Optical Studies of Topological Semimetals in Magnetic Fields

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This thesis covers theoretical and optical studies on a selection of topological materials, namely the compounds TaP, NbAs, and GdPtBi. Topology in condensed matter physics is a recent and rapidly developing field and addresses materials that are characterized by the fact that their surfaces have have physical properties that are qualitatively distinct from their bulk. Topological insulators, for example, allow no or little electrical conduction through the bulk, impeded by a band gap. On the boundary of the material, however, we find gapless topological surface states providing conduction channels. Ideally, the charge carriers hosted by these states are spin polarized, massless and resilient against backscattering from non-magnetic impurities, making the surface currents dissipationless. Further, topological phases, which can be either inherent to the material or caused by external tuning parameters like a magnetic field, are characterized by physical observables taking quantized values proportional to topological invariants, for example, the discrete steps of electrical resistance measured in the quantum Hall effect. Interestingly, even though these topological invariants provide us with information about the physics on the boundary of a material, they are derived from electron wavefunctions that inhabit the material bulk. We call this connection the bulk-boundary correspondence. Importantly, these topological invariants do not require to be delicately tuned and can be robust against finite changes of external tuning parameters such as pressure, temperature or lattice imperfections, before the material is forced through a topological transition into the trivial regime. These properties raise interest in topological materials for potential computing applications, such as race-track memories powered by skyrmions, or topological Majorana qubits for quantum computing, which can be realized at interfaces with superconduc-
This thesis focuses on a subset of topological phases called the *Weyl semimetals*. These are hosts of bulk electronic bands, which form conical intersections, namely the Weyl cones. The associated charge carriers, the Weyl fermions, are chiral, meaning that they are massless and spin polarized. Weyl cones always come in pairs of opposite chiralities and are interconnected by a unique type of surface state called a *Fermi arc*, whose detection identified the first successfully realized Weyl semimetal, TaAs, in 2015. Weyl semimetals host a variety of interesting bulk properties, chiefly among them the *chiral anomaly*, an anomalous current between Weyl cones of opposite chirality. Weyl fermions and the associated chiral anomaly originally started as propositions in particle physics. The discovery of Weyl semimetals, bears not only great potential for physical applications, but has also brought a flurry of fundamental research connecting condensed-matter and particle physics.

In the scope of this thesis, the Weyl semimetals TaP, NbAs, and the potentially field-tunable variant GdPtBi are investigated using infrared-spectroscopy. The aim is to detect massless Weyl fermions by exploiting a property of linearly dispersing bands: when subjected to magnetic fields their Landau-level energies are proportional to $\sqrt{B}$. This makes them easily distinguishable from the linear-in-B Landau levels of massive electrons, which are described by parabolically dispersing bands. Though, as we will find, this low-energy approximation often fails to grasp the essential character of real materials. We will evaluate magneto-optical experiments performed in fields up to 33 T in close combination with detailed band structure analyses for TaP, NbAs and GdPtBi. The thesis is organized as follows:

- In Chapter 1, the introduction briefly outlines the connection between topological phases and particle physics, with a focus on the Weyl fermions and the chiral anomaly.
In Chapter 2, we introduce the matter of topology first on geometric examples and work towards the topological description of Weyl cones in $k$-space using Berry’s quantities.

In Chapter 3, we derive the Landau quantization of band electrons commonly encountered in low-energy approximations of electronic band structures. Additionally, the chapter provides a short description of the chiral anomaly of Weyl semimetals in magnetic fields.

In Chapter 4, we provide a derivation for the relevant Kubo formula of the optical conductivity. The derived equation can be used to evaluate the reflectivity spectra in the forthcoming experimental chapters.

In Chapter 5 and 6, the experimental setup is described and the details of the experiments and materials are outlined.

In Chapter 7, the Landau level spectrum of the Weyl semimetal TaP is discussed. Most prominently and unexpectedly it features traces of the band inversion, which is an essential ingredient for the presence of Weyl cones. Experimental results and preliminary analysis have been presented earlier in the author’s Master thesis [1].

In Chapter 8, the Landau level spectrum of the Weyl semimetal NbAs is discussed. Unlike in TaP, the band inversion and spin-orbit coupling are much weaker in this compound. Nonetheless, we show that the bulk bands, which mostly just touch, have a hyperbolic character. To describe the Landau levels of touching hyperbolic bands an elaborate low-energy description is used.

In Chapter 9, the magneto-optical spectrum of the magnetic triple-point semimetal GdPtBi is discussed. Its triple points, intersections of a singly and a doubly degenerate band, are ex-
pected to split and give rise to Weyl cones in external magnetic fields. Rather than a Landau level spectrum of Weyl fermions, broad features are observed that we associate directly to the induced splitting by an effective Zeeman term, which arises from the paramagnetic alignment of free $f$-electron spins in the Gadolinium atoms. Some of the experimental results on this compound have been reported earlier in the author’s Master thesis [1].

- In Chapter 10, we summarize and conclude on the experimental part of the thesis.


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Part I

Theory
This thesis covers magneto-optical experiments on several Weyl semimetals. These materials host electronic bands with linear dispersions, which intersect in a point in the Brillouin zone, forming the so-called Weyl cones. Similarly to the Dirac cones in graphene, Weyl cones describe massless Bloch electrons. Unlike the Dirac electrons, however, Weyl electrons are additionally spin-polarized. As we will explore presently, these special types of charge carriers combine concepts of the two seemingly unrelated subjects of topological condensed-matter physics and particle physics. This section provides a brief timeline of these connections and motivates the recent interest in these materials. Two developments are of special importance in this regard: the discovery of the quantum Hall effect (QHE) and the chiral anomaly.

To begin with, the QHE was the first experiment connecting topology and condensed-matter physics. It was found in 1980 while Klaus von Klitzing measured the Hall current of quasi-two-dimensional conductors subjected to a large magnetic field[2]. The application of an external magnetic field orthogonal to the sample plane, binds
the free electrons into orbits, which breaks the continuous energy spectrum into a set of discrete, gapped constant-energy levels, the Landau levels. Unexpectedly, the Hall conductivity is not zero whenever the Fermi level falls between two Landau levels, but instead maintains a constant value that changes in discrete steps as the field strength is tuned. Soon, it became clear that the quantized conductivity was a universal effect, unaffected by irregularities of the crystal and realizable in any conductor. The resistivity steps are found to be quantized by a material-independent constant, the von Klitzing constant \( R_K = h/e^2 \approx 25813 \ \Omega \), which is measurable with an accuracy of one part in \( 10^{10} \) today [3, 4]. The intuitive picture is that electrons in the bulk of the system are bound in orbits and do not contribute to the current. At the edges of the sample, however, the formation of these orbits is obstructed, unavoidably leading to a conduction channel along the edge via skipping orbits, see Fig. 1.1. Thouless, Kohmoto, Nightingale and den Njis (TKNN) formulated the observed quantized conductivity in terms of a topological number called the \textit{TKNN invariant} [5]. A short introduction to topology is provided in Chapter 2. The TKNN invariant can be related to the Berry flux [6]. Berry’s quantities provide a convenient tool to quantify topological properties. They measure the change of wave functions \( \psi(R) \) of a system in some parameter space spanned by a vector \( R \), in terms of a geometric phase, the \textit{Berry phase} [6]. On closed loops in parameter space, the integrated phase must be gauge invariant and thus allows only discrete values, namely \( 2\pi n \), where \( n \) is an integer. Since these are discrete values, changing them requires a discontinuity in the wave function or the Hamiltonian of the system. This occurs in the QHE whenever a Landau level crosses the Fermi energy and leads to jumps in the conductivity, as we will discuss in detail in Chapter 4. In 1988, Haldane has shown that the quantum Hall effect can in principle be realized without the application of an external tuning parameter like the magnetic field [7]. He considered a honeycomb model, in which the time-reversal symmetry is broken magnetic doping instead, giving rise to the \textit{quantum anomalous Hall effect} (QAHE), a spin-polarized version of the QHE.
Prior to this, a different development originated on the side of particle physics. In 1969, S. L. Adler, and J. S. Bell and R. Jackiw worked on the decay of the pion π⁰ → γγ into two photons. The theoretical decay time of the process could not be brought into agreement with the much smaller experimental values. The mentioned authors found a solution to the puzzle in the ABJ anomaly [8, 9], also called the chiral anomaly. The famous Feynman-diagram of the pion decay is shown in Fig. 1.2, which provides an extra decay channel, thus shortening the decay time. The name “chiral anomaly” refers to the consequences this effect has for the chiral conservation laws of Weyl fermions, which we want to explore below. Therefore, consider the Hamiltonian of a massless Dirac fermion [10],

\[ \hat{H} = c \alpha \hat{p} = \begin{pmatrix} -c \sigma \cdot \hat{p} & 0 \\ 0 & c \sigma \cdot \hat{p} \end{pmatrix} = \begin{pmatrix} \hat{H}_L & 0 \\ 0 & \hat{H}_R \end{pmatrix}, \] (1.1)

in the chiral basis. Here, \( c \) is the speed of light, \( \hat{p} = -i\hbar \nabla \) is the momentum operator, \( \sigma = (\sigma_x, \sigma_y, \sigma_z) \) is the vector of Pauli matrices and \( \alpha = (\alpha_x, \alpha_y, \alpha_z) \) are the \( \alpha \)-matrices given as \( \alpha_i = -\sigma_z \otimes \sigma_i \) in the chiral basis, with \( i \in \{x, y, z\} \). In this massless Dirac equation, the spinor \( \psi \) separates into two chiral sectors \( \psi = (\psi_L, \psi_R) \). The chirality refers to the spin and momentum vectors being aligned in antiparallel for \( \psi_L \) (left-handed) and in parallel for \( \psi_R \) (right-handed). The 2 × 2-Hamiltonians \( \hat{H}_{L,R} \) are known as the Weyl Hamiltonians and the chiral wavefunctions \( \psi_{L,R} \) describe the Weyl fermions. The associated relativistic four-current of the Dirac fermions is \( J^\mu = \psi^\dagger \gamma^0 \gamma^\mu \psi = (c \hat{\rho}, \hat{j}) \), with \( \mu \in \{0, 1, 2, 3\} \). The \( \gamma \) matrices in the chiral basis are \( \gamma^0 = \sigma_x \otimes 1, \quad \gamma^i = i \sigma_y \otimes \sigma_i \). The four-current contains the particle density \( \hat{\rho} = \psi^\dagger \psi \) in the zero-component, while the remaining entries contain the particle current \( \hat{j} = c \psi^\dagger \alpha \psi \), where \( \alpha_i = \gamma^0 \gamma^i \).

Figure 1.2: Feynman diagram of the ABJ anomaly in the pion decay π⁰ → γγ.
This particle current fulfills the continuity equation
\[ \partial_{\mu} J^\mu = 0, \]  
\[ \partial_t \hat{\rho} + \nabla \cdot \hat{j} = 0, \]  
\[ \partial_t \hat{N} + \int_S dS \hat{j} = \Sigma, \]

where the last equation is its integral form. The continuity equation implies that the change of the particle number \( \hat{N} \) within a volume \( V \) is associated with a current through the surface \( S \) of the volume, visualized in Fig. 1.3. If a source of particles is present within \( V \), then \( \Sigma \) will be a positive number, if a sink is present, \( \Sigma \) will be negative, and if the particle number is conserved, then \( \Sigma = 0 \). We can define chiral currents \( \hat{j}_{L,R} = c \psi_{L,R}^\dagger \alpha \psi_{L,R} \) that fulfill their own continuity equations, in particular for the difference in left- and right-handed particles,

\[ \partial_{\mu} J_5^\mu = 0 \]
\[ \partial_t (\hat{\rho}_L - \hat{\rho}_R) + \nabla \cdot (\hat{j}_L - \hat{j}_R) = 0, \]

where \( J_5^\mu = \psi^\dagger \gamma^0 \gamma^\mu \gamma^5 \psi \) is the chiral particle four-current, with \( \hat{\rho}_{L,R} = \psi_{L,R}^\dagger \psi_{L,R} \) and \( \gamma^5 = i\gamma^0 \gamma^1 \gamma^2 \gamma^3 = -\sigma_z \otimes 1 \). The chiral particle density \( \hat{\rho}_L - \hat{\rho}_R \) quantifies the difference in left- versus right-handed particle densities and \( \hat{j}_L - \hat{j}_R \) defines a chiral current density. This chiral continuity equation should preserve the amount of left- and right-handed fermions within a volume if there are no currents exchanged through the surface. This is where the discovery of the chiral anomaly in 1969 becomes important. The chiral anomaly happens to be the very first quantum anomaly: It turns out that when a “classical” field theory is quantized, some symmetries may
break down. In this case, the chiral symmetry is broken and an anomalous chiral particle current is generated. The chiral current operator then fulfills

$$\partial_\mu J_5^\mu = \frac{e}{16\pi^2\hbar^2} F_{\mu,\nu} \tilde{F}^{\mu,\nu} = \frac{e^2}{4\pi^2\hbar^2} \mathbf{E} \cdot \mathbf{B} \neq 0 \quad (1.7)$$

with $\tilde{F}^{\mu,\nu} = \frac{1}{2} \epsilon^{\mu,\nu,\rho,\lambda} F_{\rho,\lambda}$, where $F_{\mu,\nu}$ is the field-strength tensor and $\epsilon^{\mu,\nu,\rho,\lambda}$ is the Levi-Civita tensor. This implies that while in a classical field theory the chirality of a system is preserved, the application of an electric field $\mathbf{E}$ and a magnetic field $\mathbf{B}$ in parallel instead changes the net chirality of the system when quantum effects are accounted for. This generates an imbalance in the number of chiral particles. The total particle density, however, remains conserved of course. In the case of the massless Weyl fermions, the application of parallel fields permanently changes the net chirality of the system. The presence of a mass term on the off-diagonal entries in the Hamiltonian of Eq. (1.1) would mix the chiral subspaces and allow for a relaxation of such a chiral imbalance. At the time, Weyl fermions were thought to be realized in the form of neutrinos [11, 12]. Today we know that neutrinos have finite masses, as indicated by neutrino flavor oscillations [13], and thus no free Weyl fermion is currently known to exist.

Let us take a look at a special application of the chiral anomaly, which is the axion [14–16]. The anomalous current modifies the Lagrangian of quantum field theory by a term [14]

$$\mathcal{L} = \frac{\theta g^2}{32\pi^2} F_{\mu,\nu} \tilde{F}^{\mu,\nu}, \quad (1.8)$$

and breaks the chiral symmetry of the field theory. The parameter $g$ describes the coupling strength between the electromagnetic field and the fermion, whereas the parameter $\theta$ is a free parameter that can take any value between 0 and $2\pi$. The chiral anomaly exists in an equivalent fashion for gluon-fields $G_{\mu,\nu}$ (replace $F_{\mu,\nu}$ by $G_{\mu,\nu}$ in
the above equation) in quantum chromodynamics (QCD) [17]. The gluon fields are responsible for binding the quarks together that make up neutrons and protons. One observable for which the effect of the chiral anomaly should become relevant, is the electric dipole moment of the neutron, which changes when the $\theta$-term is included in the theory. Theoretical estimates yield values of the order $d_n^{\text{theo}} = 4.5 \times 10^{-16} \theta \text{ em}$, where $e$ is the electron charge, whereas measurements provide an upper bound $d_n^{\text{exp}} < 2.9 \times 10^{-26} \text{ em}$. This implies that $\theta$ must be of the order of $\theta < 10^{-11}$. Since $\theta$ is allowed to take any value between 0 and $2\pi$, it is odd that it is this small for no concrete reason. This constitutes the strong CP problem (CP: Charge-Parity). The most promising solution to date is to promote $\theta$ to a (periodic) field $\theta(r)$, such that the potential energy is minimized if the particle fields of QCD settle in the minima of $\theta(r)$. This is the Peccei-Quinn theory, and it leads to a bosonic particle called the axion [14–16].

This axion formalism provided one of the contact points between topological condensed-matter and particle physics. In 1986 Fradkin et al. proposed the existence of a surface state in the semiconductor PbTe [18, 19]. The claim of Fradkin et al. was that a surface state can give rise to “quasi quantum Hall currents” in PbTe without the necessity to break time-reversal or inversion symmetry, arising solely from the action of spin-orbit coupling. Wilczek’s subsequent work showed that this surface state can be described by the axion Lagrangian of particle physics. His paper of 1987, the “Two applications of axion electrodynamics” [20], established a connection between the chiral anomaly of particle and condensed matter physics, where in the condensed matter version the presence of the $\theta$-term is tied to the existence of the surface state. In the bulk of the sample, they set the angle $\theta$ to $\pi$, while at the surface of the sample, bounded to vacuum, $\theta$ drops to 0. Areas within which $\theta$ is constant produce no new physics. Thus, the bulk of a sample with a constant $\theta = \pi$ is physically identical to the same system with a constant $\theta = 0$, or without a $\theta$-term. The axion electrodynamics
exclusively take place on the surface of the sample, where a sudden change of $\theta = \pi$ to $\theta = 0$ occurs, giving rise to a conducting surface channel. With that, the **axion insulator** was born, the first **topological insulator** [21, 22].

On the other hand H. B. Nielsen and M. Ninomiya found in 1983 that the ABJ anomaly is more readily accessible without the need for the axion formalism [23]. As C. Herring already suggested in 1937 [24], an analog of the Weyl fermion can potentially be realized in the electronic structure of condensed matter systems in which non-degenerate electronic bands cross at singular points in the shape of a cone. Materials hosting such Weyl cones are now known as a Weyl semimetal. Nielsen and Ninomiya had found that in such systems, parallel to the magnetic field conical bands can produce a single linear Landau level per cone with opposite dispersions for particles of opposite chirality. Here, the chiral anomaly arises in a very intuitive way when an electric field is applied in parallel to the magnetic field, see Fig. 1.4. Due to the opposite dispersions of the energies $E(k)$ of the linear Landau levels ($k$ is the wave number), the electric field forces the electrons down below $E_F$ in one cone, effectively emptying it, and up the other cone, filling it, creating a chiral imbalance.

With the Weyl semimetals, we now have a platform on which we can investigate quantum field theory effects in bulk sensitive measurements. This garnered much attention when electronic band structure calculations had begun to find real potential candidates [25].
As mentioned, the boundary of the Weyl semimetal surface to vacuum induces a change in $\theta$, giving rise to a special type of surface states. These surface states interconnect Weyl cones of opposite chirality in momentum space and are called Fermi arcs. Angle resolved photoemission spectroscopy (ARPES) provided the smoking-gun experiment in 2015 [26, 27], by detecting these arcs in TaAs, which is now accepted as the first experimentally confirmed Weyl semimetal. After that, the TaAs “family”, which includes the four members TaAs, TaP, NbAs and NbP, enjoyed much popularity in the research community and so, likewise, our group started investigating these materials [26, 28–35].

In an effort to investigate their Weyl characteristics, in the scope of this thesis optical studies in magnetic fields on the candidates TaP, NbAs, and GdPtBi were performed. The experiment of choice, infrared magneto-optics, cannot probe the vanishingly small topological surface states directly. However, we can investigate the Weyl fermions of the bulk, by making use of the characteristic $\sqrt{B}$-dependence of Landau levels originating from linear dispersing electron bands, differentiating them from trivial massive electrons, as we will explore in Chapter 3. We will find that there are many subtle details to bulk Landau levels, which help uncover the inner life of topological materials. First, however, let us take a short look at the topological formalisms relevant to condensed-matter physics.
Topological materials constitute phases of matter whose physical surface properties differ from those in their bulk. Topological insulators, for example, host gapless electron bands on the surface that provide conducting channels. The bulk of the material, however, remains gapped and insulating [36–38]. These effects can be quantified by topological invariants, which are derived from wavefunctions of the system. The beauty of topological invariants is that we need only to evaluate properties of the bulk to derive qualitative physical properties of the surface, i.e. whether the surface bands are gapless or not. We call this relationship between bulk and surface of a material the bulk-boundary correspondence. To get a feeling for the subject, we will first take a look at concepts geometric topology.

