\mathbf{k}$ on the Fermi surface. Arbitrary $\Omega(k)$ can be considered in the Hamiltonian $H_0$ such as the two-dimensional Bychkov-Rashba SOC or the three-dimensional Dresselhaus case discussed in the thesis. Note that for a complicated distribution of the SOC fields, the $\mathbf{k}$ integration in Eq. (A.4) might not be performed analytically.

When this calculation is performed according to Eq. (A.3) for different model Hamiltonians such as those given in the thesis, numerically identical results were obtained (data not shown) as for the Monte Carlo, and the analytic result for the dynamic spin susceptibility given in Eq. (3.23) is reproduced as well.

From Eq. (A.3) it is obvious that in a two-dimensional case, i.e. when $\Omega_z = 0$: 

$$S_z(t, \mathbf{k}) = \cos(\Omega(k)t). \quad (A.5)$$

By taking the Bychkov-Rashba SOC, $\Omega(k) = \frac{\epsilon}{\hbar k_F}[k_x, k_y, 0]$, $\Omega(k)$ becomes $\mathbf{k}$ independent as $\Omega(k) = \mathcal{L}/\hbar = \Omega$, which means a single oscillating component in $S_z(t)$ as it is shown by dashed curve in Fig. 3.8 in the thesis, and two Dirac-delta peaks at $\pm\Omega$ in the real part of the Fourier transform $S(\omega)$. In three-dimensional cases,
Figure A.4: Schematic depiction of the spin precession around the SOC fields and the corresponding $\Omega(k)$ vectors when the spins start along the $z$ direction at $t = 0$. Note that for an arbitrary $\Omega(k)$ which is not in the $x-y$ plane, the precession retains a finite positive $s_z$ value.

there is always a $t$-independent non-zero term in $S_z(t)$ coming from the last term in Eq. (A.3), which explains the finite $S_z$ value in Fig. 3.12 with $\Gamma = 0$ in the thesis. This non-zero and time independent term corresponds to a Dirac-delta function centered on $\omega = 0$ in $S(\omega)$.

The origin of this effect is further supported by a geometric consideration which is depicted in Fig. A.4. The presence of the Dirac-delta peak for $S(\omega = 0)$ is a generic feature and its absence for the two-dimensional electron gas and the Bychkov-Rashba SOC is an exception. For the latter, when the spins are aligned perpendicular to the 2D plane, all SOC fields are in the plane, i.e. the precession of the spins around the built in $\Omega(k)$ results in a zero-averaged net magnetization. However, for a general distribution of the SOC fields and the corresponding $\Omega(k)$ vectors, the precession of the spins retains a finite positive $s_z$ component as Fig. A.4
depicts. A straightforward geometric consideration shows that the \( \langle s_z \rangle \), i.e. the Dirac delta function strength is given by \( \Omega_z^2/\Omega^2 \) (\( \Omega_z \) and \( \Omega \) are the \( z \) component and the magnitude of the \( \Omega(k) \) vector, respectively) for a particular \( \Omega(k) \) component. Similarly, the amplitude of the oscillation goes as \( 1 - \Omega_z^2/\Omega^2 \), in full agreement with Eq. (A.3).

![Figure A.5: Comparison of the Monte Carlo based \( \text{Re} S(\omega) \) (solid line) and the histogram of the \( |\Omega(k)| \) (open symbols). Note that there are no scaling parameters between the two kinds of data.](image)

In Fig. A.5 I present the comparison between the \( \text{Re} S(\omega) \) and the histogram of the internal Larmor frequency distribution for the three-dimensional Dresselhaus model. The latter data integrated for the positive frequencies gives the average of the SOC fields which coincides with the frequency value where it is peaked and its weighted integral, i.e. the integral of the histogram values multiplied by the related frequency gives unity since all weights are summed up in this way.

### A.4 Spin relaxation for multiple types of SOC

In this section I investigate the spin-susceptibility inside a bulk zincblende conductor in the presence of electric field. I assume Dresselhous and Bychkov-Rashba spin-
orbit coupling for the bulk intrinsic SOC and the electric field induced SOC respectively.