2.1 Geometric Topology

The term “topology” was coined by Johann Benedikt Listing, a student of Carl Friedrich Gauss, in his “Vorstudien zur Topologie” published in 1848. Listing defined the term to relate to “the study of modal relationships of spatial structures and the study of laws of correlation in the position and sequence of points, lines, planes, bodies and their constituents or aggregates in space”. In his work he compiles many known examples that are topological, such as, different loops of string with joint ends that can be differentiated by how many
knots they contain. Or how there are two ways to print a six-sided dice. The common theme being how to arrange physical objects or mathematical sequences that are similar to one another in character, but cannot be transformed into each other. The two different dices cannot be matched by rotations and two loops with different numbers of knots cannot be disentangled in some way into the same shape without cutting the string, which in Listing’s words constitutes topological inequivalence. By that time, some important discoveries in geometric topology were already made. In 1758 Leonard Euler formulated the Euler characteristic $\chi$, relating the number of vertices (V), edges (E) and faces (F) for polyhedra as

$$\chi = V - E + F = 2. \quad (2.1)$$

This makes the polyhedra depicted in Fig. 2.1 topologically equivalent. We can see that, if we increase the number of vertices indefinitely, we arrive at a smooth sphere, for which the same relation should hold. We can also deduce that any deformations of the polygons will not change the outcome. From these considerations it is intuitive then, that a continuous formulation of the theorem should exist. In 1827, Gauss states such a relation in his remarkable theorem [39, 40].
CHAPTER 2. TOPOLOGY

Theorema Egregium - “The Remarkable Theorem”

“Si superficies curua in quacunque aliam superficiem explicantur, mensura curuaturae in singulis punctis inuarata manet. [...] Quaeuis pars finita superficie curvae post explicationem in aliam superficiem eandem curvaturam integram retinebit.”

“If a curved surface is a development of any other surface, the measure of the [Gaussian] curvature remains unchanged in every single point. [...] Any finite part of the curved surface will retain the same integral [Gaussian] curvature after development upon another surface.”

Curvatures at a point on a surface are calculated as $\kappa = 1/r^2$, where $r$ is the radius of a circle that is tangential at that point. At any point, we can find a maximum and a minimum curvature, the principle curvatures $\kappa_1$ and $\kappa_2$. We obtained the Gaussian curvature $K$ by multiplying them

$$K(r) = \kappa_1(r)\kappa_2(r). \quad (2.2)$$

The remarkable theorem states that if a surface is transformed, without distorting distances, the curvature stays the same at any point. For example, a crumpled sheet of paper has the same Gaussian curvature as a flat one at any point, namely zero. Conversely, the surface of a sphere has a constant Gaussian curvature at any point. This implies that a sheet of paper cannot have a spherical dent. This is why no projection of a sphere onto a flat map can exist that preserves lengths, a problem we are familiar with from cartography.
when trying to project earth onto a map. It also follows that the integral of the curvature over the surface stays invariant. As an example, for the surface $S$ of the unit sphere, we have $\kappa_1 = \kappa_2 = 1$, which integrates to

\[
\text{Sphere: } \int_S K(r) = 4\pi. \tag{2.3}
\]

The Gauss-Bonnet theorem, was formulated by Pierre Ossian Bonnet in 1848 [41] and states that such an integral of the Gaussian curvature over a closed surface fulfills the relation

\[
\int_S KdS = 2\pi\chi, \tag{2.4}
\]

where $\chi$ is in fact the same Euler characteristic as for the discrete case of polyhedra in Eq. (2.1). Moreover, Bonnet finds a rather surprising connection: The Euler characteristic $\chi$ can be related to the number of holes $g$, the genus, in the structure we integrate over via

\[
\chi = 2 - 2g. \tag{2.5}
\]

This statement remains true even for transformations that are not length-preserving, that is, for surfaces without a boundary.

Figure 2.3: A torus and a sphere with hair combed flat onto their surface. The sphere’s topology enforces the existence of two vortices or radial points.
The integration of a sphere then yields
\[ \chi_{\text{Sphere}} = 2. \] (2.6)

Conversely, for the torus, which has one hole, we have \( \chi_{\text{Torus}} = 0 \). This means that the torus and the sphere are topologically distinct, however, a donut and a coffee cup are topologically equivalent. This connection between genus and Euler characteristic seems magical. Rather than having to count vertices, or perform possibly difficult surface integrals, simply counting the number of holes of two objects tells us whether they can be continuously transformed into each other. This makes topological invariants, such as the genus, powerful tools. In the case of geometric shapes, a physical consequence of topology is, for example, the hairy ball theorem. It was proven by Henri Poincaré in 1885 and states that hair on a sphere cannot be combed flat without the occurrence of at least two singular points, i.e., vortices. Conversely, on a torus all hair can be combed flat. This is visualized in Fig. 2.3.

Another example is the parallel transport of a vector on a sphere. If one moves along a closed path on the surface of a sphere such that vector locally never changes direction, the starting and end vector do generally not coincide. A succinct way to illustrate this is shown in Fig. 2.4, where a closed curve with three 90° angles is depicted. This is unlike the situation on a flat plane, where a closed path with 90° angles must contain four vertices, and the starting vector is identical with the end vector. In the next section, we find that similar concepts hold true for electrons described by Bloch functions. Here, we may also define curvatures and invariant surface integrals, which can be related to physical observables. Whether the Bloch functions of two adjacent systems can be transformed into one another leads
to observable physical consequences, which we touched on in the introductory Chapter 1: the surface currents.

2.2 Berry’s quantities

We can also formulate topological quantities for electronic systems by evaluating the relationships between continuous wavefunctions. In 1984, Michael Berry pointed out [6] that eigenfunctions \( |n(r, t)\rangle \) of Hamiltonians

\[
\hat{H} |n(r, t)\rangle = E_n(t) |n(r, t)\rangle,
\]

collect not only a dynamical phase factor

\[
\Phi_n(t) = \frac{1}{\hbar} \int_0^t E_n(t') dt','
\]

but can additionally host a geometric phase \( \gamma_n(r) \), also known as the Berry phase. The parameter \( r \) is the real space coordinate, but the Berry phase can be equivalently defined for other parameter spaces, such as the wave vector \( k \) in lattices. A Berry phase is collected when we adiabatically vary a wave function over the parameter space. For a scalar parameter \( r \), an infinitesimal variation yields a phase contribution

\[
\langle n(r)|n(r + dr)\rangle = e^{-id\gamma_n(r)}.
\]

The continuous version is obtained as

\[
\langle n|\partial_r|n\rangle := \frac{\langle n|(|n(r + dr)\rangle - |n(r)\rangle)}{dr},
\]

\[
dr \langle n|\partial_r|n\rangle = e^{-id\gamma_n} - 1 = -id\gamma_n, \quad \partial_r \gamma_n = i \langle n|\partial_r|n\rangle,
\]
where we use the relation $e^{id\gamma_n} = 1 + id\gamma_n + \mathcal{O}(d\gamma_n)$. We can generalize this for vector-valued parameters $\mathbf{r}$ and write

$$\nabla_{\mathbf{r}}\gamma_n(\mathbf{r}) = i \langle n | \nabla_{\mathbf{r}} | n \rangle = \mathbf{A}_n(\mathbf{r}), \quad (2.13)$$

$$\gamma_n(\mathbf{r}) = \int \mathbf{A}_n(\mathbf{r}) \, d\mathbf{r}, \quad (2.14)$$

where we define the *Berry connection* $\mathbf{A}(\mathbf{r})$. The Berry phase $\gamma_n(\mathbf{r})$ is not gauge invariant, meaning that transformations of the form $|\tilde{n}(\mathbf{r})\rangle = e^{i\beta(\mathbf{r})} |n(\mathbf{r})\rangle$, where $\beta(\mathbf{r})$ is an arbitrary phase factor, alter the Berry phase. However, the Berry phase is gauge invariant modulo $2\pi c$ along closed paths [42]

$$\gamma_n(\mathbf{r}) = \oint \mathbf{A}_n(\mathbf{r}) \, d\mathbf{r} + 2\pi c, \quad c \in \mathbb{Z}. \quad (2.15)$$

It is often more useful to work with the quantity called the *Berry flux*

$$\Omega_n(\mathbf{r}) = \nabla_{\mathbf{r}} \times \langle n | \nabla_{\mathbf{r}} n \rangle = \nabla_{\mathbf{r}} \times \mathbf{A}_n(\mathbf{r}). \quad (2.16)$$

With Stokes’ theorem, we have

$$\int_S \Omega_n d^2\mathbf{r} = \oint_{\partial S} \mathbf{A}_n d\mathbf{r} = \gamma_n. \quad (2.17)$$

Integrating the Berry flux on a surface $S$ yields the Berry phase around its boundary. As we know, we can subdivide the integral over a rotation field on a surface into plaquettes. The contribution to the line integral at touching borders of two neighboring plaquettes cancels out in the integral, as implied by Stokes’ theorem. If we apply this logic to any closed surface, we expect the line integral to disappear. However, this is true only if we can find a continuous parametrization $\Omega$ on the entire surface. Though, this is not always possible as we will see in the prominent example of a spin-$\frac{1}{2}$ particle in the presence of a magnetic monopole. It turns out that for closed surfaces, the integral of the Berry flux is quantized in steps of
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This defines the quantized measure $C$, called the *Chern number* [43]

$$\int_S d^2 r \, \Omega_n(r) = 2\pi C, \quad C \in \mathbb{Z}. \quad (2.18)$$

Before we move on to the example case, we summarize the most important quantities below.

**Topology – Formula Overview**

- **Berry Phase**: $\gamma_n(r) = \oint A_n(r) dr + 2\pi n, \quad n \in \mathbb{Z}$
- **Berry Connection**: $A_n(r) = i \langle n(r)|\nabla_r|n(r)\rangle$
- **Berry Curvature**: $\Omega_n(r) = \nabla_r \times A_n(r)$
- **Chern number**: $C_n = \frac{1}{2\pi} \int_S \Omega_n(r) d^2 r, \quad C \in \mathbb{Z}$

It is common to rewrite the Berry flux as [6]

$$\Omega_n = -i \sum_{m \neq n} \frac{\langle n|(\nabla \hat{H})|m\rangle \times \langle m|(\nabla \hat{H})|n\rangle}{(E_n - E_m)^2}, \quad (2.19)$$

where we dropped the $r$-dependencies for brevity. Eq. (2.19) is useful, since this way we do not need to know the analytical expression for the eigenstates $|n\rangle$, required for the derivative $\nabla |n\rangle$.

### 2.2.1 Spin-$\frac{1}{2}$ particle and magnetic monopole

We pointed out that the integral of the Berry flux on a closed surface yields a quantized Chern number. However, using Stokes’ theorem we had argued also that due to a vanishing surface contour, we
expect the integral of the Berry flux to be zero. This apparent con-
tradiction can be resolved. Vanderbilt states 

“[...]When the Chern index is non-zero, it is impossible to
construct a smooth and continuous gauge over the entire
surface \( S \). If such a gauge did exist, then we could apply
Stokes’ theorem directly to the entire surface and conclude
that the Chern number vanishes.”

A useful example to demonstrate this is the spin-\( \frac{1}{2} \) particle in an
external magnetic field. The magnetic field is varied adiabatically,
such that the spin of the particle stays exactly aligned with the
field. This is equivalent to a spin-\( \frac{1}{2} \) particle that is adiabatically
moved around a magnetic monopole with magnetic field \( \mathbf{B}(r) = B_0 \frac{r}{r^2} \),
where \( r = (x, y, z)^T \) is the position vector and \( r = |r| \). The
corresponding Hamiltonian is

\[
\hat{H}(r) = -g \mu_B s \mathbf{B}
\] (2.20)

with \( s = \mathbf{\sigma}/2 = (\sigma_x, \sigma_y, \sigma_z)/2 \) and the Pauli matrices \( \sigma_i \). It describes
the Zeeman splitting of a fermion in the presence of a magnetic
monopole. When the magnetic field is applied along the \( z \)-axis, the
eigenstates correspond to \( |s_{z+}\rangle = (1, 0)^T \) and \( |s_{z-}\rangle = (0, 1)^T \). However,
since the problem is spherically symmetric, no spin direction
is favoured. Then, as we move a single spin adiabatically around
the magnetic monopole, its spin direction follows the alignment of
the hedgehog or anti-hedgehog structure depicted in Fig. 2.5. This
continuous alignment is associated with a Berry phase that one picks
up as a path around the monopole is traversed. A general solution
is thus given by any eigenstate on the Bloch sphere. We may now
choose the continuous gauge

\[
|s(\vartheta, \varphi)\rangle = \left( \begin{array}{c} \cos(\vartheta/2) \\ \sin(\vartheta/2)e^{i\varphi} \end{array} \right)
\] (2.21)
where

\[ |\uparrow\rangle_x = (1, 1)^T / \sqrt{2} = |s(\pi, 0)\rangle, \quad |\downarrow\rangle_x = (1, -1)^T / \sqrt{2} = |s(\pi, \pi)\rangle \]
\[ |\uparrow\rangle_y = (1, i)^T / \sqrt{2} = |s(\pi, \pi/2)\rangle, \quad |\downarrow\rangle_y = (1, -i)^T / \sqrt{2} = |s(\pi, 3\pi/2)\rangle \]
\[ |\uparrow\rangle_z = (1, 0)^T = |s(0, \varphi)\rangle, \quad |\downarrow\rangle_z = (0, e^{i\varphi})^T = |s(2\pi, \varphi)\rangle. \]

Note that near the “south pole” (\(\vartheta = \pi\)) of our chosen gauge we have \(|\downarrow\rangle_z = (0, e^{i\varphi})^T\). The wave function is not uniquely defined and in fact, no matter how we choose the gauge, we can not avoid this topological obstruction [42, 44]. It is easy to see that integrating an infinitesimally small loop around the south pole yields a Berry phase of \(\theta = \pm 2\pi\). This is nothing but the Chern number multiplied by \(2\pi\).

However, we can use Eq. (2.19) instead to calculate the Chern number. In this way, we do not have to concern ourselves with the parametrization of the wavefunction, as mentioned earlier. After some calculation, one obtains

\[ \Omega_{\pm}(r) = \pm \frac{1}{2} \frac{r}{r^3}, \quad (2.22) \]

which describes a monopole of Berry flux. Calculating the Berry phase on a closed loop around the monopole corresponds to integrating the Berry flux within the area of the closed loop. One finds
that the surface integral of Eq. (2.22) yields the solid angle, multiplied by a factor of $\pm \frac{1}{2}$

$$
\int\int_S \Omega_{\pm}(\mathbf{r})d^2\mathbf{r} = \pm \frac{1}{2} \int\int_S \sin(\varphi) d\varphi d\theta.
$$

(2.23)

Since the solid angle of a sphere is $4\pi$, the Chern number associated with an integration around the entire monopole is

$$
C_{\pm} = \pm 1.
$$

(2.24)

Let us now investigate Bloch wavefunctions in condensed matter, which are subjected to the crystal fields. There, we will find the physics of special band structure features, called the Weyl cones, to turn out very similar to the discussion above.

### 2.3 Topology in $k$-space

Their electronic wavefunctions of periodic lattices are given by the Bloch functions

$$
\psi(\mathbf{r})_{n,k} = \frac{1}{\sqrt{V}} e^{i\mathbf{k}\cdot\mathbf{r}} u_n(\mathbf{r}), \quad \hat{H} |n, \mathbf{k}\rangle = E_n(\mathbf{k}) |n, \mathbf{k}\rangle
$$

(2.25)

where we denote $\psi(\mathbf{r})_{n,k} = \langle \mathbf{r}|n, \mathbf{k}\rangle$. Here, $V$ is the volume of the crystal and the wave vectors, $\mathbf{k}$. The state $u_n(\mathbf{r})$ is a lattice-periodic
Figure 2.7: Weyl cones of opposite helicities. The cone with left-handed helicity (spins anti-parallel to momentum) plotted on the left, right-handed (spins parallel to momentum) on the right.

\[ u_n(r) = \sum_j a_{nj} e^{i\mathbf{G}_j \cdot \mathbf{r}}, \quad (2.26) \]

where the \( \mathbf{G}_j \) are wave vectors with the periodicity of the lattice, \( a_j \) are the amplitude coefficients. The wave vectors \( \mathbf{k} \) span Brillouin zones and the energies \( E_n(\mathbf{k}) \) define electronic bands, which may intertwine in complicated ways, giving rise to various topologies. We define a Berry phase analogously to the previous section, but for the parameter space spanned by \( \mathbf{k} \). We note, however, that in \( k \)-space, two neighbouring states in \( |n, \mathbf{k}\rangle \) and \( |n, k + d\mathbf{k}\rangle \) are orthogonal, since the parameter \( \mathbf{k} \) is also an eigenvalue to the Hamiltonian. Though, for closed paths nothing changes and the Berry phase can be calculated as usual. The Hamiltonian describing condensed matter systems generally contains too much information. Typically, we want to describe a few bands near the Fermi surface. Fig. 2.6 displays a part of the band structure of TaAs near the Fermi surface is plotted. We are interested in the two intersecting bands near the Fermi surface. In the immediate vicinity of this intersection, the
Hamiltonian can be expressed as

$$\sum_{n} \langle n, k | \hat{H} | n, k \rangle = \begin{pmatrix} \ddots & H(k)_{2 \times 2} \\ H(k)_{2 \times 2} & \ddots \end{pmatrix}$$

(2.27)

The right-hand side of Eq. (2.27) is the Hamiltonian in \( k \)-space representation, in which we work from here on out. We are only interested in the two intersecting bands and thus isolate the Hamiltonian \( H_{2 \times 2} \)

$$H(k)_{2 \times 2} = H_{\text{Weyl}} = v_F \hbar k \sigma.$$ 

(2.28)

For simplicity, we assume an isotropic dispersion relation. This equation describes nothing other than a Weyl cone, the condensed-matter analog of the Weyl fermion [10], which we discussed in the introduction, Chapter 1. Its energy spectrum is given by

$$E_{\text{Weyl}} = \pm v_F \hbar \sqrt{k_x^2 + k_y^2 + k_z^2},$$

(2.29)
which describes electronic bands that intersect linearly in a point. It is obtained from the Dirac equation in the limit of a vanishing mass, which then consists of two copies of the Weyl Hamiltonian with opposite signs (for free fermions replace $v_F \hbar \mathbf{k} \to c\mathbf{\hat{p}}$ in Eq. (2.28)). In Fig. 2.7 we have plotted the spectrum in the $k_x$-$k_y$ plane to illustrate its characteristic cone shape. The Weyl Hamiltonian closely resembles that in Eq. (2.20). Here, instead of the presence of a magnetic monopole, it is the action of the spin-orbit coupling that aligns the spins in a hedgehog formation. Similar to the discussion of Berry’s Hamiltonian, Chern numbers can be calculated for these cones. $C = 1$ for the all-out configuration and $C = -1$ for the all-in configuration of spins.

An interesting point was raised by Dirac [45] concerning magnetic monopoles. From the discussion in Section 2.2.1, we remember that such a pole comes with an anomalous point, where a smooth gauge cannot be defined. Since this is obviously true at any distance from the magnetic monopole, we expect this anomalous point to extend to a nodal line. We call this the **Dirac strings**. Such a Dirac string must either extend to infinity or end on a magnetic monopole of
opposite magnetic charge. In the context of Weyl points, a Dirac string connects two monopoles of opposite chirality, or equivalently, opposite Chern numbers. At first, this seems like unimportant mathematical trickery, but it turns out that these Dirac strings are in fact experimentally observable in the form of Fermi arcs. These are surface states between Weyl cones of different chiralities [46]. The detection of these Fermi arcs in ARPES measurements is considered the smoking gun experiment to detect Weyl fermions in matter. Their successful detection of Fermi arcs confirmed TaAs as the first inversion-symmetry broken Weyl semimetal [26, 27, 47]. The Fermi arcs of TaAs are qualitatively sketched in Fig. 2.9.

2.3.1 Chern number and Hall conductivity

As pointed out earlier, the hallmark of topological materials are their conducting surface states. Take, for example, a gapped topological insulator, which we can obtain by replacing the $k_z$ dispersion of the Weyl Hamiltonian with a constant term $\Delta$,

$$H(k) = \hbar v_F (k_x \sigma_x + k_y \sigma_y) + \Delta \sigma_z.$$ 

Its energy dispersion is plotted in Fig. 2.10 for the parameters $\hbar v_F / a = \frac{4}{3} \Delta$, where $a$ is the length of the unit cell, assumed to be isotropic along $x$ and $y$. The arrows on the energy bands indicate the Berry flux for each band (arbitrary scale), which contains only a $z$-component for this model. The Berry flux integrates to $C = +1$ and $C = -1$ for the conduction and valence band respectively. As a consequence, two singly degenerate bands must cross the energy}

Figure 2.10: Berry flux profile of a gapped topological insulator model in arbitrary units.
gap as illustrated in Fig. 2.11. These topological states give rise to quantized currents. The formula of Thouless, Kohmoto, Nightingale and den Nijs [5], provides the integer quantum Hall conductivity for a two-dimensional conductor in magnetic fields

$$\sigma_{x,y} = \frac{ie^2}{h} \sum_{E_n > E_F} \sum_{E_m < E_F} \frac{\langle m|\partial k_x \hat{H}|n\rangle \langle n|\partial k_y \hat{H}|m\rangle - \langle m|\partial k_y \hat{H}|n\rangle \langle n|\partial k_x \hat{H}|m\rangle}{(E_n - E_m)^2}$$

with $A$ being the surface area. Comparing with Eq. (2.19), we see that Eq. (3.36) contains the Berry flux and we obtain the term for the conductivity

$$\sigma_{x,y} = \frac{e^2}{h} C_n. \quad (2.30)$$

We obtain a surface current analogous to the integer quantum Hall effect. For a real material, with two orientable surfaces, we find two oppositely spin polarized surface states, as depicted in Fig. 2.11. These topological states are interesting due to their robustness. To eliminate them, one needs to change the Chern number of the system. This requires a discontinuous jump of the Chern number that can only be achieved by closing a band gap, forcing the system through topological phase transition. Lattice imperfections that locally break the topological phase will be skirted around by these surface states, as the boundary of such an imperfection is just another interface between a topological and a normal insulator and thus the surface currents remain dissipationless. This makes topological compounds interesting in the context of spintronics, quantum computing and metrology among many other fields. A prime example for this robustness of topological states is the experiment of Chang et al. [48] who were able to observe a near perfect quantization of the Hall current in a ferromagnetic thin film \(^1\), with the Hall terminal scratched

\(^1\)That is, the quantum anomalous Hall effect (QAH), in which the two spin channels are energetically well separated due to ferromagnetic exchange, allowing the measurement of a single spin channel.
CHAPTER 2. TOPOLOGY

Figure 2.11: Qualitative energy spectrum of the gapped topological insulator described by Eq. (2.3.1) on the left and a topological edge state carrying a conductance quantum at the edge of a topological insulator in real space on the right.

directly into the sample. In the Hamiltonian of Eq. (2.3.1), the topological transition occurs at \( m = 0 \), which corresponds to the Weyl Hamiltonian in two dimensions. The Weyl semimetal state can be understood as an intermediate phase between a topological insulator and a normal insulator. Thus, early proposals to realize a Weyl semimetal included layered heterostructures of normal and topological insulators [49, 50]. While in the present case of Eq. (2.3.1) the Weyl cone exists only exactly at \( m = 0 \), that is not universally the case. The Weyl semimetal state can in fact span a finite region in parameter space. Further, its existence has lax symmetry requirements: Weyl cones can occur if either the time-reversal symmetry or the inversion symmetry is broken [42, 51, 52].

In the case of a broken inversion symmetry, the presence of spin-orbit coupling can lift the degeneracy of spins, leading to pointlike intersections similar to the Rashba [53] or Dresselhaus effect [54]. While the strength of spin-orbit coupling effects is generally rather small, modern density functional theory (DFT) calculations can predict them reliably and a large database of topologically classified ma-
materials is available [55]. On the other hand, breaking time-reversal symmetry involves either external magnetic fields, the presence of free spins, or both. Since the exchange interaction of spins can lead to much larger band shifts compared to spin-orbit coupling effects, there remains a great potential in the search of magnetic topological phases.

In this chapter, we covered the basics of topological materials, which can be quantified using Chern numbers. As we have seen, Weyl semimetals belong to the topologically non-trivial materials. The associated topological obstruction gives rise to the Fermi arcs, which are a special type of surface states interconnecting Weyl cones of opposite chiralities. These surface states are not only an ideal footprint for identifying topological phases, but are also host to a quantized Hall conductivity with potential applications due to their inherent robustness. In the next chapter, we are discussing the Landau quantization of Bloch electrons in an external magnetic field. Since topological materials typically come with characteristic band dispersions, optically probing the Landau level spectrum offers an alternative route to identify material phases.
Chapter 3

LANDAU QUANTIZATION OF BAND ELECTRONS

In a strong external magnetic field, electrons condense into Landau levels. By investigating Landau-level energies, we can extract information about band electrons in condensed matter systems. In this chapter, we discuss the Landau quantization for three essential models: massive electron in parabolic bands, massless Weyl electrons and briefly also massive Dirac electrons. Further, the chiral anomaly of Weyl semimetals in magnetic fields will be shortly addressed.

3.1 Schrödinger Electrons

The Schrödinger Hamiltonian of a massive free electron is

$$\hat{H} = \frac{\hat{p}^2}{2m_e},$$

(3.1)

with momentum $\mathbf{p} = -i\hbar \nabla$ and the free electron mass $m_e$. For free electrons, an external magnetic field is introduced by the minimal coupling $\hat{p} \rightarrow \hat{p} + e\hat{A}$, where $e = |e|$ is the electron

Figure 3.1: Parabolic dispersion of a Schrödinger electron.
charge and $\hat{A}$ is the vector potential. We apply a magnetic field along the $z$-axis, for which the vector potential is given as

$$\hat{A} = (0, B \hat{x}, 0), \quad \mathbf{B} = \nabla \times \hat{A} = (0, 0, B).$$ (3.2)

The choice of $\hat{A}$ is not unique and Eq. (3.2) is referred to as the Landau gauge. For electrons in a lattice, we have to take the lattice periodic potential into account $\hat{H}_{\text{lattice}} = (\hat{p} + e\hat{A})^2/(2m_e) + V_{\text{lattice}}$. This is not trivial, however, and instead we use the Peierls’ substitution $k \rightarrow \hat{p} + e\hat{A}$ [56], which is a good approximation. For a parabolic band electron, the Hamiltonian with Peierls’ substitution in the Landau gauge reads

$$\hat{H} = \frac{1}{2m} \left( \hat{p} + e\hat{A} \right)^2 = \frac{1}{2m} \left[ \hat{p}_x^2 + (\hat{p}_y + eB\hat{x})^2 + \hat{p}_z^2 \right],$$ (3.3)

where $m$ is the effective mass. The justification of the Peierls’ substitution is difficult [57]. Early attempts to justify it were undertaken by Kohn [58] and Luttinger [59]. It is a an assumption used for decades, whose results are consistent with experimental data.

To solve the Hamiltonian of Eq. (3.3), we substitute

$$\hat{w} = \frac{\hat{x} - \hat{x}_0}{l_B},$$ (3.4)

where $l_B = \sqrt{\hbar/(eB)}$ is the magnetic length and $\hat{x}_0 = l_B^2 \hat{p}_y/\hbar$. With this, we can rewrite the Hamiltonian to

$$H = \left[ \frac{\hbar eB}{2m} \left( -\partial_w^2 + w^2 \right) + \frac{\hbar^2 k_z^2}{2m} \right].$$ (3.5)

Now the Hamiltonian closely resembles the equation for the quantum harmonic oscillator

$$-\frac{1}{2} \partial_w^2 \varphi_n + \frac{1}{2} w^2 \varphi_n = \left( n + \frac{1}{2} \right) \varphi_n,$$ (3.6)
CHAPTER 3. LANDAU QUANTIZATION

whose solutions we know. The wavefunctions $\varphi_n$ of the harmonic oscillator are given by

$$\varphi_n = \frac{1}{\sqrt{2^n n!}} \pi^{-\frac{1}{4}} e^{-\frac{w^2}{2}} H_n(w),$$

(3.7)

where the $H_n(w)$ are the Hermite polynomials

$$H_n(w) = (-1)^n e^{w^2} \frac{d^n}{dw^n} e^{-w^2}.$$

(3.8)

Then, the corresponding Schrödinger equation to Eq. (3.3) is solved by

$$\phi_{n,k_z}(r) = \sqrt{\frac{1}{2^n n! \sqrt{l_B}}} e^{-\frac{(x-x_0)^2}{2l_B^2}} H_n \left( \frac{x-x_0}{l_B} \right) \psi_{k_y,k_z}(y,z).$$

(3.9)

where $\psi_{k_y,k_z}(y,z) = u(y,z)e^{ik_y y+ik_z z}/\sqrt{L_y L_z}$ is a Bloch function for two dimensions $k_y$ and $k_z$, with $u(y,z)$ being a lattice-periodic function and with the lengths of the system along the $y$ and $z$ directions given by $L_{y,z}$. The Hamiltonian in Eq. (3.3) then produces the following eigenenergies

$$E_{n,k_z} = \hbar \omega_c \left( n + \frac{1}{2} \right) + \frac{1}{2m} \hbar^2 k_z^2,$$

(3.10)

where $\omega_c = eB/m$ is the cyclotron frequency.

An example of the probability density $|\phi_n(x)|^2$ is given in Fig. 3.2. The associated energy spectrum as a function of $k_z$ is plotted in Fig. 3.3. Along $k_z$, the energy spectrum corresponds to a series of parabolic levels, equidistantly spaced in units of $\hbar \omega_c$. Thus, the distance between Landau-levels scales linear with respect to $B$. The $B$-dependence of the Landau-level shifts is plotted on the right of Fig. 3.3. In the case of a fermion with parabolic dispersion, the lowest Landau level is displaced by $\frac{1}{2} \hbar \omega_c$ from $E = 0$, which is the zero-point energy.
3.1. SCHRÖDINGER ELECTRONS

The Hamiltonian and its eigenfunctions can be expressed conveniently by using the following dimensionless ladder operators

$$\hat{a}^\dagger = \frac{l_B}{\sqrt{2\hbar}}[\hat{p}_x + i(\hat{p}_y + eB\hat{x})]$$  \hspace{1cm} (3.11)

$$\hat{a} = \frac{l_B}{\sqrt{2\hbar}}[\hat{p}_x - i(\hat{p}_y + eB\hat{x})]$$  \hspace{1cm} (3.12)

The Hamiltonian then takes the form

$$\hat{H} = \hbar\omega_c \left(\hat{a}^\dagger \hat{a} + \frac{1}{2}\right) + \frac{\hat{p}_z^2}{2m}.$$  \hspace{1cm} (3.13)

The ladder operators fulfill the commutation relation

$$[\hat{a}, \hat{a}^\dagger] = 1.$$  \hspace{1cm} (3.14)

For simplicity we adopt the notation $|\phi_{n,k_z}\rangle = |n\rangle$ and summarize the action of the ladder operators below

$$\hat{a}^\dagger \hat{a} |n\rangle = \hat{n} |n\rangle = n |n\rangle,$$  \hspace{1cm} (3.15)

$$\hat{a}^\dagger |n\rangle = \sqrt{n + 1} |n + 1\rangle,$$  \hspace{1cm} (3.16)

$$\hat{a} |n\rangle = \sqrt{n} |n - 1\rangle.$$  \hspace{1cm} (3.17)

Figure 3.2: The probability density $|\phi_n(x)|^2$ for $n = 8$, $B = 10$ T and $m = m_e$ the free electron mass.
Figure 3.3: The Landau-level energies as a function of $k_z$ (left) with $m = m_e$, the free electron mass and $B = 100$ T. The energy spectrum of Landau levels for $k_z = 0$ as a function of magnetic field $B$ (right), with $m = m_e$.

In the number basis, we can then express the quantized Hamiltonian as a matrix. We write an arbitrary state $|\Psi\rangle$ as

$$|\Psi\rangle = \sum_n c_n |n\rangle. \quad (3.18)$$

Then the Schrödinger equation reads

$$\hat{H} |\Psi\rangle = \begin{pmatrix} \cdots & \hbar \omega_c (n - 1 + \frac{1}{2}) + \frac{\hbar^2 k_z^2}{2m} & \hbar \omega_c (n + \frac{1}{2}) + \frac{\hbar^2 k_z^2}{2m} & \cdots \\ \hbar \omega_c (n - 1 + \frac{1}{2}) + \frac{\hbar^2 k_z^2}{2m} & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \end{pmatrix} \begin{pmatrix} \cdots \\ c_{n-1} \\ c_n \\ \cdots \end{pmatrix}.$$

In this representation, the Hamiltonian for the Landau-quantized parabolic band is a diagonal matrix. However, as we will see for
3.2. WEYL ELECTRONS

the Weyl electrons, the Hamiltonian in the number basis is not always diagonal, though now we can use numerical methods to solve it.

3.2 Weyl Electrons

In light of our interest in Weyl semimetals, in this section we discuss a massless Weyl electron in an external magnetic field. We make use of the massless Dirac equation

\[ \hat{H} = v \alpha \hat{p}, \]  

(3.19)

with effective velocity \( v \), the matrices \( \alpha = (\alpha_x, \alpha_y, \alpha_z) \) where \( \alpha_i = \sigma_z \otimes \sigma_i \) with \( i \in \{x, y, z\} \), and \( \beta = \sigma_x \otimes 1 \) in the Weyl representation. The energy spectrum \( E(k) \) is a linear Dirac cone as shown for the \( k_x-k_y \)-plane in Fig. 3.4. Written in matrix form

\[ \hat{H} |\Psi\rangle = \left( \begin{array}{cc} \hat{H}^{R}_{\text{Weyl}} & 0 \\ 0 & \hat{H}^{L}_{\text{Weyl}} \end{array} \right) \left( \begin{array}{c} |\psi^{R}\rangle \\ |\psi^{L}\rangle \end{array} \right) = E |\Psi\rangle, \]  

(3.20)

we see that the Hamiltonian consists of two Weyl Hamiltonians \( \hat{H}^{R,L}_{\text{Weyl}} = \pm v \sigma \hat{p} \) placed on the diagonal. The spinor \( |\Psi\rangle = (|\psi^{R}\rangle, |\psi^{L}\rangle) \) separates into two chiral subspaces \( |\psi^{R}\rangle \) and \( |\psi^{L}\rangle \), which the massless Dirac equation does not mix. These are the Weyl fermions with right- and left-handed chirality, which refers to their parallel or antiparallel alignment of the spin and momentum vectors. We apply the Peierls’ substitution in the Landau gauge and obtain

\[ (E - v \sigma (\hat{p} + e \hat{A})) |\psi^{R}\rangle = 0. \]  

(3.21)
for the right-handed case. To obtain the solution to $\psi_R$, we consider the auxiliary equation

$$0 = [E - v\sigma(\hat{p} + e\hat{A})][E + c\sigma(\hat{p} + e\hat{A})] |\phi^R\rangle$$

$$= \left( E^2 - \hat{H}_{\text{aux}} \right) |\phi^R\rangle,$$

with

$$\hat{H}_{\text{aux}} = [v\sigma(\hat{p} + e\hat{A})]^2$$

$$|\psi^R\rangle = (E + v\sigma(\hat{p} + e\hat{A})) |\phi^R\rangle.\quad (3.25)$$

Note that $\hat{H}_{\text{aux}} = (\hat{H}^{R}_{\text{Weyl}})^2 = (\hat{H}^{L}_{\text{Weyl}})^2$. Finding the eigenenergies to $\hat{H}_{\text{aux}}$ yields the squared energies to both $|\psi^R\rangle$ and $|\psi^L\rangle$. The auxiliary Hamiltonian is quadratic in $\hat{p}$. Expanding Eq. (3.23)

$$E^2 |\phi^R\rangle = v^2 [\hat{p}_x^2 + (\hat{p}_y + eB\hat{x})^2 + \hat{p}_z^2 + \hbar eB \sigma_z] |\phi^R\rangle,\quad (3.26)$$

we notice that it is now similar to the massive free electron case, Eq. (3.3), except for the replaced mass $1/2m \rightarrow v^2$ and the additional term $\hbar eB \sigma_z$. We use the same procedure employed for Eq. (3.3) and arrive at

$$(H_{\text{aux}} - \hbar eB \sigma_z) \phi_{n,m_s,k_z}(r) = [v^2 \hbar eB (-\partial_w^2 + w^2) + v^2 \hbar^2 k_z^2] \phi_{n,m_s,k_z}(r).$$

We now make an ansatz similar to Eq. (3.9)

$$\phi_{n,m_s,k_z}(r) = \sqrt{\frac{1}{2^n n! \sqrt{l_B}}} \frac{\pi^{-\frac{1}{4}}}{2^{l_B}} e^{-\frac{(x-x_0)^2}{2l_B^2}} H_n \left( \frac{x-x_0}{l_B} \right) \chi_{m_s} \psi_{k_y,k_z}, \quad (3.27)$$

though now containing spinors $\chi_{m_s}$, which are the eigenvectors of the $\sigma_z$-matrix

$$\chi_{m_s = \frac{1}{2}} = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad \chi_{m_s = -\frac{1}{2}} = \begin{pmatrix} 0 \\ 1 \end{pmatrix}.\quad (3.28)$$
3.2. WEYL ELECTRONS

Figure 3.5: The probability density $|\psi_{n,m_s,k_z}^R|^2$ for $n = 8$, $m_s = -1/2$, $v = 3 \times 10^8 \text{ m/s}$, and $B = 10 \text{ T}$.

where $m_s = \pm 1/2$ is the spin magnetic quantum number. We find

$$H_{\text{aux}} \phi_{n,m_s,k_z}(x,y,z) = E^2 \phi_{n,m_s,k_z}(x,y,z),$$  \hspace{1cm} (3.29)

$$E^2 = \hbar^2 \omega_D^2 \left( n + \frac{1}{2} + m_s \right) + v^2 \hbar^2 k_z^2,$$  \hspace{1cm} (3.30)

and a new cyclotron frequency

$$\omega_D^2 = 2v^2 eB/\hbar.$$  \hspace{1cm} (3.31)

Then we find the eigenenergies

$$E = \pm \sqrt{\hbar^2 \omega_D^2 (n + \frac{1}{2} + m_s) + v^2 \hbar^2 k_z^2}.$$  \hspace{1cm} (3.32)
Figure 3.6: (a) Landau levels for the left-handed Weyl cone and (b) the right-handed Weyl cone on the right plotted with respect to $k_z$ for $B = 20$ T and $v = 5 \cdot 10^5$ m/s.

The wavefunctions $\psi^R$ and $\psi^L$ can be found by evaluating

$$\psi_{n,m_s,k_z}^R = (E + v\sigma(\hat{p} + e\hat{A}))\phi_{n,m_s,k_z},$$

$$\psi_{n,m_s,k_z}^L = (E - v\sigma(\hat{p} + e\hat{A}))\phi_{n,m_s,k_z}.$$  \hspace{1cm} (3.33)

$$\psi_{n,m_s,k_z}^L = (E - v\sigma(\hat{p} + e\hat{A}))\phi_{n,m_s,k_z}.$$  \hspace{1cm} (3.34)

The calculation is provided in Appendix A. Two special solutions, the “0th” Landau levels, $\psi_{0,-\frac{1}{2},k_z}^R = 0$ with $E = \nu \hbar k_z$ and $\psi_{0,-\frac{1}{2},k_z}^L = 0$ with $E = -\nu \hbar k_z$ are of particular interest, since they remain linear in $k_z$ and chiral [23]. Each Weyl cone hosts a non-degenerate chiral Landau level with opposite dispersion and opposite chirality. The Landau level dispersion with respect to $k_z$ is plotted in Fig. 3.6 for the two Weyl Hamiltonians. Note that the Landau level spectrum of the Dirac Hamiltonian in Eq. (3.20) is the superposition of these two Weyl spectra.
The Landau level energies for the Weyl cones at \( k_z = 0 \) have a characteristic \( \sqrt{B} \)-dependence, which is plotted in Fig. 3.7. This makes them easily differentiable from the Landau levels of the massive parabolic bands, a property that we can exploit in Landau level spectroscopy. Note, that unlike the parabolic case, the lowest Landau level for the Weyl or Dirac cones does not scale with \( B \) at all and when plotted over \( k_z \) it retains its linear dispersion.

In the following, we want to also express the Weyl Hamiltonian in the language of ladder operators, which are given by Eqs. (3.11) and (3.12). In the right-handed case we obtain

\[
\hat{H}_{\text{Weyl}}^R = v(\hat{p} + e\hat{A})\sigma = \begin{pmatrix} v\hat{p}_z & \hbar\omega_D a \\ \hbar\omega_D a^\dagger & -v\hat{p}_z \end{pmatrix}.
\]  

(3.35)

Compared to the parabolic case in the previous section, the Hamiltonian is not diagonal in the number basis for the case of the Weyl fermion. With Eqs. (3.16) and (3.17), the ladder operators take the form

\[
\hat{a} = \begin{pmatrix} 0 & \sqrt{1} \\ \sqrt{2} & 0 \\ \sqrt{3} & \ddots \end{pmatrix} \quad \text{and} \quad \hat{a}^\dagger = \begin{pmatrix} 0 & \sqrt{1} \\ 0 & \sqrt{2} \\ \ddots \end{pmatrix}.
\]  

(3.36)

Note that this matrix representation is technically only correct for infinite matrix sizes, taking into account all possible \( n \in [0, \infty) \). Thus, if we solve the corresponding number-basis Hamiltonian numerically,
we have to truncate its matrix to a finite size, which produces small artifacts that can be filtered out.