When the electric field in the zincblende structure is along the [0, 0, 1], both the Dresselhaus and the Bychkov-Rashba Hamiltonian has the usual form which is mentioned in the thesis and the resulting Larmor-frequency distribution reads:

\[
\Omega(k) = \frac{L_D}{\hbar k_F^3} \begin{bmatrix} k_x (k_y^2 - k_z^2) \\ k_y (k_x^2 - k_z^2) \\ k_z (k_x^2 - k_y^2) \end{bmatrix} + \frac{L_R}{\hbar k_F} \begin{bmatrix} -k_y \\ k_x \\ 0 \end{bmatrix},
\] (A.6)

When the electric field direction is along the [1, 1, 1], the z-axis along this direction is taken, therefore the Bychkov-Rashba Hamiltonian is retained and the Dresselhaus one is transformed. The resulting Larmor-frequency distribution reads:

\[
\Omega(k) = \frac{L_D}{2\sqrt{3}\hbar k_F^3} \begin{bmatrix} -k_y (k_x^2 + k_y^2) - (k_y^2 - 2k_x k_y - k_z^2) k_z + 4k_y k_x k_z \\ k_x (k_z^2 + k_y^2) + (k_z^2 - 2k_x k_y - k_y^2) k_z - 4k_x k_y^2 \\ (k_x - k_y) (k_x^2 + 4k_x k_y + k_y^2) \end{bmatrix} + \frac{L_R}{\hbar k_F} \begin{bmatrix} -k_y \\ k_x \\ 0 \end{bmatrix}.
\] (A.7)

The direction in the zincblende structure are illustrated in Fig. A.6.

We set \(L_D = L_R = L\) in our examples but the simulation could be performed for arbitrary values of the SOC.

In Fig. A.7, the components of Re\(S(\omega)\) for the various SOC configurations and for different values of \(\Gamma\) are shown. The first column is for the pure Dresselhaus SOC and is an identical result to that given above. \(\Gamma\) values in the rows are 0, 0.02\(L\) and 5\(L\) respectively. The scales are the same for the first two rows of data but are different for the third one (with large \(\Gamma\)).

<table>
<thead>
<tr>
<th>SOC model and direction</th>
<th>(\Gamma_s) in units of (\mathcal{L})</th>
</tr>
</thead>
<tbody>
<tr>
<td>pure Dresselhaus</td>
<td>0.017</td>
</tr>
<tr>
<td>Dresselhaus+Rashba [0, 0, 1] (xx) direction</td>
<td>0.091</td>
</tr>
<tr>
<td>Dresselhaus+Rashba [0, 0, 1] (zz) direction</td>
<td>0.161</td>
</tr>
<tr>
<td>Dresselhaus+Rashba [1, 1, 1] (xx) direction</td>
<td>0.088</td>
</tr>
<tr>
<td>Dresselhaus+Rashba [1, 1, 1] (zz) direction</td>
<td>0.161</td>
</tr>
</tbody>
</table>

Table A.1: The spin-relaxation broadening parameter, \(\Gamma_s\), in the D’yakonov-Perel’ regime for the various SOC combinations in Fig. A.7 for \(\Gamma = 4\mathcal{L}\).

The important observations are:
Figure A.6: [0, 0, 1] and [1, 1, 1] directions in the zincblende crystal structure.

1. the overall SOC field becomes larger as expected with a singular (step-like) feature (indicated by an arrow in the figure),

2. the spin-relaxation becomes anisotropic when the Bychkov-Rashba term is also present and even an $xy$ term is present for the [0, 0, 1] electric field direction, which however vanishes in the D'yakonov-Perel' regime,

3. the anisotropy remains for the [1, 1, 1] electric field direction but the $xy$ term is zero,

4. a single Lorentzian component is observed in the D'yakonov-Perel' regime for all cases, but with varying widths which are summarized in Table A.1. The broadening parameter, $\Gamma_a$ is the HWFM of the Lorentzian curves in the figures.
A.4. **SPIN RELAXATION FOR MULTIPLE TYPES OF SOC**

Figure A.7: The components of $\text{Re}\mathcal{S}(\omega)$ for the various SOC configurations and for different values of $\Gamma$. $\Gamma$ from top to bottom is 0, 0.02$\mathcal{L}$ and $5\mathcal{L}$. Note that the $\omega$ axes are scaled differently for the last two columns of figures.