### 3.2.1 Chiral Anomaly

In particle physics Weyl fermions are associated with the chiral anomaly. The application of a parallel electric field $\mathbf{E}$ and a magnetic field $\mathbf{B}$, creates a chiral current that can transform fermions of one chirality into its opposite, creating a chiral imbalance [8, 9]. An analogous effect exists in Weyl semimetals, as shown by H. B. Nielsen and M. Ninomiya [23]. In the previous section, we saw that the Landau quantization of the Weyl cones gives rise to a single linearly dispersing, chiral Landau level per cone. Next, we consider what happens if an electric field is applied along the $k_z$-direction. For the right-handed Landau level with a positive slope, the electrons climb to higher energies, filling the band, whereas the left-handed band is emptied, as is depicted in Fig. 3.8. This process creates a chiral imbalance between the cones: an effective chiral anomaly. To derive the chiral current we require the number of states in each Landau level. Without an applied magnetic field, we know that the number of states for linear bands in the $k_x$-$k_y$ plane is

$$N_{xy} = \pi k^2 \frac{4\pi^2}{L_x L_y} = \frac{\pi E^2}{\hbar^2 v^2} \frac{4\pi^2}{L_x L_y},$$

(3.37)

where $k = |\mathbf{k}|$ and $E = \hbar v k$. Under the application of a magnetic field along $z$, the electrons in the $k_x$-$k_y$-plane condense into Landau levels, whose energies are given by $E^2 = \hbar^2 \omega_D^2 (n + \frac{1}{2} + m_s)$. Inserting the energy into Eq. (3.37), we obtain the number of states per
Landau level and unit area

\[ \rho_{\text{LL}} = \frac{\partial_n N}{L_x L_y} = \frac{\pi \omega_D^2}{c^2} = \frac{eB}{\hbar}. \]  

(3.38)

Alternatively, we can consider the shift \( x_0 = l_B^2 k_y \), which is present in the Landau quantized wave functions, see Eq. (3.27). The crystal momentum takes quantized values \( k_y = \frac{2\pi n_y}{L_y} \). In the Landau gauge, the electrons thus condense into Landau stripes with the distance \( \Delta x_0 = \frac{2\pi l_B}{L_y} \), as sketched in Fig. 3.9. The number of states along the length of \( L_x \) is \( N_{xy} = L_x / \Delta x_0 \) and thus we find the same equation

\[ \rho_{\text{LL}} = \frac{1}{L_x L_y} \frac{L_x}{\Delta x_0} = \frac{eB}{\hbar}. \]  

(3.39)

For the \( k_z \)-direction, the number of states of the right-hand chiral Landau level is \( N_z^R = k_z L_z \frac{L_z}{2\pi} \). To drive the current, we apply an electric field \( \mathbf{E} = (0, 0, E) \) along the \( z \)-direction and use the Peierls’ substitution \( \hbar k_z \rightarrow \hat{p}_z - eEt \). It follows that the current density is

\[ j_z^R = \partial_t \frac{1}{L_z} N_z^R = -\frac{eE}{\hbar}. \]  

(3.40)

for the right-hand chiral carrier density along the \( z \)-direction. Multiplied with \( \rho_{\text{LL}} \), taking the electrons condensed into the Landau level into account, we obtain the total current

\[ j^R = j_z^R \rho_{\text{LL}} = -\frac{e^2}{\hbar^2} EB. \]  

(3.41)

For the left-hand chiral fermions, it follows that \( \rho^L = -\rho^R \) and the
imbalance in the chiral fermion density is
\[ j_{\text{chiral}} = \partial_t \frac{N^L - N^R}{V} = \frac{2e^2}{\hbar^2} eB. \] (3.42)

In practice, this effect could be measured in terms of a negative longitudinal magneto-resistance (NLMR). This was reported for example, for GdPtBi in multiple publications [60–62], making it attractive as a Weyl-semimetal candidate for Landau-level spectroscopy.

### 3.3 Massive Dirac Electrons

Finally, we want to take a look at a Dirac electron with finite mass. For massive particles in the Weyl representation, the Hamiltonian is given by
\[ \hat{H} = \begin{pmatrix} \hat{H}^R_{\text{Weyl}} & mv^2 \\ mv^2 & \hat{H}^L_{\text{Weyl}} \end{pmatrix}. \] (3.43)

We square the Hamiltonian, keeping in mind that \( \hat{H}^R_{\text{Weyl}} = -\hat{H}^L_{\text{Weyl}} \), and obtain
\[ \hat{H}^2 = \begin{pmatrix} m^2v^4 + \left(\hat{H}^R_{\text{Weyl}}\right)^2 & 0 \\ 0 & m^2v^4 + \left(\hat{H}^L_{\text{Weyl}}\right)^2 \end{pmatrix}. \] (3.44)

Following the procedure of Section 3.2, we obtain the Landau level energies for the massive Dirac Hamiltonian as
\[ E_n = \pm \sqrt{2v^2\hbar eB(n + \frac{1}{2} + m_s) + v^2 k_z^2 + m^2v^4}. \] (3.45)

The chiral subspaces \( \hat{H}^{R,L}_{\text{Weyl}} \) are coupled by the off-diagonal mass terms producing a gap in the Dirac cone as shown in Fig. 3.10 (a)
3.3. MASSIVE DIRAC ELECTRONS

Figure 3.10: (a) Gapped Dirac cone in arbitrary units. (b) Landau levels along $k_z$ for $v = 4 \times 10^5$ m/s and $m = m_e/20$. (c) Landau levels at $k_z = 0$ for $v = 4 \times 10^5$ m/s and $m = m_e/20$ with respect to $B$.

in the $k_x$-$k_y$-plane. The corresponding Landau level spectrum is plotted in Fig. 3.10 (b). For large masses $m$, the hyperbolic profile of the gapped Dirac cones in the fieldless case asymptotically approach the parabolic dispersion of the Schrödinger electron. Nonetheless, unlike the parabolic case, the lowest Landau level of the massive Dirac electron does not scale with the magnetic field and there is no field-dependent zero-point energy. As the mass gap increases, the separated Landau level fans of Fig. 3.10 (c) approach linear dispersions. Using the Taylor series $\sqrt{A} + v \approx \sqrt{A} + v/(2\sqrt{A}) +$
\[ O(v^2), \] we find that for large masses

\[
E_{n,m_s,k_z} = \pm \sqrt{2v^2 \hbar eB(n + \frac{1}{2} + m_s) + v^2 k_z^2 + m^2 v^4} \tag{3.46}
\]

\[
\approx \pm mv^2 \pm vk_z \pm \hbar \omega_c \left( n + \frac{1}{2} + m_s \right) + \ldots \tag{3.47}
\]

We find that all bands can be described by

\[
E_{n,m_s,k_z} \approx \pm mc^2 \pm \hbar \omega_c n' + \ldots \tag{3.48}
\]

\[
E_{n,m_s,k_z} \propto \pm \hbar \omega_c n' \tag{3.49}
\]

where \( n' = (n + \frac{1}{2} + m_s) \in \mathbb{Z} \), keeping in mind that the \( n' = 0 \) levels are non-degenerate, while \( |n'| > 1 \) levels are doubly degenerate. This is different from the massive parabolic case, which produces Landau levels of the form

\[
E_{n,k_z} = \hbar \omega_c \left( n + \frac{1}{2} \right), \tag{3.50}
\]

see Eq. (3.10). The lowest energy state of a Dirac electron is not offset by a zero-point energy like its Schrödinger equivalent, due to the extra term \( m_s \), which correspond to the Zeeman-splitting in the free-electron case. This leads to constant-in-\( B \) Landau-levels, as can be seen in Fig. 3.10 (c).

In summary, in an external magnetic field, the quasifree electrons of condensed matter systems are bound to quantized orbits, the Landau levels. These have characteristic energy spectra that can be related to the dispersion relation of the electrons. This allows us to differentiate between sections of the band structure that are described by massive or massless carriers. In the next chapter, we will investigate the optical response of a material using the Kubo formalism.
4.1 Susceptibility Functions

In this chapter, we derive the optical response of electrons subjected to an external magnetic field. Using perturbation theory, we obtain the Kubo formula \([63, 64]\) for the (optical) conductivity tensor \(\sigma_{\alpha,\beta}\). The unperturbed system is described by a Hamiltonian \(H_0\). We apply a time-dependent perturbation \(\Delta \hat{H}(t)\),

\[
\hat{H}(t) = \hat{H}_0 + \Theta(t - t_0)\Delta \hat{H}(t).
\]  

(4.1)

Here, \(\Theta\) is the Heaviside function, which turns on the perturbation at time \(t = t_0\). First order perturbation theory yields the eigenvalue of an observable \(\hat{F}\)

\[
\langle \hat{F} \rangle(t) = \langle \hat{F}(t) \rangle_0 + \langle \Delta \hat{F} \rangle(t) \approx \langle \hat{F}(t) \rangle_0 - \frac{i}{\hbar} \int_{t_0}^t dt' \langle [\hat{F}(t'), \Delta \hat{H}(t')] \rangle_0 ,
\]

(4.2)

where \(\langle \rangle_0\) denotes the expectation value taken with the undisturbed eigenvectors of \(\hat{H}(t)\) at \(t = t_0\). The probability distribution for the occupation of eigenstates of the system is described via the density matrix operator

\[
\hat{\rho} = \frac{1}{Z} e^{-\beta(\hat{H} - \mu \hat{N})}.
\]  

(4.3)
4.1. SUSCEPTIBILITY FUNCTIONS

The constant $\beta = 1/k_B T$, the occupation operator is denoted as $\hat{N}$ and $\mu$ is the chemical potential. For the temperature $T = 0$, the chemical potential $\mu$ coincides with the Fermi energy $E_F$. The partition function $Z$ is

$$Z = \text{Tr}(e^{-\beta(\hat{H} - \mu \hat{N})}). \quad (4.4)$$

The expectation value of an operator $\hat{F}$ is obtained by the trace [65],

$$\langle \hat{F} \rangle = \text{Tr}(\hat{F}) = \frac{1}{Z} \sum_n \langle n | e^{-\beta(\hat{H}_0 - \mu \hat{N})} \hat{F} | n \rangle. \quad (4.5)$$

where the sum $\sum_n$ sums over all energy eigenstates in our many-body ensemble. We get the familiar Fermi-Dirac distribution by calculating the expectation value of $\hat{N}_n$, the occupation number operator associated with the energy level $E_n$

$$f(E_n) = f_n = \langle \hat{N}_n \rangle = \frac{1}{e^{\beta(E_n - \mu)} + 1}. \quad (4.6)$$

We now define the retarded correlation function

$$\chi_{F, \Delta H}(t, t') = -\Theta(t' - t_0) \frac{i \hbar}{\hbar} \langle [\hat{F}(t), \Delta \hat{H}(t')] \rangle_0, \quad (4.7)$$

and express the expectation value $\langle \Delta \hat{F} \rangle(t)$ as

$$\langle \Delta \hat{F} \rangle(t) = \int_{-\infty}^t dt' \chi_{F, \Delta H}(t, t'). \quad (4.8)$$

It is often convenient to define $\chi_{F, \Delta H}$ in terms of other, more familiar operators. We rewrite the perturbation as

$$\Delta \hat{H} = \hat{G}(t)g(t), \quad (4.9)$$

where $\hat{G}(t)$ is an operator and $g(t)$ is a scalar time-dependent function. Then, we can define a $\chi_{F, G}$

$$\chi_{F, G}(t, t') = -\Theta(t' - t_0) \frac{i \hbar}{\hbar} \langle [\hat{F}(t), \hat{G}(t')] \rangle_0, \quad (4.10)$$
and instead of Eq. (4.8), \( \langle \Delta \hat{F} \rangle (t) \) is then given by the equation

\[
\langle \Delta \hat{F} \rangle = \int_{-\infty}^{t} dt' \chi_{F,G}(t, t')g(t').
\]

(4.11)

A special case is \( \hat{G} = \hat{F} \), meaning that the quantity that we observe, \( \hat{F} \), is proportional to the perturbation \( \Delta \hat{H} = \Delta \hat{F}g(t) \). Then \( \chi_{F,F}(\omega) \) is an auto-correlation function. If the perturbations are applied fields, we know the correlation functions \( \chi_{F,G} \) also as susceptibility functions. A familiar example is that of an external electric field \( \mathbf{E}_{\text{ext}} \) applied to a charge density, which induces a field \( \mathbf{E}_{\text{ind}} \), such that the total field is

\[
\mathbf{E}_{\text{tot}} = \mathbf{E}_{\text{ext}} + \mathbf{E}_{\text{ind}}.
\]

(4.12)

We know that an electric field \( \mathbf{E} \) is related to an electrostatic potential \( \phi \) via \( \mathbf{E} = -\nabla \phi \). The Hamiltonian associated with a charge density \( \hat{\rho}_e \) of electrons in an electric field is given by

\[
\hat{H}(t) = \int dr \hat{\rho}_e(r, t)\phi(r).
\]

(4.13)

To find the induced field \( \mathbf{E}_{\text{ind}} \), we require the induced charge density \( \rho_{\text{ind}} \). Then, we obtain the electric field via Poisson’s equation \( \nabla^2 \phi_{\text{ind}} = -\rho_{\text{ind}}/\varepsilon_0 \), where \( \varepsilon_0 \) is the vacuum permeability, and \( \mathbf{E}_{\text{ind}} = -\nabla \phi_{\text{ind}} \). We identify \( \Delta \hat{\rho}_e = \hat{\rho}_{\text{ind}} \) and calculate

\[
\langle \hat{\rho}_{\text{ind}} \rangle \approx -\frac{i}{\hbar} \int_{t_0}^{t} dt' \langle [\hat{\rho}_e(r, t), \Delta \hat{H}(t')] \rangle
\]

(4.14)

\[
= -\frac{i}{\hbar} \int_{t_0}^{t} dt' \int d\mathbf{r}' \langle [\hat{\rho}_e(r, t), \hat{\rho}_e(\mathbf{r}', t')\phi(\mathbf{r}')] \rangle
\]

(4.15)

\[
= \int_{-\infty}^{t} dt' \int d\mathbf{r}' \chi_{\rho_e,\rho_e}(\mathbf{r}, \mathbf{r}', t, t')\phi(\mathbf{r}')
\]

(4.16)

with \( \chi_{\rho_e,\rho_e}(\mathbf{r}, \mathbf{r}', t, t') = -\Theta(t - t_0)\frac{i}{\hbar} \langle [\hat{\rho}_e(r, t), \hat{\rho}_e(\mathbf{r}', t')] \rangle_0 \). Here, \( \chi_{\rho_e,\rho_e} \) represents the charge-charge correlation function or the electric susceptibility, which we refer to as the electric permeability. We
simplify the problem to be isotropic and time-independent. Then, by multiplying Eq. (4.12) with \( \varepsilon_0 \) and abbreviating \( \chi_e = \chi_{\rho_e, \rho_e} \), we identify the familiar relation

\[
D = \varepsilon_0 E + P = \varepsilon_0 (1 + \chi_e) E, \quad \text{where} \quad P = \varepsilon_0 \chi_e E.
\]

(4.17)

relating the displacement field \( D = \varepsilon_0 E_{\text{tot}} \), the polarization field \( P = \varepsilon_0 E_{\text{ind}} \), and the electric field \( E \) to another.

In the same manner we can relate the magnetic induction \( B \) in a material to the external magnetic field \( H \) and the magnetization \( M \)

\[
B = \mu_0 (H + M) = \mu_0 (1 + \chi_m) H, \quad \text{where} \quad M = \chi_m H,
\]

(4.18)

with the magnetic susceptibility \( \chi_m = \chi_{\rho_m, \rho_m} \). Here, \( \rho_m (r) \) is the density of magnetic moments and \( \mu_0 \) is the vacuum permeability. For a system of \( S = 1/2 \) spins, where the spins and the magnetic field \( H \) are aligned along \( z \), the perturbation reads

\[
\Delta \hat{H} = \sum_s \int d^3 r \, \hat{\rho}_m (r) H(r).
\]

(4.19)

Then the general form of the susceptibility is

\[
\chi_m (r, r', t, t') = -\frac{i}{\hbar} \Theta (t - t') \langle [\hat{\rho}_m (r, t), \hat{\rho}_m (r', t')] \rangle_0
\]

(4.20)

with the density of magnetic moments

\[
\hat{\rho}_m = -\mu_B (\psi_\uparrow^\dagger (r, t) \psi_\uparrow (r, t) - \psi_\downarrow^\dagger (r, t) \psi_\downarrow (r, t))
\]

(4.21)

where \( \psi_{\uparrow, \downarrow} (r, t) \) are the field operators of the spins and \( \mu_B \) is the Bohr magneton.

### 4.2 Conductivity Tensor

In infrared spectroscopy, we are interested in one particular susceptibility function, which is the optical conductivity. It relates the
current to an external electric field

$$\mathbf{j} = \sigma \mathbf{E}. \quad (4.22)$$

The electronic current density is

$$\mathbf{j} = -\rho \mathbf{v}, \quad (4.23)$$

with the electron velocity $\mathbf{v}$. In the following we are interested in electrons on a lattice, thus consider the Bloch electron eigenstates $|n, \mathbf{k}\rangle$, where $n$ is the band index and $\mathbf{k}$ is the wave vector. To obtain an expression for the conductivity, we first consider a single electron and introduce a perturbation with an electric field using Peierls’ substitution

$$\hat{H} = \frac{1}{2m} \left[ \hat{\mathbf{p}} + e \hat{\mathbf{A}}(t) \right]^2. \quad (4.24)$$

The vector potential $\hat{\mathbf{A}}$ is

$$\hat{\mathbf{A}} = \hat{\mathbf{A}}_0 + \Delta \hat{\mathbf{A}}(t), \quad (4.25)$$

where $\hat{\mathbf{A}}_0$ is constant and $\Delta \hat{\mathbf{A}}(t)$ is a time-dependent perturbation. Note that we assume no $\mathbf{r}$-dependence and thus the associated electric field $\mathbf{E}(t) = \partial_t \Delta \mathbf{A}$ is only time dependent and spatially uniform. Later on, we will introduce a static magnetic field and Landau quantize the Hamiltonian via the term $\hat{\mathbf{A}}_0$. For now though, we neglect the term, setting $\hat{\mathbf{A}}_0 = 0$, such that

$$\hat{\mathbf{A}} = \Delta \hat{\mathbf{A}}(t). \quad (4.26)$$

Expanding the Hamiltonian and keeping terms only up to first order in $\hat{\mathbf{A}}(t)$, we arrive at

$$\hat{H} \approx \frac{1}{2m} \hat{\mathbf{p}}^2 + \frac{e}{m} \hat{\mathbf{p}} \hat{\mathbf{A}} = \frac{1}{2m} \hat{\mathbf{p}}^2 + e\hat{\mathbf{v}} \hat{\mathbf{A}} = \hat{H}_0 + \Delta \hat{H} \quad (4.27)$$

with $\Delta \hat{H} = \hat{\mathbf{J}} \hat{\mathbf{A}}$, where we define $\hat{\mathbf{J}} = e\hat{\mathbf{v}}$ and we use that $\hat{\mathbf{A}}$ commutes with $\hat{\mathbf{p}}$ in the Coulomb gauge ($\nabla \hat{\mathbf{A}} = 0$). The expectation
value of the current $\hat{J}$ is then given by

$$\langle \hat{J} \rangle = \langle \hat{J} \rangle_0 + \int_{-\infty}^{\infty} dt \chi_{J,J}(t,t') \hat{A}(t'),$$  \hspace{1cm} (4.28)

see Eq. (4.2). The associated susceptibility tensor $\chi_{J,J}$ is the current-current correlation function, whose entries are given by

$$\chi_{J,\alpha,J,\beta}(t,t') = -\frac{i}{\hbar} \Theta(t-t_0) \langle [\hat{J}_\alpha(t),\hat{J}_\beta(t')] \rangle_0,$$  \hspace{1cm} (4.29)

with $\alpha, \beta \in \{x,y,z\}$. The first term in Eq. (4.28) reads

$$\langle \hat{J}_\alpha \rangle_0 = \frac{e^2}{m} \hat{A}_\alpha$$  \hspace{1cm} (4.30)

where we assume no initial current to be present before turning on the perturbation. In the case of an oscillating field, $\hat{E}_\alpha \propto e^{i\omega t}$, the relation $E_\alpha = -\partial_t \hat{A}_\alpha = i\omega \hat{A}_\alpha$ holds and we can express $\chi$ in terms of the electric field. In the following, we switch to the frequency space by applying the Fourier transform

$$h(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} H(\omega) e^{-i\omega t} d\omega,$$  \hspace{1cm} (4.31)

$$H(\omega) = \int_{-\infty}^{\infty} h(t) e^{i\omega t} dt,$$  \hspace{1cm} (4.32)

where $h(t)$ and $H(\omega)$ are some arbitrary functions related by the Fourier transform. For now, we assert that the expectation value of $\Delta\hat{F}$ then takes the form

$$\langle \Delta\hat{F} \rangle (\omega) = \chi_{F,G}(\omega)g(\omega)$$  \hspace{1cm} (4.33)

in frequency space. With this, we obtain the current

$$\langle \hat{J}(\omega) \rangle = -i \frac{e^2}{m\omega} E(\omega) + \chi_{J,J}(\omega)E(\omega).$$  \hspace{1cm} (4.34)
The conductivity relates the electric field to the current density

\[
\langle \hat{j}(\omega) \rangle = \frac{1}{V} \langle \hat{J}(\omega) \rangle = \sigma(\omega)E(\omega).
\]  

(4.35)

So far, we had looked at the optical conductivity for a single electron. In the context of condensed matter materials we deal with many-body states. We have \(N\) electrons on the Fermi-surface that contribute to the conductivity and assume the electrons to be independent of one another. Summing the electrons in \(k\)-space over the Fermi surface, we arrive at [66, 67]

\[
\sigma_{\alpha,\beta}(\omega) = -i \frac{N e^2}{m \omega V} \delta_{\alpha,\beta} + i \frac{\omega V}{\omega} \sum_{k} \chi_{J_{\alpha},J_{\beta}}(k,\omega),
\]  

(4.36)

where the current operator \(J_{\alpha}\) is now \(k\)-dependent. We want to bring Eq. (4.36) into a more familiar form. To do so, we revise the Fourier transform. Using the Fourier transform of Eq. (4.31) and (4.32), a perturbation of the form \(A(t) \propto e^{i \omega_0 t}\) yields a response in the form of a delta-function centered around \(\omega_0\) in frequency space. Physical processes, however, are subject to relaxation. Thus, we introduce the quantity

\[
\omega^{\pm} = \omega \pm i \gamma,
\]  

(4.37)

and revise our Fourier transform into the following Laplace transform:

\[
\sigma_{\alpha,\beta}(\omega) \propto \frac{N e^2}{m \omega V} \delta_{\alpha,\beta} + \frac{i}{\omega V} \sum_{k} \chi_{J_{\alpha},J_{\beta}}(k,\omega),
\]

(4.36)
4.2. CONDUCTIVITY TENSOR

The quantities we transform contain Heaviside functions $\Theta(t - t_0)$. We choose $t_0 = 0$ without loss of generality and adjust the integration limits in the Fourier transform accordingly. The presence of $\omega^+$ introduces a relaxation process, as we can see by transforming an oscillating function $h(t) = e^{-i\omega_0 t}$, which yields

$$H(\omega^+) = \int_0^\infty h(t)e^{i\omega^+ t} dt. \quad (4.39)$$

This gives rise to the natural lineshape, described the Lorentzian function, as visualized in Fig. 4.1 with a scattering rate $\gamma$.

With this, Eq. (4.36) becomes

$$\sigma_{\alpha,\beta}(k, \omega^+) = -i \frac{\sigma_0}{\omega} \delta_{\alpha,\beta} + \frac{i}{\omega V} \sum_k \chi_{J_\alpha, J_\beta}(k, \omega^+). \quad (4.42)$$

with $\sigma_0 = \frac{N_f e^2}{mv}$, where $N_f$ is number of free electrons on the Fermi surface and $\tau = 1/\gamma$. The first term should not be confused with the Drude contribution. Rather, it is the so-called diamagnetic response \cite{68}. The second term is the paramagnetic response. An expression for $\chi(\omega^+)$ is derived in Appendix B. The diamagnetic part then cancels precisely with a part of the paramagnetic term. Thus, we

form

$$h(t) = \frac{1}{2\pi} \int_0^\infty H(\omega^+) e^{-i\omega^+ t} d\omega^+ \quad (4.38)$$

$$H(\omega^+) = \int_0^\infty h(t)e^{i\omega^+ t} dt. \quad (4.39)$$
obtain
\[
\sigma_{\alpha,\beta}(\omega^+) = -\frac{ie^2}{\hbar V} \sum_{n,m,k} \frac{f_{mn}}{\omega_{mn}} \frac{\langle n|\hat{v}_\alpha|m\rangle \langle m|\hat{v}_\beta|n\rangle}{\omega^+ - \omega_{mn}}.
\]
where \( f_{mn} = f_m - f_n = \) and \( \omega_{mn} = \omega_m - \omega_n \). Note that this conductivity contains both the interband \((n \neq m)\) and intraband contributions \((n = m)\). Taking the limit \( \omega_n - \omega_m \to 0 \) such that
\[
\frac{f_{mn}}{\omega_{mn}} \to \frac{\partial f_n}{\partial \omega_n},
\]
the intraband conductivity can be expressed as
\[
\sigma_{\text{intra}}^{\alpha,\beta} = -\frac{ie^2}{\omega^+} \frac{1}{V} \sum_{n,k} \frac{\partial f_n}{\partial \omega_n} v_{n,\alpha} v_{n,\beta},
\]
where the \( v_{n,\alpha/\beta} \) are the expectation values \( \langle n|\hat{v}_{\alpha/\beta}|n\rangle \) of the velocity operators. We note that even though we had started with a parabolic dispersion, the Kubo formula above is independent of the specific model and only requires the velocity operators to be known. It is often useful to express the optical conductivity for the circular polarizations of light, using the velocities \( \hat{v}_\pm = (\hat{v}_x \pm i\hat{v}_y)/\sqrt{2} \). For left-hand polarized light (+), we denote \( \sigma^{+,+} = \sigma_+ \), similarly for right-hand polarized light \( \sigma^{-,-} = \sigma_- \). The quantity relevant to the measurements in the experimental chapters, is the reflectivity \( R \). It can be obtained via the standard optical formulas for normal incidence \([69]\)
\[
R(\omega) = \frac{\left|\sqrt{\varepsilon(\omega)} - 1\right|^2}{\left|\sqrt{\varepsilon(\omega)} + 1\right|^2}, \quad \varepsilon(\omega) = 1 + \frac{i\sigma(\omega)}{\varepsilon_0 \omega}.
\]
To get an intuition for interband-optical conductivity, we want to discuss a few model examples in the remainder of this section. We
4.2. CONDUCTIVITY TENSOR

Figure 4.2: Depicted are three different parabolic models and their optical responses. For (a) and (d), the term $c\sigma_z$ only shifts the energies of the bands, for (b) and (e) the term $c\sigma_x$ gaps the parabolic bands near the cusps, for (c) and (f) the $k$-dependent part is not diagonal, mixing the electron and hole bands throughout $k$-space. The mass was chosen as $m = m_0/10$, the gap as $c = 50$ meV. To calculate the optical conductivity, the lattice parameters were chosen as $a_x = a_y = a_z = 6.68\,\text{Å}$.

Consider three parabolic Hamiltonians with different couplings, depicted in Fig. 4.2. The Hamiltonian of Fig. 4.2 (a)

$$H = \frac{1}{2m}\hbar^2k^2\sigma_z + c\sigma_z = \begin{pmatrix} \frac{1}{2m}\hbar^2k^2 + c & 0 \\ 0 & -\frac{1}{2m}\hbar^2k^2 - c \end{pmatrix}, \quad (4.47)$$

where $\sigma_i$ are the Pauli matrices with $i \in x, y, z$ and $c$ is a constant, evidently has eigenstates $\psi_+ = (1, 0)$ and $\psi_- = (0, 1)$. The eigenenergies are given by $E_\pm = E_0 \pm c$ where $E_0 = \hbar^2k^2/2m$. The velocities $v = i[H, r]$ are

$$v_x = i[H, x] = \frac{\hbar k_x}{m}\sigma_z, \quad v_y = \frac{\hbar k_y}{m}\sigma_z. \quad (4.48)$$
We see that for the diagonal Hamiltonian, the interband matrix
elements \langle \psi_+ | v_{x,y} | \psi_- \rangle vanish and thus no transitions occur as is
shown in Fig. 4.2 (d).

In the case of Fig. 4.2 (b), with the Hamiltonian

\begin{equation}
H = \frac{1}{2m} \hbar^2 k^2 \sigma_z + c \sigma_x = \left( \frac{1}{2m} \hbar^2 k^2 c \right) - \frac{1}{2m} \hbar^2 k^2 c \right),
\end{equation}

the eigenstates are given by \( \psi_+ = \frac{1}{\mathcal{N}_+} [-c/(E_0 - E_+), 1] \) and \( \psi_- = \frac{1}{\mathcal{N}_-} [(E_0 + E_-)/c, 1] \). Here \( \mathcal{N}_\pm \) are normalization constants and the
eigenenergies are \( E_\pm = \pm \sqrt{E_0^2 + c^2} \). The velocities are identical to
the previous case, however, the matrix elements \( \langle \psi_\pm | \hat{v}_\alpha | \psi_\mp \rangle \) now do
not vanish and we expect interband transitions near \( k = 0 \). Far
from \( k = 0 \) the eigenstates tend toward \( \psi_+(k \to \infty) = (1, 0) \) and
\( \psi_-(k \to \infty) = (0, 1) \), such that for \( k \to \infty \) the optical conductivity
vanishes again. This results in a peak in the vicinity of the interband
gap, as seen in Fig. 4.2 (e).

Finally, for the Hamiltonian of Fig. 4.2 (c), with \( k \)-dependent off-
diagonal terms

\begin{equation}
H = \frac{1}{2m} \hbar^2 (k_x^2 \sigma_x + k_y^2 \sigma_y + k_z^2 \sigma_z) + c \sigma_x
\end{equation}

\begin{equation}
= \left( \frac{\hbar^2 k_z^2}{2m} + c \right) \left( \frac{\hbar^2 k_x^2}{2m} (k_x^2 - ik_y^2) \right) - \frac{\hbar^2 k_z^2}{2m} c \right),
\end{equation}

and velocities

\begin{equation}
v_x = \frac{\hbar k_x}{m} \sigma_x, \quad v_y = \frac{\hbar k_y}{m} \sigma_y
\end{equation}

the transition matrix elements \( \langle \psi_+ | v_{x,y} | \psi_- \rangle \) are finite for all \( k_{x,y} \) also
far from \( k = 0 \). Then, the optical conductivity continuously grows
with energy and resembles the joint density of states for parabolic
bands, as shown in Fig. 4.2 (f).
4.3 Optical Conductivity and Landau Quantization

Next, we cover the optical conductivity in the presence of Landau levels. In Landau-level spectroscopy we distinguish between intra- and interband transitions, which are visually represented in Fig. 4.3. Intra-band transitions occur between Landau levels that originate from the same electronic band and require the Fermi level to intersect with it. In the small-field limit, this contribution to the optical conductivity can be described using the Drude model [70]. We are interested in large fields, however, where clear Landau-level separation dominates the spectrum. The interband transitions cover the remaining transitions between Landau levels that originate from different electronic bands. For completeness, and given its great importance, the low-frequency limit of the intraband transitions, which result in the quantum Hall effect, is treated in Appendix C.

Recall the interband optical conductivity from the previous section

$$\sigma_{\alpha,\beta}(\omega^+) = -\frac{i e^2}{\hbar V} \sum_{n,m,k} \frac{f_{mn}}{\omega_{mn}} \frac{\langle n | \hat{v}_\alpha | m \rangle \langle m | \hat{v}_\beta | n \rangle}{\omega^+ - \omega_{mn}}, \quad (4.43 \text{ revisited})$$

with the notation $|n\rangle \equiv |n, k_x, k_y, k_z\rangle$, where $n$ is the band index. In the presence of a magnetic field, the states are fully described by $|n, k_z\rangle$ and the quantum numbers $k_x$ and $k_y$ can be discarded. Here, $n$ and $m$ index the Landau levels. They run over the Landau levels of all bands, however, to shorten the notation, an extra band
CHAPTER 4. OPTICAL CONDUCTIVITY

index has not been written out but is implicit. The degeneracy of the Landau levels in the $k_x$-$k_y$-plane is given by

$$N = \frac{BL_x L_y}{\Phi_0} = \frac{eBL_x L_y}{2\pi \hbar},$$  \hspace{1cm} \text{(4.53)}$$

which is the total magnetic flux divided by the magnetic flux quantum $\Phi_0 = 2\pi \hbar/e$. Replacing the sum over $k_z$ with an integral $\sum_{k_z} \rightarrow \frac{L_z}{2\pi} \int_{k_z} dk_z$ and the sum over $k_x$ and $k_y$ with the factor $N$, we arrive at the Landau-quantized optical conductivity $\sigma_{LL}$ [71]

$$\sigma_{LL\alpha,\beta}^{\alpha,\beta}(\omega) = -iG_0 \hbar l^2 B \sum_{n,m} \int_{k_z} \frac{dk_z}{2\pi} \frac{f_{mn}}{\hbar \omega_{mn}} \frac{\langle n | \hat{v}_\alpha | m \rangle \langle m | \hat{v}_\beta | n \rangle}{\omega^+ - \omega_{mn}}.$$ \hspace{1cm} \text{(4.54)}$$

Note, that we introduced the conductance quantum $G_0 = e^2/(2\pi \hbar)$. For spin-degenerate systems, this formula can be found with a prefactor of 2 in the literature. In our case, however, we consider $\sum_{n,m}$ to sum over each individual band, including possibly spin-degenerate ones. For the Faraday configuration, the integration over $k_z$ effectively multiplies the equation with the number of states along $k_z$.

To illustrate the optical response of Landau levels, we treat the examples from the previous Section 4.2 once more. For the first case $H = \frac{\hbar k^2}{2m} \sigma_z + c\sigma_z$, however, the optical transition elements vanish again, like in the fieldless case. Thus, we start with the second example, given by the Hamiltonian $\hat{H} = \frac{1}{2m} \hat{p}^2 \sigma_z + c\sigma_x$ in Eq. (4.49). Using Peierls’ substitution, the Hamiltonian becomes

$$\hat{H} = \hbar \omega_c \left( \hat{n} + \frac{1}{2} \right) \sigma_z + \frac{\hat{p}_z^2}{2m} \sigma_z + c\sigma_x.$$ \hspace{1cm} \text{(4.55)}$$

The velocities can also be obtained via Peierls’ substitution and
4.3. LANDAU QUANTIZATION

Figure 4.4: Interband optical conductivity $\sigma_{+}^{LL}$ of the first 120 Landau levels for an isotropic pair of parabolic bands obtained from the Hamiltonian given in Eq. (4.49). Left: Color plot of $\sigma_{+}^{LL}$, showing the Landau level spectrum at $B = 10 \text{T}$. Right: Stacked plot of $\sigma_{+}^{LL}$ for different values of $B$. For the plots we chose the mass $m = m_0/20$ with free-electron mass $m_0$, $c = 20 \text{ meV}$, a scattering rate of $\hbar \gamma = 2 \text{ meV}$ and $E_F = 0$.

read

\[ \hat{v}_+ = \frac{1}{\sqrt{2}} \frac{\hat{p}_x + i(\hat{p}_y + eB\hat{x})}{m} = \frac{\hbar}{m l_B} \hat{a}^\dagger, \quad (4.56) \]
\[ \hat{v}_- = \frac{1}{\sqrt{2}} \frac{\hat{p}_x - i(\hat{p}_y + eB\hat{x})}{m} = \frac{\hbar}{m l_B} \hat{a}. \quad (4.57) \]

The resulting interband optical conductivity $\sigma_{+}^{LL}$ is plotted in Fig. 4.4, showing the Landau level transitions. While the first term in Eq. (4.55) is clearly linear in $B$, the band coupling introduced by the term $\sigma_x$ leads to a strongly positive curvature near the edges of the band gap. We can also see, that the strength of the interband transitions strongly decreases for photon energies $E = \hbar \omega$ much larger than the band gap like in the fieldless case, see Fig. 4.2 (b). For
large $B$, the Landau levels approach linear dispersion far from the gap.

For the Hamiltonian in Eq. (4.51), to obtain the Landau quantized version, we make use of the fact that we can write the Peierls’ substituted momenta in terms of the ladder operators

\[
\hat{p}_x = \frac{1}{\sqrt{2} l_B} \hbar (\hat{a} + \hat{a}^\dagger), \\
\hat{p}_y + eB\hat{x} = \frac{i}{\sqrt{2} l_B} \hbar (\hat{a} - \hat{a}^\dagger).
\]

Then, the Peierls’ substituted Hamiltonian reads

\[
\hat{H} = \frac{1}{2} \hbar^2 \left[ (\hat{a} + \hat{a}^\dagger)^2 \sigma_x - (\hat{a} - \hat{a}^\dagger)^2 \sigma_y \right] + \frac{\hbar^2}{2m} \sigma_z + c\sigma_z. 
\]

The substituted velocities are

\[
\hat{v}_+ = \frac{\hbar}{2ml_B} \left[ (\hat{a} + \hat{a}^\dagger)\sigma_x - (\hat{a} - \hat{a}^\dagger)\sigma_y \right] \\
\hat{v}_- = \frac{\hbar}{2ml_B} \left[ (\hat{a} + \hat{a}^\dagger)\sigma_x + (\hat{a} - \hat{a}^\dagger)\sigma_y \right].
\]

Here, we expect the bands to host transitions regardless of whether $c \neq 0$. This is confirmed by the calculated optical response for $c = 0$, shown in Fig. 4.5. In this case, the transitions are perfectly linear in $B$ and of the same strength for all Landau levels.

Finally, we consider one additional example and look at the optical conductivity for the case of a Landau quantized isotropic Dirac cone

\[
H = v\alpha\hat{p}, 
\]

with linearly dispersing bands in the zero-field case. Here $\alpha = (\alpha_x, \alpha_y, \alpha_z)$ is the vector of $\alpha$- matrices, which are defined as $\alpha_i = \sigma_z \otimes \sigma_i$ in the chiral basis, where $i \in x, y, z$. The Dirac cone consists
4.3. LANDAU QUANTIZATION

Figure 4.5: Interband optical conductivity $\sigma_{LL}^{+}$ of the first 120 Landau levels for an isotropic pair of parabolic bands obtained from the Hamiltonian given in Eq. (4.51). Left: Color plot of $\sigma_{LL}^{+}$, showing the Landau level spectrum at $B = 10$ T. Right: Stacked plot of $\sigma_{LL}^{+}$ for different values of $B$. For the plots we chose the mass $m = m_0/20$ with free-electron mass $m_0$, $c = 0$ meV, a scattering rate of $\hbar \gamma = 2$ meV and $E_F = 0$.

of two superimposed Weyl cones and the corresponding Landau-quantized Hamiltonian is discussed in Section 3.2. The velocity operators in this case are rather simple, as they contain no momentum operators

$$\hat{v}_+ = \frac{v}{\sqrt{2}} \sigma_z \otimes (\sigma_x + i \sigma_y), \quad (4.64)$$

$$\hat{v}_- = \frac{v}{\sqrt{2}} \sigma_z \otimes (\sigma_x - i \sigma_y), \quad (4.65)$$

and they are no different from the zero-field operators. The optical response is plotted in Fig. 4.6. At $B = 10$ T, the transitions peak at the interband energies $E_{n}^{\text{inter}} = (\sqrt{n+1} + \sqrt{n})\sqrt{2\hbar e v^2}B \approx (\sqrt{n+1} + \sqrt{n}) \times 46$ meV with the typical $\sqrt{B}$-dependency for linear
bands, as discussed in Section 3.2. A clear distinction between the Landau level transitions of the parabolic bands and the linear bands is apparent, making massless carriers easy to identify using Landau level spectroscopy.

To summarize, we have derived a general susceptibility function using the Kubo formalism. We focused our attention on the optical conductivity, which is the auto-correlation function of the electrical current density. We have seen how distinct parabolic models with similar dispersions can yield very different optical responses, not necessarily with an obvious linear-in-B dependency and that the Landau levels of linear bands yield a characteristic $\sqrt{B}$-dependence. In the next chapter, we discuss the experimental setup.
4.3. LANDAU QUANTIZATION
Part II

Experimental Techniques
5.1 Spectrometry and Cryostat

We perform measurements in reflection geometry using the Bruker Vertex v80 spectrometer. The core of the spectrometer is based on a Michelson interferometer, depicted in Fig. 5.1. The source light is collimated by a parabolic mirror into the interferometer, consisting of a beam splitter, a static mirror and a movable mirror. Displacing the movable mirror, the output signal can be tuned through several periods of constructive and destructive interferences. The intensity of the signal varies as

\[ I(dL) = I_0 \cos^2(kdL) \]  

(5.1)

where \( k = 2\pi/\lambda \), \( dL \) is the mirror displacement and \( \lambda \) the wavelength of a monochromatic input signal. The input wavelength can then be obtained by a fast Fourier transform of the interference pattern, as
5.1. SPECTROMETRY AND CRYOSTAT

Figure 5.2: Intensity profile versus mirror displacement for a wavelength of $\lambda = 600$ nm.

illustrated in Fig. 5.2. Due to $I \propto \cos^2(kx) \propto \cos(2kx)$, the Fourier transform peaks at half the input wavelength. In Fig. 5.3, the full beam path within the Bruker Vertex 80v is shown, as given in [72], adjusted in accordance with the magneto-reflectivity setup at the Laboratoire National des Champs Magnétiques Intenses (LNCMI) in Grenoble. The source radiation is provided by either a Globar or a Hg-lamp. Infrared magneto-optical measurements were performed in the range $\approx 5$-225 meV. In addition, the spectrometer in Fig. 5.3 includes an aperture wheel in the beam path, placed before the Michelson interferometer.

After the beam passes through the Michelson interferometer, it is collimated again, onto a parabolic gold mirror with a large focus length. The mirror focusses the beam onto the sample, fixed at the bottom of a sample holder that is roughly 1.5 meters in length and is inserted into the cryostat below. Before the light reaches the sample, it passes through another beam splitter installed at the top of the sample holder, which redirects a part of the reflected signal into a bolometer for detection.
A detailed sketch of the setup, consisting of the Bruker Vertex 80v in conjunction with a Oxford Spectromag cryostat, is provided in Fig. 5.4. The parabolic gold mirror in Fig. 5.3 is contained within the indicated box in front of the spectrometer. Below it, the sample probe is attached, which is inserted into a liquid helium bath cryostat. The probe contains a finite amount of helium exchange gas, cooling the inside of the probe down to liquid-helium temperature of $T = 4.2$ K. Optionally, the bath can be pumped to lower pressures to achieve a temperature of around $T = 1.8$ K. The sample is attached on a gold cone and inserted into a sample holder, which can be rotated back and forth between two sample positions. This allows to measure a gold reference signal besides the sample, which is used to optimize the beam path prior to the measurement. The Spectromag cryostat contains a superconducting coil, delivering a field of $B = 13$ T. As of 2021, the setup was upgraded with a new coil delivering up to $B = 16$ T. All measurements are performed exclusively in Faraday configuration, meaning that the wave vector of the probing light impinging on the sample is in parallel to the applied magnetic field.

Figure 5.3: Simplified Bruker Vertex 80v spectrometer setup with optical beam path.
Figure 5.4: Shown on the left is the Bruker Vertex 80v spectrometer coupled to a cryostat via a box containing a parabolic mirror. The signal, which is reflected off the sample, is redirected into a bolometer. A sketch of the probe, containing the sample, is shown on the right. The sample holder can be rotated back and forth between two sample positions.

5.2 High-Field Magnet

The LNCMI Grenoble is part of the European Magnetic Field Laboratories (EMFL) spanning currently over four high-field institutions: the LNCMI Grenoble, the LNCMI Toulouse, the HZDR in Dresden and the HFML in Nijmegen. Grenoble is equipped with a series of continuous water-cooled high-field magnets. The magnetic field of superconducting coils is limited due to their critical field beyond which the superconducting phase breaks down. To achieve higher fields, copper coils are used. These are limited mainly by their tensile
strength, as retaining a magnetic field exerts large magnetic pressure on the coils. The high-field coil used in Grenoble is rated for a maximum field of $B = 36$ T. At the time of measurement, due to safety precautions, it was limited to a maximum of 33 T. To reach this field, a large continuous current of $\approx 28$ kA is driven through the coil with a total power consumption of 18.7 MW. The entire load is given off as heat and thus the system requires sophisticated water cooling. Up to 300 liters of cooling water per second are pumped through the coils. The outer Bitter coil stack consists of interchanging metal and insulator plates. The slits in each individual plate provide cooling channels when stacked. The larger circular holes serve as screw holes for fixture. The inner polyhelix consists of solid copper pieces insulated by layers of epoxy resin mixed with fiber glass. Here, the small gaps between each stack serve as cooling slits, growing wider towards the center.

![Schematic diagram of the resistive coils in the high-field magnet at the LNCMI Grenoble](image)

Figure 5.5: Schematic diagram of the resistive coils in the high-field magnet at the LNCMI Grenoble [73], consisting of an outer Bitter coil stack and the inner polyhelix.
5.3 Bolometer

The cryogenic bolometer is a suitable detector for infrared radiation, with an example depicted in Fig. 5.6. The detection scheme of a bolometer is based on an electric circuit called a Wheatstone bridge. The Wheatstone bridge describes the arrangement of resistors depicted in Fig. 5.7. The resistor $R_T$ is thermally sensitive. It is referred to as a barretter if its resistance drops, or a thermistor if it rises as its temperature is lowered. A barretter (positive temperature coefficient), is for example a metallic wire. Thermistors (negative temperature coefficient) are made of a semiconductor material. The voltage $V$ is given by

$$V = \frac{R_T}{R_3 + R_T} V_S + \frac{R_1}{R_2 + R_1} V_S.$$  \hspace{1cm} (5.2)

The circuit is in balance ($V = 0$) if $R_2/R_1 = R_3/R_T$. Thus, changes in the value of $R_T$, due to the heating by impinging radiation, can be read out as a voltage across the bridge. Modern bolometers include an oscillating circuit on one of their arms as well as a differential amplifier to separate the measured signal from static background.
noise and improve the signal-to-noise ratio.
5.3. BOLOMETER
MATERIALS AND TECHNICAL DETAILS

TaP

The magneto-reflectivity data was taken in reflectivity mode in the Faraday configuration on the (001)-oriented facet of a TaP crystal. The facet area was roughly 2 by 2 mm$^2$, and the sample was kept in helium exchange gas cooled to $T = 1.8$ K during the measurements. The sample was placed in a superconducting solenoid for measurements in fields up to 13 T or a resistive coil up to 33 T. Far-infrared radiation ($\sim 5 - 223$ meV) from a globar or Hg lamp was delivered via light-pipe optics. For data below 71 meV, a Mylar beam splitter (T222) was inserted into the interferometer, for data above 71 meV a KBr beam splitter was chosen. On the return path, reflected off the sample, the light passes through a CdTe or silicon beam splitter. The light is partially reflected onto a liquid-helium-cooled bolometer, which is placed outside the magnet for detection and connected to a Bruker Vertex80v Fourier-transform spectrometer. Data was taken in steps of 0.25 T for fields below 13 T and in steps of 0.5 T above 13 T.

Single crystals of TaP were obtained according to the procedure reported in [74]: a polycrystalline TaP powder was synthesized in a direct reaction of pure Ta and P, while the single TaP crystals were grown from the powder via chemical vapour transport with
iodine.

**NbAs**

The magneto-optical data was collected in reflectivity mode in the Faraday configuration from a (001)-oriented facet of a NbAs crystal. The facet area was roughly 2 by 2 mm$^2$, and the sample was kept at $T = 1.8$ K in helium exchange gas during the measurements. The sample was placed in a superconducting coil, which provided magnetic fields up to 13 T. Far-infrared radiation ($\sim 5 - 150$ meV) from a globar or Hg lamp was delivered to the sample via light-pipe optics. For data below 46 meV, a Mylar beam splitter (T222) was inserted into the interferometer, for data above 46 meV a KBr beam splitter was chosen. On the return path, reflected off the sample, the light passes through a silicon beam splitter. The light is partially reflected onto a liquid-helium-cooled bolometer, which is placed outside the magnet for detection and connected to a Bruker Vertex80v Fourier-transform spectrometer. Data was taken in steps of 0.25 T. For the colormaps the data was linearly interpolated to a step size of 0.0625 T to obtain a smoother picture.

Single crystals of NbAs were obtained according to the procedure reported in [74]: a polycrystalline NbAs powder was synthesized in a direct reaction of pure Nb and As, while the single NbAs crystals were grown from the powder via chemical vapour transport with iodine.

The *ab initio* calculations were performed with the WIEN2k code [75], which is based on the (linearized) augmented plane-wave and local orbitals [(L)APW+lo] method to solve the Kohn-Sham equations [76] of DFT. The exchange-correlation potential was calculated using the PBE-GGA functional [77]. The unit cell was defined on space group No. 109, $I4_1md$ with lattice constants $a = b = 3.4517$ Å and $c = 11.680$ Å. The self-consistent field calculations converged
on a $10 \times 10 \times 10$ $k$-mesh (charge convergence below $10^{-5}$ e, energy below $10^{-6}$ Ry) for the two-dimensional and on a $30 \times 30 \times 30$ $k$-mesh (charge convergence below $10^{-6}$ e, energy below $10^{-7}$ Ry) for the one-dimensional band structure cuts.

GdPtBi

The magneto-optical data were collected in reflectivity mode from a (111)-oriented facet with the area of roughly 2 by 1.1 mm$^2$. The sample was kept at $T = 4.2$ K in helium exchange gas during the measurements. The sample holder was placed in the new superconducting coil, which provided magnetic fields up to 16 T. Far-infrared radiation ($\sim 4 - 150$ meV) from a Hg lamp (below 45 meV) and a globar (above 45 meV) was delivered to the sample via light-pipe optics. For data below 45 meV, a solid state beam splitter (T230) was inserted into the interferometer, for data above 45 meV a Mylar multilayer beam splitter (T222) was chosen. An Si beam splitter directed the reflected sample signal into a liquid-helium-cooled bolometer placed outside the magnet for detection and was analyzed by a Bruker Vertex80v Fourier-transform spectrometer. The sample reflectivity $R_B$ at a given magnetic field $B$ was normalized by the sample’s reflectivity $R_0$ measured at $B = 0$. A recently installed heater on the sample holder allowed to measure GdPtBi at elevated temperatures above $T = 4.2$ K. Additional data was taken for $T = 25$ K and $T = 50$ K up to 45 meV, above the antiferromagnetic transition temperature of $T_N \approx 9$ K. Data was taken in steps of 0.25 T for the measurements at $T = 4.2$ K and in steps of 0.5 T for the measurements at $T = 25$ K and $T = 50$ K. For the colormaps, the data was linearly interpolated to a step size of 0.0625 T for $T = 4.2$ K and to 0.125 T for $T = 25$ K and $T = 50$ K to obtain a smoother picture.

GdPtBi single crystals were grown by the solution method from a Bi flux. Freshly polished pieces of Gd, Pt, and Bi, each of purity
larger than 99.99%, in the ratio Gd:Pt:Bi = 1:1:9 were placed in a tantalum crucible and sealed in a dry quartz ampoule under 3 mbar partial pressure of argon. The filled ampoule was heated at a rate of 100 K/hr up to 1200°C, followed by 12 hours of soaking at this temperature. For crystal growth, the temperature was slowly reduced by 2 K/hr to 600°C. Extra Bi flux was removed by decanting it from the ampoule at 600°C. Overall, the crystal-growth procedure followed closely the ones described in Ref. [61]. The crystals’ composition and structure (noncentrosymmetric F$$\overline{4}$$3m space group) were checked by energy dispersive x-ray analysis and Laue diffraction, respectively.

The DFT calculations were performed with the WIEN2K code [75], which is based on the (linearized) augmented plane-wave and local orbitals [(L)APW+lo] method to solve the Kohn-Sham equations [76] of DFT. The exchange correlation potential was calculated using the PBE-GGA functional [77]. The unit cell was defined on space group No. 216, I$$\overline{4}$$1md with lattice constants \(a = b = c = 6.680\ \text{Å}\). The on-site Coulomb potential for the 4f-electrons of Gd was chosen as \(U = 10\ \text{eV}\). Results were cross-checked with runs performed on a \(40 \times 40 \times 40\ k\)-mesh, with the energy converged below \(10^{-8}\ \text{Ry}\) and charge converged to \(10^{-8}\ \text{e}\) without spin-polarization and a \(50 \times 50 \times 50\ k\)-mesh converged below \(10^{-7}\ \text{Ry}\) and \(10^{-7}\ \text{e}\) with polarized spins.
Part III

Results
Chapter 7

TaP - BAND INVERSION MADE VISIBLE

The Weyl equation is results from reducing the massless Dirac equation from a four-component to a two-component theory. However, this lifts the symmetry of left and right, referring to the chirality of the fermion [10]. In 1937, Herring proposed that Weyl fermion quasiparticles could be realized in condensed matter systems [24], though such systems would not be realized for still quite a long time. Nearly 50 years later, Nielsen and Ninomya suggested that a “quasi” chiral anomaly could be realized in crystals [23] hosting Herring’s Weyl fermions.

Notable early ideas to realize Weyl fermions on a lattice involved the influence of magnetic order on the pyrochlore iridates [25], layered magnetic hetero-structures [49, 79], the inversion-broken HdTe/CdTe hetero-structures [50], photonic crystals [80] and non-centrosymmetric solid solutions LaBi$_{1-x}$Sb$_x$Te$_3$ and LuBi$_{1-x}$Sb$_x$Te$_3$ [81]. Only as late as 2015 could experiments provide the first proof of a Weyl semimetal, TaAs, by detecting the associated open Fermi arcs using ARPES [26, 27, 46, 82–84]. This naturally attracted much interest in the entire “TaAs family”, as it is often referred to. The TaAs family is a series of the chemi-
cally closely related compounds consisting of TaAs, TaP, NbAs and NbP and which share similar band structures [28, 85]. Beyond fundamental research, such as on the chiral anomaly, the topology of Weyl semimetals harbors the potential for many technical applications. This includes, for example, a use in hydrogen catalysis [86], the realization of Majorana fermions in a superconducting phase [87] or when interfaced with superconductors [88–92], a dissipationless transfer of charges via the surface, which may pave the way to low-energy-consuming electronic or spintronic devices [93].

As we had explored in Chapter 3 and 4, one way to probe the massless Weyl fermions in condensed matter, is to subject the material to a magnetic field and detect its Landau levels by investigating the infrared response. In this thesis, magneto-optical measurements are discussed for two of these TaAs-family compounds. Namely, we are going to investigate the materials TaP and NbAs, starting off with the former in the present chapter. The crystal structure of TaP, shown in Fig. 7.1, is tetragonal and belongs to the space group $I4_1md$, No. 109, with the lattice constants being $a = b = 3.437\, \text{Å}$ and $c = 11.656\, \text{Å}$. Evidently, the unit cell lacks inversion symmetry. The positions of the Weyl cones within the Brillouin zone of TaP are shown in Fig. 7.2. Here, we define directions in the Brillouin zone as $k_z$ being along unit cell axis $c$, and $k_x$ and $k_y$ along $a$ and $b$. The space group $I4_1md$ contains two mirror planes, shown in yellow, as well as a $C_4$ rotation around the $z$-axis. The 24 shown Weyl points can be separated into two distinct types, related by the mirror symmetries. These are four pairs, denoted $W_1$, in the $k_z = 0$ plane and the remaining eight pairs, denoted $W_2$. We note that mirror symmetries relate cones of opposite chirality, whereas inversion relates cones of the same symmetry [25]. This can be understood from the fact that the Berry curvature $\Omega$ behaves like a pseudo-vector, in analogy to a magnetic field $B$. The behavior of pseudo-vectors with respect to symmetry operations is illustrated in Fig. 7.3. Not shown are rotations, which leave the chirality of the Weyl cones unaffected.
Much information of the electronic band structure can be obtained using infrared spectroscopy. Though, analysing an optical spectrum can prove very difficult since contributions from the entire band structure may not easily separated. One way of restricting transition rules is by using an applied magnetic field, forcing the electrons into circular orbits. This yields the quantized Landau levels with well separated gaps, as shown in chapter 3. The transition rules now obey $|\Delta n| = 1$, where $n$ is the quantum number associated with the Landau levels. The characteristic energy-field dependencies $E(B)$ can then be used to identify their origin within the band structure. If different parts of the band structure contribute to the Landau level spectrum simultaneously, the associated Landau-level transitions will overlap in obvious ways, leading to crossing sets of lines. Further, often the Landau quantization is detectable only in a few sections of the band structure, where either the band couplings are strong enough, the electron density is high enough or the scattering time low enough. This has both a positive and a negative aspect to it. Some compounds might show no field-dependent response at all, while in other cases it singles out a feature within the band structure. Landau level spectroscopy is especially suited to detect massless Weyl fermions: For massive, meaning quadratically dispersing, massive bands, the Landau levels disperse linearly with the applied magnetic field, whereas for linear, massless bands they...
Figure 7.3: The Berry flux behaves like a pseudovector, analogous to the magnetic field created by a current density. Mirror planes and inversion centers relate Weyl cones of opposite chirality. Breaking the inversion symmetry thus allows cones of the same chirality to lie at opposite $k$-vectors.

Disperse proportional to $\sqrt{B}$, allowing for easy distinction. With this, let us move on to the experiment.
CHAPTER 7. TaP - BAND INVERSION MADE VISIBLE

7.1 Measurement

The results of the infrared-Landau-level spectroscopy on TaP are shown in Fig. 7.4. The single crystal TaP sample has been prepared by the method described in Refs. [94–96]. The magneto-reflectivity data was taken in the Faraday configuration on the (001)-oriented facet. A macroscopic area of the sample ($\approx 4 \text{ mm}^2$), kept at $T = 2 \text{ K}$ in the helium exchange gas and placed in a conducting coil. For magnetic fields below $B = 13 \text{ T}$, superconducting solenoid was used, for fields above $B = 13 \text{ T}$ a water cooled resistive coil delivered up to $B = 33 \text{ T}$. The source radiation was emitted by a globar or Hg lamp, which was analyzed by a Fourier-transform spectrometer and delivered via light-pipe optics, see Fig. 5.4. The reflected light was detected by a liquid-helium-cooled bolometer placed outside the magnet. The setup is identical for both the superconducting and the resistive magnet. The sample reflectivity $R_B$ at a given magnetic field $B$ was normalized by the sample reflectivity $R_0$ measured at $B = 0$. This yields the relative change of the spectrum in the applied magnetic field. The measured spectrum is exceptionally rich with features and traces of the features have been plotted on the right-hand panel Fig. 7.4. The spectrum shows several interesting characteristics:

1. We observe a series of tightly spaced lines marked dashed green, which likely originate from interband transitions. While they are slightly nonlinear in $B$ towards large fields, they disperse relatively linear in the low field region, indicating transitions between mostly parabolic bands.

2. A constant energy feature emerges around 42 meV, marked orange, which is increasing in size with the magnetic field. Its origin is not obvious and is associated with a dip in the zero field conductivity, discussed shortly.

3. A set of low energy features marked with dotted blue lines,
Figure 7.4: In-field reflectivity $R(B)$ divided by the zero-field reflectivity $R(0)$ showing relative changes of the energy spectrum $E(B)$ with magnetic field showing the traces of over a dozen of Landau level transitions, following [1]. For visibility, they have been extracted and plotted on the right. We focus our attention on three sets of features. First, the unusual downwards dispersing lines that have been marked red. Second, the dense fan of regularly spaced dashed green lines, indicating interband transitions between the crossing electron and hole bands near the Fermi level. Third, the strongly pronounced constant-energy feature at 42 meV, marked with a thick orange line [34, 97].
containing among them strongly curved Landau levels, which intersect with the plasma edge feature. As we expect to measure Weyl fermions and thus Landau levels dispersing with $\sqrt{B}$, these lines are of special importance.

4. Finally, and most importantly, we observe a fan of lines extrapolating to roughly 70 meV at $B = 0$. This set of features contains downwards dispersing Landau levels, marked red. They are uniquely indicative of the present band inversion, which is a necessity for a band structure to host Weyl cones.

7.2 Discussion: Downwards Dispersing Landau Levels

It is clear from Fig. 7.4 that the regular conical approximations, which are often employed to describe Weyl semimetals [98–100], are not sufficient to describe the present experimental observations. Instead, we have to additionally account for the inverted nature of the band structure. To describe the time-reversal invariant Weyl semimetal, we start with a generic $2 \times 2$ Hamiltonian, often applied to systems with inverted bands

$$H = \begin{pmatrix} \Delta - \frac{\hbar^2 q^2}{2M} & \gamma(q) \\ \gamma^*(q) & -\Delta + \frac{\hbar^2 q^2}{2M} \end{pmatrix}.$$  \hspace{1cm} (7.1)

Here, we employ the common notation $q_i = k_i/a_i$ where $a_i$ are the lattice constants, $i \in \{x, y, z\}$, and $k_i \in [-\pi, \pi i)$. The solutions to this Hamiltonian can be concisely given as

$$E_{\pm} = \pm \sqrt{(\Delta - \frac{\hbar^2 q^2}{2M})^2 + |\gamma(q)|^2} \hspace{1cm} (7.2)$$

The model hosts two parabolic bands with particle-hole symmetry. The displacement $\Delta$ overlaps the parabolic bands, intersecting them.
7.2. DISCUSSION: DOWNWARDS DISPERSING LANDAU LEVELS

Figure 7.5: The bands of the coupled parabolic model (left) which hosts conical intersections wherever the diagonal terms and off-diagonal terms of Hamiltonian (7.1) simultaneously vanish. This is shown in on the right, which plots the ellipses on which \( \pm(\Delta - \hbar^2 q^2/(2M)) = 0 \) and \( \gamma(q) = 0 \) respectively. The crossing points of the ellipses correspond to the conical intersections on the left [34, 97].

on a circular line called a nodal ring. Different choices for the off-diagonal couplings \( \gamma(q) \) will yield a variety of models, such as \( \gamma = \text{const.} \) for a nodal-loop semimetal [101], or a topological insulator for \( \gamma = \hbar v(q_x + iq_y) \) [102, 103]. We choose the off-diagonal terms as

\[
\gamma(q) = \gamma_0 - \frac{\hbar^2 q_x^2}{2m_x} - \frac{\hbar^2 q_y^2}{2m_y},
\]

with positive masses \( M, m_x, m_y > 0 \), which gaps the Hamiltonian out with the exception of four points where the diagonal and off-diagonal terms vanish simultaneously and create conical intersections, see Fig. 7.5 (a) and (b). The chosen parameters are
\[ M = m_y = m_x/12 = m_0/4 \]
\[ \gamma_0 = 10 \text{ meV} \]
\[ \Delta = 35 \text{ meV}, \]

where \( m_0 \) is the free electron mass. These cones merge at higher energies via two kinds of saddle points (Lifshitz transitions of neck-collapsing type [104, 105]), see Fig. 7.5 (b).

Indeed, the Weyl cones at opposite momenta \( \mathbf{q}^w \) and \(-\mathbf{q}^w\) have the same topological charge, and a second pair of Weyl cones is thus required to ensure the sum of topological charges to be zero. The proposed model thus may be relevant for any time-reversal invariant 3D Weyl semimetal. In the case of TaP, the \( \mathbf{q} = 0 \) point of the model bands may be straightforwardly associated with the \( \Sigma \) point [106, 107]. Since our approximation is local to the \( \Sigma \) point and not the \( \Gamma \) point, we simplified the model to not include any inversion symmetry breaking terms in the model. The simplicity of the model allows us to find analytically the positions and topological charges of the Weyl nodes, given as

\[
\gamma_0 = \frac{\hbar^2 q_x^2}{2 m_x} + \frac{\hbar^2 q_y^2}{2 m_y} \\
\Delta = \frac{\hbar^2 q_x^2}{2 M} + \frac{\hbar^2 q_y^2}{2 M}
\]

\[
\rightarrow (\pm q_x^w, \pm q_y^w) = \frac{2}{\hbar^2} \left( \pm \sqrt{\frac{m_x (M \Delta - \gamma_0 m_y)}{m_x - m_y}}, \pm \sqrt{\frac{m_y (M \Delta - \gamma_0 m_x)}{m_y - m_x}} \right).
\]

as well as of the locations of the saddle points, which are obtained by searching for the minimum of the equation

\[
E^2 = \left( \Delta - \frac{\hbar^2 q_y^2}{2 M} \right)^2 + \left( \gamma_0 - \frac{\hbar^2 q_y^2}{2 m_y} \right)^2 \tag{7.5}
\]
7.2. DISCUSSION: DOWNWARDS DISPERSING LANDAU LEVELS

for the $y$-component, and a corresponding expression for the $x$-component, to arrive at the coordinates

$$
\hbar q_{sp}^y = \pm \sqrt{\frac{2}{1/M^2 + 1/m_y^2}} \left( \frac{\Delta}{M} + \frac{\gamma_0}{m_y} \right) \tag{7.6}
$$

and

$$
\hbar q_{sp}^x = \pm \sqrt{\frac{2}{1/M^2 + 1/m_x^2}} \left( \frac{\Delta}{M} + \frac{\gamma_0}{m_x} \right) \tag{7.7}
$$

Simple expressions were also found for the energy of the saddle points, given as

$$
E_{sp}^y = E(0, q_{sp}^y, 0) = \pm \frac{|\Delta M - \gamma_0 m_y|}{\sqrt{M^2 + m_y^2}} \tag{7.8}
$$

and

$$
E_{sp}^x = E(q_{sp}^x, 0, 0) = \pm \frac{|\Delta M - \gamma_0 m_x|}{\sqrt{M^2 + m_x^2}} \tag{7.9}
$$

which yields $E_{sp}^y = \pm 17.7$ meV and $E_{sp}^x = \pm 7.9$ meV for our parameter choices (7.4). The local band extrema at $q = 0$, the cusps we referred to earlier, are given by

$$
E(q = 0) = E^{\text{cusp}} = \pm \sqrt{\Delta^2 + \gamma_0^2}, \tag{7.10}
$$

which yield $E^{\text{cusp}} = \pm 33.5$ meV. Let us now introduce a quantizing magnetic field along the $z$-axis in the Hamiltonian Eq. (7.1). Be reminded that we use a two-dimensional Hamiltonian and consider states only for $q_z = 0$. This is a common approach, as it greatly simplifies the complexity of the model and discussion, while being sufficiently representative of the band structure. Using the Peierls’ substitution and the representation of ladder operators,

$$
\hat{a} = \frac{l_B}{\sqrt{2}\hbar} [\hat{p}_x + i(\hat{p}_y + eB\hat{x})]
$$

$$
\hat{a}^\dagger = \frac{l_B}{\sqrt{2}\hbar} [\hat{p}_x - i(\hat{p}_y + eB\hat{x})]
$$

90
we obtain the following Hamiltonian in magnetic fields

\[
\hat{H}_B = \begin{pmatrix}
\Delta - \hbar \omega_c (a \dagger a + \frac{1}{2}) & \gamma (a, a \dagger) \\
\gamma^* (a, a \dagger) & -\Delta + \hbar \omega_c (a \dagger a + \frac{1}{2})
\end{pmatrix},
\]

(7.11)

with

\[
\gamma (a, a \dagger) = \gamma_0 - \frac{(a + a \dagger)^2}{m_y l_B^2} - \frac{(a - a \dagger)^2}{m_x l_B^2}.
\]

(7.12)

Here, \( \omega_c = eB/M \) is the cyclotron frequency and \( l_B = \sqrt{\hbar/(eB)} \) is the magnetic length. The search for eigenvalues of Hamiltonian (7.11) leads to diagonalization of an, in principle, infinitely large matrix. As pointed out in Chapter 3, we must limit ourselves to an approximate solution by truncating the matrix, taking account of a finite but sufficiently large number of Landau levels.

An example of a calculated Landau-level spectrum is presented on the left of Fig. 7.6. At low energies, the Landau levels follow an almost perfect \( \sqrt{B} \) dependence, characteristic for massless charge carriers \[108\]. For small magnetic fields, the Landau levels tend towards a fourfold degeneracy. This is due to the fact that the proposed model holds four Weyl cones, whose Landau levels must converge to the same energies as \( B \) tends towards 0 T. With a low energy approximation in the vicinity of the cones for \( \mathbf{q} = (\pm q_x^W, \pm q_y^W) \), we can calculate a effective velocities for the electrons within the conical bands, given by

\[
v_{\text{eff}} = \sqrt{|v_+ v_-|} = \sqrt{\frac{q_x^W q_y^W}{M} \left( \frac{1}{m_y} - \frac{1}{m_x} \right)}.
\]

(7.13)

Using the model parameters (7.4), yields \( v_{\text{eff}} = 1.37 \times 10^5 \) m/s. The
Figure 7.6: The Landau-level spectrum (left) as a function of $B$ calculated for the parameters in (7.4). The number of Landau levels considered in our calculations has been limited to $N = \pm 700$. The horizontal lines denote energies of the band extreme and saddle points in the (zero-field) band structure. False color-plot (right) of the calculated relative magneto-reflectivity, $R_B/R_0$. The reference spectrum $R_0$ has been approximated by the reflectivity trace calculated at $B = 1$ T and for $\Gamma = 5$ meV. The background dielectric constant was chosen as $\varepsilon_{\infty} = 20$ [34, 97].
Figure 7.7: Landau levels (blue) for $N = \pm 400$ and position of the saddle points (dashed green), compare Fig. 7.6. The red lines show a low-energy extrapolation for a Weyl cone with an effective velocity of $v_{\text{eff}} = 1.37 \times 10^5$ m/s.

Landau levels for the low energy model then disperse as

$$E_{\pm,n} = \pm \hbar \sqrt{\frac{2 v_{\text{eff}}^2}{l_B} n}. \quad (7.14)$$

A comparison between a conical model with $v_{\text{eff}}$ and the low energy region of the full model is given in Fig. 7.7, indeed matching quite well below $B \approx 3T$. The near-degeneracy is lifted with increasing $B$, and a pair of doubly degenerate levels emerges, and subsequently a quartet of non-degenerate Landau levels can be identified. These bifurcations occur near the saddle point energies indicated by horizontal dashed lines on the left in Fig. 7.7. In the intermediate energy range, the Landau levels exhibit rich (anti)crossing behavior. This happens above the saddle points, but still below the local maximum of the conduction band ($\mathbf{k} = 0$), indicated by dotted lines in the left panel Fig. 7.6. They originate from the cusps of the parabolic bands, extrapolating towards the values $E_{\text{cusp}}$, see Eq. (7.10), for $B \to 0$ T. At energies beyond $E_{\text{cusp}}$, all Landau levels follow nearly linear-in-B dependence, which is governed by the diagonal terms in the Hamiltonian Eq. (7.11).
To calculate the optical response of the model Hamiltonian in magnetic fields, we calculate the magneto-optical conductivity within linear-response theory, see Chapter 4. For bulk materials and circularly polarized light we have

$$\sigma^\pm (\omega, B) = 2iG_0 \frac{\bar{l}_\omega^2}{\hbar} \sum_{n,m,q_z} \frac{(f_m - f_n)}{E_n - E_m - \hbar\omega + i\Gamma} \langle m | \hat{v}_\pm | n \rangle (7.15)$$

where $G_0 = e^2/\hbar$ is the quantum of conductance, $\Gamma$ is the broadening parameter, and $\hat{v}_\pm = \hat{v}_x \pm i\hat{v}_y$ are the circular velocity operators with $\hbar v_i = \partial H / \partial q_i$. A closer analysis of the matrix elements, $\langle m | \hat{v}_\pm | n \rangle$, shows that the response is dominated by transitions following the selection rule $n = m \pm 1$, typical of electron-dipole excitations in isotropic materials. At the same time, the pronounced anisotropy of the studied system also activates excitations beyond this selection rule, especially when the final or initial state is nearby the saddle point. Consistently with Eq. (7.11), we consider only states around $q_z = 0$. Therefore, we have replaced the sum over $q_z$ in Eq. (7.15) by the integral of the characteristic $1/(E_m - E_n - \hbar\omega)^{1/2}$ profile in the joint density of states over the interval of $(E_m - E_n; E_m - E_n + \Gamma)$.

The right-hand side of Fig. 7.6 displays a false-color plot of the relative magneto-reflectivity $R_B/R_0$ calculated from Eq. (7.15), for the set of parameters in (7.4). This map displays fairly complex behavior. We may identify three main sets of inter-Landau-level excitations in Fig. 7.6 (b).

1. The transitions at low photon energies with $\sqrt{B}$, or more generally, sub-linear-in-$B$ dependence, which are related to excitations within the Weyl cones

2. The negatively dispersing transitions originating from the extrema of the inverted parabolic bands, which extrapolate to $2\sqrt{\Delta^2 + \gamma_0^2}$ in the zero-field limit.
3. A series of regularly spaced and linear-in-B excitations at high energies, an indicator for regular interband transitions between parabolic bands.

4. A feature just below 40 meV that corresponds to the transitions between the saddle-points $E_{sp} = \pm 17.7$ meV.

Considering the simplicity of the model Hamiltonian, it qualitatively captures the character of the observed features surprisingly well. Particularly striking is the fact that the parabolic Hamiltonian, which contains no linear terms, successfully reproduces the $\sqrt{B}$ scaling in the low field limit. Unexpectedly, we further find transitions between the saddle points of the intersecting bands feature near 40 meV, coinciding well with the horizontal feature in the measurement around 42 meV, see Fig. 7.4.
In the experimental data, however, the interpretation is less straightforward. The zero-field optical data in Fig. 7.8 and also the preceding optical study by Kimura et al. [95], indeed show in this spectral range a dissipative feature interpreted previously as due to excitations between the saddle points. Nevertheless, this feature nearly coincides in energy with the plasma edge in Fig. 7.8, which reflects the response due to all free electrons, including topologically trivial pockets. According to classical magneto-plasma theory [70], the plasma edge in the reflectivity exhibits splitting with $B$ and may thus mask effects related to excitations between saddle points.

7.2.1 Landau Levels along $k_z$

In the Weyl fermion approach, with purely linear Landau levels, one finds that the level for $n = 0$ remains a linear band along the non-quantized direction, $q_z$, with opposite dispersions for Weyl cones of opposite chirality. We had found that the low-energy limit in the Landau level spectrum approaches the same $\sqrt{B}$ scaling just as the purely linear Weyl fermions do. One may ask then whether the modelled Landau levels also produce the linear bands along $k_z$? To investigate this, we choose a parabolic dispersion along $k_z$, meaning we add a term $-\frac{\hbar^2 q_z^2}{2m_z} \sigma_z$, where $a_z = 11.544$ nm. For a qualitative discussion, we choose $m_z = M/8$. This choice seems low, but since $a_z \approx 3.34a_x$, the square of which appears in the $q_z^2$-term, the low mass $m_z$ just scales the bands back to a convenient energy frame. These bands are plotted in Fig. 7.9 (a). In Fig. 7.9 (b), (c) and (d) the Landau levels of the model for $B = 1$ T and $N = 240$, and $B = 2$ T and $N = 120$, and $B = 9$ T and $N = 120$ are shown.

For the lowest plotted field, one can see that the conical shape of the unquantized model is still visible, but there are no linear Landau levels. Interestingly, the cones are connected by a constant-energy Landau level. For higher fields, as in (d), the twisted Landau levels
Figure 7.9: (a) The bands of the coupled parabolic model in the $k_y$-$k_z$ plane, if a parabolic $k_z$ term with mass $m_z = M/8$ is added to the model. Subfigures (b)-(d) show the calculated Landau levels for $B = 1, 2$ and $9$ T, and $N = 240, 120$, and $120$ respectively.

that we had encountered in Fig. 7.5 (c) already, begin to appear. From these plots it also becomes more intuitively clear why the $\sqrt{B}$-dependent Landau levels are reproduced by the large-$N$ limit of the model. Fig. 7.10 compares the zero-field case with the case of $B = 1$ T. For the zero-field case, the dispersion along $k_z$ represents the band structure at $k_y = 0$. The case of $B = 1$ T we had already seen in Fig. 7.10 (b), but here we plotted it for lower $N$. Since for small $B$, the Landau levels are dispersing towards the center, for an accurate description it is necessary to use a sufficiently large $N$. This is because they disperse proportional to $B$, and if not enough
7.3. CONCLUSIONS

Figure 7.10: Illustration (left) of the zero field band structure with a parabolic dispersion along \( k_z \) discussed in the text and (right) the Landau levels for \( B = 1 \) T and \( N = 20 \).

levels are taken into account, they will not reach the center, meaning that we would overlook the low energy regime. For any finite field, the intersection of Landau levels are reproduced if \( N \) is chosen large enough.

7.3 Conclusions

In this chapter we presented the results of infrared spectroscopy measurements of the Landau levels in TaP in high magnetic fields up to \( B = 33 \) T. We observed an unconventionally rich Landau level fan, including a series of downwards dispersing levels. Using a parabolic nodal line model, which hosts Weyl cones at the intersections of the oppositely dispersing parabolic bands, we demonstrated that the downwards dispersing levels are a clear indicator of the band inversion. For energies below 42 meV we observe weak signatures of strongly curved Landau levels that we associate with the \( \sqrt{B} \)-scaling of Weyl fermions. Even though our model Hamiltonian is parabolic, containing only \( k^2 \)-terms, it is able to successfully reproduce the \( \sqrt{B} \)-dependence at low energies. Further, we could identify a pro-
nounced, constant-in-$B$ signature at 42 meV of what is likely the plasma edge of TaP.

To see whether the low-energy analogy to the Weyl cones can be driven further, we investigated the $k_z$ dispersion of the Landau levels parallel to the external field $B$. We extended the model by a parabolic dispersion along $k_z$, forming a nodal line in the $k_y$-$k_z$ plane. We found that the linearly-in-$k_z$ dispersing Landau levels, which would occur for ideal Weyl cones, are missing. Instead we saw that the cones are interconnected by flat Landau levels for low magnetic fields. With this simple addition to the model, we can conclude that the existence of $\sqrt{B}$-dependent Landau levels, or point-like intersections with a linearly dispersing low-energy extrapolation, do generally not necessitate the existence of chiral lowest Landau levels.
7.3. CONCLUSIONS
NbAs, like TaP, belongs to the TaAs-family and is likewise expected to show traces of Weyl fermions under magneto-optical investigation. A typical approach to describe the Weyl fermions in condensed matter systems is the linear band approximation

\[ H(k) = \sum_i v_i k_i \sigma_i, \quad (8.1) \]

where \( v_i \) are velocities, and \( \sigma_i \) are the Pauli matrices and which is valid in close proximity to the Weyl point. However, there are exceptions to this rule, as we have seen in the last section of the previous chapter. This linear-approximation approach was taken prior by a different group [99] to describe the results of their own Landau level studies on NbAs. Indeed, as we will see shortly, we measure a series of non-linear Landau levels ourselves in NbAs. However, with Nb being lighter than Ta, the spin-orbit coupling is significantly weaker.
8.1 Measurement

The NbAs sample is cooled via an exchange helium gas that remains in the sample chamber, which is inserted into the helium bath. By lowering the pressure of the helium bath using a vacuum pump, a temperature of $T = 1.8$ K is achieved. A magnetic field up to $B = 13$ T is provided by a superconducting coil and reflectivity data of NbAs is collected in Faraday geometry. Fig. 8.2 shows the relative reflectivity, that is the change of the reflectivity in fields $R(B)$ divided by the zero-field reflectivity $R(0)$. All reliably traceable features have been marked with red dots, which are shown superposed onto the measurement on the right hand panel in Fig. 8.2. The observed sharp lines correspond to transitions between quantized Landau levels. Three qualitatively distinct sets of Landau level transitions can be identified: First, a fan of slightly curved lines starting at just
CHAPTER 8. NbAs - HIDDEN WEYL FERMIONS

Figure 8.2: Relative reflectivity $R(B)/R(0)$ of NbAs. On the right, clearly identifiable features have been traced. A flurry of closely positioned curved Landau-levels is visible above 50 meV. Another curved line is visible below 50 meV as well as two parallel, almost linear lines below 25 meV. Lastly, another lone line is seen above 110 meV [110, 111].

about 50 meV, which are likely associated to interband transitions between non-parabolic bands. Second, a set of features just below 50 meV, hosting one curved and a set of two nearly linear lines. Third, a lone line that is found above 110 meV. This third feature
8.2 DISCUSSION: HYPERBOLIC BAND APPROXIMATION

seems to have no connection to the former two sets of features, likely originating from different bands or a different part of the Brillouin zone. On the other hand, the lines within the fan above 50 meV must be related to a common source. We find that we can successfully reproduce this fan as well as the three low-energy transitions within a single model Hamiltonian.

8.2 Discussion: Hyperbolic Band Approximation

Like TaP, the crystal structure of NbAs, see Fig. 8.3, hosts a $C_4$ four-fold rotational symmetry around the long axis, which we identify with the $z$-direction, and two mirror symmetries, but lacks an inversion symmetry. As for all members of the family, the Weyl cones of NbAs originate from a set of intersecting bands, which form nodal lines. The nodal lines gap out due to the influence of spin-orbit coupling, with the exception of a few points: the Weyl points. Fig. 8.4 shows band structure calculations for NbAs without the inclusion of spin-orbit coupling. The $\Gamma-\Sigma$ directions will be referred to as $k_x$ and $k_y$. In (c) and (d), calculated band structure cuts of NbAs are shown within the $k_x$-$k_y$-plane. The corresponding planes are indicated in Fig. 8.4(a), where the plane for Fig. 8.4(c) is positioned at the center of the Brillouin zone ($k_z = 0$), and Fig. 8.4(d), which is offset by roughly $k_z = \pm 43\pi/80$. In both cuts, we find sets of Dirac cones, related by the $C_4$ symmetry mentioned previously. We can gain more insight by looking at the band structure in the $k_x$-$k_z$-plane, see Fig. 8.4(b). It provides a look down
Figure 8.4: (a) The bcc Brillouin zone for NbAs. The green planes correspond to the $k_x$-$k_y$ planes for which band structures are illustrated in (c) and (d). (b) The band structure in the $k_x$-$k_z$ plane for $k_y = 0$ as viewed from above, on top of the energy surface. Gaps smaller than 50 meV have been colored orange, outlining the nodal rings within this plane. The dashed line represents the outline of the first Brillouin zone at $k_y = 0$. (c,d) Band structure renderings for the green planes illustrated in (a). Depending on the cut, the size and distance of neighbouring Dirac cones varies.

onto the $k_x$-$k_z$-plane of the three-dimensional band structure. By extending the plot beyond the first Brillouin zone and marking gaps smaller than 50 meV in orange, this plot visualizes the position of the nodal rings in NbAs within the $k_y = 0$-plane. Then, Fig. 8.4(c) and (d) are represented by line-cuts through (b). Wherever such a cut intersects with a nodal ring, Fig. 8.4(c) and (d) host a Dirac cone. Thus, by varying $k_z$ of the line cut, we can see in Fig. 8.4(b) how the distance between the Dirac cones is varied. Fig. 8.5 shows an angled view Fig. 8.4(b), visualizing the dispersion in the $k_x$-$k_z$-plane. The cut of Fig. 8.4(d) at $k_z = \pm 43\pi/80$, which grazes by the outer edge of the nodal line, yields Dirac cones of comparatively low velocities when contrasted with the cones of Fig. 8.4(c) and thus a high density of states.
Figure 8.5: Band structure of NbAs in the \( k_x-k_z \)-plane \((k_y = 0)\) with gaps smaller than 50 meV marked in orange. The orange markings outline the nodal rings of the NbAs band structure, compare Fig. 8.4(b).

When trying to model the measured Landau levels, it becomes immediately apparent that in order to fit the slopes of the lines, the interband transitions for linear bands (either Dirac or Weyl cones) \( \propto (\sqrt{(n + 1)B} + \sqrt{nB}) \) are too strongly curved and too widely spaced in energy to fit to the recorded interband fan above 50 meV. Clearly, neither are parabolic bands with purely linear-in-\( B \) scaling Landau levels going to fit to the spectrum. If we regard the dispersions of Fig. 8.5 and Fig. 8.5, we see that the overlapping bands are quite linear close to the point of intersection, but strongly curved near the cusps. The dispersion of the bands remind of the hyperbolic profile of the gapped cones of a massive Dirac particle. Thus, constant-\( k_z \) cuts through the nodal lines likewise demonstrate hyperbolic dispersions. Clearly, the smaller the overlap of the bands within the cut,

Figure 8.6: Actions of individual terms of \( H_4 \) in Eq. (8.2) on its energies.
the larger we can expect the influence of the hyperbolic band profile in shaping the Landau levels, rather than the linear profile near the intersection. Since the density of states within the $k_x$-$k_y$ plane is particularly high whenever this overlap is small, as discussed, the hyperbolic band profile might indeed be relevant for the experimental observations.

With this in mind, we aim to create a model Hamiltonian that yields Weyl points from an intersection of hyperbolic bands. A way to achieve this sketched in Fig. 8.6, the Hamiltonian for which is shown given as

$$H_4 = \begin{pmatrix} m\sigma_0 + c_1\sigma_z + c_2\sigma_x & \hbar v_x q_x\sigma_x + \hbar v_y q_y\sigma_y \\ \hbar v_x q_x\sigma_x + \hbar v_y q_y\sigma_y & -m\sigma_0 - c_1\sigma_z - c_2\sigma_x \end{pmatrix}.$$  \hspace{1cm} (8.2)$$

On the left of the figure, we start out with a Dirac cone, consisting of four bands, corresponding to the parameters $m = c_1 = c_2 = 0$. A mass parameter $m$ gaps the cones into two doubly degenerate hyperbolic bands. We can lift the degeneracy by introducing an effective Zeeman shift $\propto c_1\sigma_z$, which creates a nodal ring if the shift is larger than the mass gap. The remaining two bands get further removed from the intersection. Finally, by introducing a coupling $\propto c_2\sigma_x$ the bands become gapped except for two Weyl points. This model is incomplete yet, as the cusp-to-cusp gaps between the central two bands and the outer two bands is dependent on the mass and thus the hyperbolicity of the bands. However, the changes of
the band structure upon introducing spin-orbit coupling are fairly minor and cannot correspond to the mass gap. This can be seen in Fig. 8.7 where, similar to Fig. 8.4(c), the $k_x-k_z$ band structure of the now spin-orbit coupled DFT calculations has been plotted. Now, the nodal lines are gapped with the exception of two Weyl points. However, by coloring gaps smaller than 30 meV in blue, it becomes apparent that these gaps are rather small for most of the ring. Further, a plot of the band structure in the $k_x-k_y$-plane is provided in Fig. 8.8 for $k_z = 0$, which is the side view of Fig. 8.4(c) but with spin-orbit coupling included. The zoomed in version on the right of the figure shows the $W_1$ Weyl cones. We note that the band separation in the center of Fig. 8.8 (b) is as small as 15 meV and revalidate our previous assessment that this band structure is unsuitable for the linear band approximation, as was done for example in a previous publication [99].

All in all, we now want an additional tuning parameter, independent of the mass $m$ to give rise to the Weyl cones. To achieve this, we first establish spin degeneracy again, by doubling the size of the Hamiltonian. We create a Hamiltonian $H_8$, which list the $H_4$ twice.

Figure 8.8: Band structure of NbAs with spin-orbit coupling in the $k_x-k_y$-plane for $k_z = 0$, showing two $W_1$ Weyl points on the zoomed-in right-hand figure [110, 111].
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Figure 8.9: The bands for the model Hamiltonian Eq. (8.4), viewed along \( k_y \) (left) and along \( k_x \) (center). On the right is a visual representation of \( H_8 \), containing two copies of \( H_4 \) coupled by off-diagonal matrices.

\[
H_8 = \begin{pmatrix} H_4 & \text{Coupling} \\ \text{Coupling} & H_4 \end{pmatrix}.
\]

By coupling the separate spin degrees of freedom in Eq. (8.3) with off-diagonal \( 4 \times 4 \)-blocks, the Dirac cones can be split into two Weyl cones, illustrated in Fig. 8.9. Here, the bands that are split far from the Weyl cones, four in total, are not further considered and are thus not shown. The model should only reproduce the bands near the Fermi level. The final Hamiltonian is then given by Eq. (8.4)

\[
H_8 = \begin{pmatrix} H_4 & C \\ C^\dagger & H_4 \end{pmatrix}, \quad C = \begin{pmatrix} 0 & c_4 \sigma_x \\ c_3 \sigma_x & 0 \end{pmatrix}.
\]

Note, that we consider half of the bands to be redundant and we are interested only in the central four bands. The redundant bands are required only to yield the hyperbolic band shape of the bands of interest via the mass term. To obtain the quantized Hamiltonian,
we use Peierls’ substitution in Eq. (8.2) to obtain

$$\hat{H}_4 = \begin{pmatrix}
    m\sigma_0 + c_1\sigma_z + c_2\sigma_x \\
    v_x\hat{p}_x\sigma_x + v_y(\hat{p}_y + eB\hat{x})\sigma_y \\
    v_x\hat{p}_x\sigma_x + v_y(\hat{p}_y + eB\hat{x})\sigma_y \\
    -m\sigma_0 - c_1\sigma_z - c_2\sigma_x
\end{pmatrix}.$$  

We then use Eqs. (3.11) and (3.12) to write

$$\hat{p}_x = \frac{\hbar}{\sqrt{2l_B}}(\hat{a}^\dagger + \hat{a}) \quad (8.5)$$

$$\hat{p}_y = \frac{i\hbar}{\sqrt{2l_B}}(\hat{a}^\dagger - \hat{a}). \quad (8.6)$$

With this, we can express $\hat{H}_8$ with ladder operators and the Landau level spectrum can be obtained. We calculate the optical conductivity $\sigma_\pm$, see Eq. (4.54), and the reflectivity via the standard optical formulas for normal incidence, as given in Eq. (4.46).

This yields the reflectivity plotted in Fig. 8.10. So far, however, this does not look convincing. The Landau levels are very strongly pronounced in the relative reflectivity $R(B)/R(0)$, whereas the measurements show the reflectivity hover much closer around $R(B)/R(0) = 1$. To improve on this result, we have to consider that the Fermi surface only partially condenses into Landau levels, while the remaining part still contributes to the conductivity as free carriers. This can be achieved, by modifying the permittivity.
Table 8.1: Parameters for the spin-orbit coupled model Hamiltonian, see Figs. 8.11 and 8.14. A positive $c_4$ creates small gaps rather than Weyl cones.

\[\varepsilon = 1 + \frac{i\sigma}{\varepsilon_0\omega} = 1 + \alpha \frac{i\sigma_{\text{meas}}}{\varepsilon_0\omega} + (1 - \alpha) \frac{i\sigma_{\text{LL}}}{\varepsilon_0\omega}\] (8.7)

or

\[\varepsilon = 1 + \alpha (\varepsilon_{\text{ZF}}^{\text{meas}} - 1) + (1 - \alpha) \varepsilon_{\text{LL}}.\] (8.8)

Here, $\varepsilon_{\text{ZF}}^{\text{meas}}$ is the measured zero-field permittivity for NbAs. For the calculations an $\alpha = 0.35$ was used. The result of this calculation is shown in Fig. 8.11(a), which is in good agreement with the experimental features traced in red. This reflectivity has been produced with the parameters listed in Table 8.1 and the resulting energy bands are plotted in Fig. 8.11(b) and (c). We note that the parameters $c_3$ and $c_4$ are substantially smaller than $c_1$ and $c_2$, as is to be expected given that we had argued that the spin-orbit coupling is small. The required zero-field data of NbAs, measured at $T = 10$ K, is shown in Fig. 8.11(d).

Table 8.2: Velocities of the Weyl cones $W_1$ and $W_2$, taken from C. Lee et al. [28] and D. Grassano et al. [85].
8.2. DISCUSSION: HYPERBOLIC BAND APPROXIMATION

Figure 8.11: (a) Calculated Landau-level spectrum for $H_8$, with the parameters in Table 8.1, superposed with the measurement (red dots). (b,c) Associated energy bands of Hamiltonian $H_8$. (d) Zero-field complex permittivity of NbAs obtained following the procedure described in Refs. [112, 113].

At this point, we note another important point. To account for the required density of Landau level transitions in their spectrum, Yuan et al. [99] considered that both types of Weyl cones, $W_1$ and $W_2$, contribute to the spectrum. We, however, argue that due to the stark difference between the velocities $v_z$ of $W_1$ and $W_2$, and thus the difference in the density of states of these features, the recorded Landau level spectrum cannot originate simultaneously from both types of Weyl cones. The velocities, taken from Refs. [28] and [85], are listed in the Table 8.2 and we note that $v_z$ is more than a magnitude larger for $W_2$ than $W_1$. To illustrate the effect on the spectrum, the reflectivity plot of Fig. 8.11(a) has been recalculated for $v_z = 10^5$ m/s in Fig. 8.12 on the right, contrasted with the original on the left.
One magnitude of difference drastically changes the line intensities, compare $v_z, W_1$ and $v_z, W_2$ in Table 8.2 [110, 111]. Clearly, the features of $W_2$ would be overshadowed by those of $W_1$ and we cannot take both cones in order to produce sufficiently many Landau levels, thus further validating our approach of a hyperbolic band approximation.

Comparing the energy bands of our model with those of the DFT calculations, we notice that the Weyl cones in our model are estimated to be much smaller. This is not exactly unexpected however, since in our experimental spectrum of Fig. 8.2 we notice no clearly distinguishable $\sqrt{B}$-dependent Landau level fan. Further, if the cusp-to-cusp distance between the Weyl cones is around 15 meV, as we read from Fig. 8.8 earlier, we should be able to resolve an associated feature in Fig. 8.2, since we reliably measure down to 5 meV. We argue
8.2. DISCUSSION: HYPERBOLIC BAND APPROXIMATION

Figure 8.13: Band structure plane for \( k_y = 0 \) in the extended Brillouin zone. The green line along \( k_z \) grazes by the slightly gapped nodal rings, along which the dispersion is low. Three red lines mark the cuts in Fig. 8.14 [110, 111].

then, that the influence of Weyl cones on the spectrum is not directly observable. Further, as we have seen and argued in Fig. 8.4, cuts through the Brillouin zone grazing by a nodal ring result in much larger densities of states near the band crossings. In fact, our model estimates the central gap between the pairs of Weyl cones, see left illustration of Fig. 8.9 to be substantially smaller than the bands in Fig. 8.8 indicate (towards \( \Sigma \) the gap grows to beyond 100 meV). Thus instead, we search for a different feature more similar to our model, which we can expect to occur near the cuts along the perimeter of the nodal lines, like Fig. 8.4(d) where the Dirac cones approach each other, leading to a more flat dispersion along the \((k_x,0)\)- or \((0,k_y)\)-lines that pass through the Dirac points. Such cuts, in the spin-orbit coupled calculations for \( k_y = 0 \) are illustrated in Fig. 8.13 including spin-orbit coupling. Three different cuts are shown with different offsets along \( k_z \), with the lowest path corresponding to the \( \Gamma\)-N-\( \Gamma \) high symmetry line. As the offsets \( \Delta k_z \) are increased, the paths approach and pass through the gapped nodal ring, colored blue. These cuts, which contain a variable \( k_z \)-component, are chosen for illustration
Figure 8.14: (a) Calculated Landau level structure for the model with a positive $c_4 = 2$ meV. The bands are slightly gapped here. (b-d) Band structures along the red lines in (e) for different offsets $\Delta k_z$ from the point N. (e) Extended Brillouin zone with the $k$-paths for band structures (b-d). These are equivalent to the cuts shown in Fig. 8.13. (f) Surface rendering of the band structure in the plane containing N in the center, illustrating how the cuts (b-d) extend in two $k$-dimensions. (g) Bands of the model Hamiltonian of Eq. (8.4) for a positive $c_4 = 2$ meV. The bands are now slightly gapped and contain no Weyl cones. It compares qualitatively well to DFT band structure in (f), which resolves the good match for the Landau levels in (a).

purposes, as the band structure long these cuts is $\Gamma$-N-$\Gamma$ is symmetric around N, and the paths with finite offsets $\Delta k_z$ retain some of that symmetry. This can be seen Fig. 8.14(b), (c) and (d), where the band structure is plotted along these paths. For Fig. 8.14(c) the
8.2. DISCUSSION: HYPERBOLIC BAND APPROXIMATION

Figure 8.15: Landau level spectra for different couplings. Except $c_3$ and $c_4$ all parameters are chosen as listed in Table 8.1. For $c_3 = 10$ meV and a negative $c_4 = -2$ meV in the left plot, the model hosts Weyl cones as shown in Fig. 8.11 (b,c). In the middle plot we have $c_3 = c_4 = 0$, corresponding to the case without spin-orbit coupling and Dirac cones only. Here, notably the lowest transition is not split. The spectrum looks overall very similar, reinstating the little effect the spin-orbit coupling has on the spectrum in accordance with expectations. The right-hand figure with a positive $c_4 = -2$ meV does not host Weyl cones, but is slightly gapped, compare Fig. 8.14. The lowest line is pushed further down. Their slopes are even more agreeable with the measured features [110, 111].

band structure has been plotted over two dimensions in $k$-space in (f), comparing qualitatively well with the model in (g). Note, however, that since along this path there are no Weyl points, and in fact no band intersections, the model parameter $c_4$ has been chosen positive here, to produce small gaps instead. This change barely affects
the Landau level spectrum, as illustrated in the plot of the magneto-optical reflectivity $R(B)/R(0)$ in Fig. 8.14(a). A direct comparison of the magneto-optical reflectivity spectra for positive and negative $c_4$ is provided in Fig. 8.15. Further, a spectrum for $c_4 = 0$ has been calculated. We see that a finite but small $c_4$ leads to the splitting of the lowest transition line and, even though it is small, cannot be neglected.

8.3 Conclusions

To conclude, NbAs, a member of the TaAs-family and a Weyl semimetal was optically investigated in magnetic fields up to $B = 13$ T in reflection infrared spectroscopy. A previous magneto-reflectivity study modelled the Landau-level spectrum using both types of the Weyl cones $W_1$ and $W_2$ present in the band structure [99]. We noted though, that this approach is likely not valid, due to the stark difference in velocities $v_z$ and the associated difference in the density of states. Thus, only the features of one cone could be visible at a time. Nonetheless, the reflection spectra show non-linear Landau levels and we found that we can model these successfully with an $8 \times 8$ model Hamiltonian based on gapped Dirac cones with hyperbolic band dispersions. However, while the model is able to host pairs of Weyl cones, we noted that due to their negligible size, they do not qualitatively affect the spectrum. Extensive band analysis was undertaken using the Wien2k package. We found appropriate sections of the band structure, with high densities of states, which are positioned around the perimeter of the nodal lines in the spin-orbit-less band structure. With spin-orbit coupling taken into account, these nodal lines split, but the dispersion of electronic bands around former nodal lines remains slow. We found that paths near the $\Sigma$-N-$\Sigma$ high-symmetry line are well in line with the energy bands of the suggested model Hamiltonian.
8.3. CONCLUSIONS
GdPtBi is a so-called half-Heusler compound. Heusler compounds have a special relationship to magnetism. The first alloys of their kind were presented in 1903 at the "Verhandlungen der Deutschen Physikalischen Gesellschaft" [114] for their unusual magnetic properties. Friedrich Heusler had observed that various bronzes, meaning alloys of copper, had shown ferromagnetic behavior even though none of its constituents where originally ferromagnetic. Specifically, he investigated compounds of composition Cu$_2$MnX, where X stands in for the metals Sn, Al, As, Sb, Bi and B. Alloys with a X$_2$YZ composition, where X and Y are transition metal elements and Z is an main-group element of groups III to V, are thus called Heusler compounds. The material GdPtBi is part of the half-Heusler compound family, which is an extension to the ordinary or full Heuslers and are described by the composition XYZ [115]. The least electronegative element X, here Gd, transfers charges onto the main group element Z, here Bi. Apart from the outer-shell electrons, which Gd gives up, it hosts an exactly half-filled 4f-shell with seven free electron spins and thus has a rich magnetic phase diagram [61].
Figure 9.2: Band inversion strengths for a variety of different Half-Heusler compounds. GdPtBi, not shown, is deep in the “negative band gap” regime with an inverted band structure. The irreducible representations $\Gamma_6$ and $\Gamma_8$ refer to two subsets of bands that are two- and four-fold degenerate at the $\Gamma$-point. Due to the shape of the $\Gamma_8$ in the inverted regime, the topological half-Heuslers are semimetallic [116, 117].

The half-Heuslers have a vast amount of family members, which can be virtually continuously tuned between trivial insulators and topological semimetals. This can be seen in Fig.9.2 [116], where a series of Half-Heusler crystals and the gaps between the bands of the irreducible representations $\Gamma_6$ and $\Gamma_8$ are shown. In the trivial regime, this gap between the $\Gamma_6$ and $\Gamma_8$ bands at $\Gamma$ is defined positive, situating the $\Gamma_6$ above the $\Gamma_8$ band. In the topological regime it defined negative with the $\Gamma_6$ bands now situated below the $\Gamma_8$ bands and the $\Gamma_8$ bands tending to form parabolic touching points. The
CHAPTER 9. GdPtBi - A FIELD-TUNABLE WEYL SEMIMETAL

topological sector consists of compounds in which a strong spin-orbit coupling is present. Since GdPtBi contains fairly heavy atoms, it is deep within the topological regime.

GdPtBi had become prominent recently for a new type of band intersection, shown in Fig. 9.3. Here, two bands of the $\Gamma_8$-representation are degenerate along the $L - \Gamma$ high-symmetry line and dip below one of the other two spin-split bands and intersect with it not far from $\Gamma$-point. This intersection, between a singly degenerate band and a doubly degenerate band has been coined the triple point $[118–121]$. Similar to Weyl cones, they are topological in nature and give rise to Fermi arcs $[120]$. But as Fig. 9.3 shows, they live on a very small energy scales and are difficult to detect experimentally. Though, what makes them especially interesting to us is that the spin-degenerate band may split in magnetic fields, forming two Weyl points from each triple point $[60, 61, 122]$. Thus, investigating these could be a worthwhile endeavor, since the realm of magnetism is governed by the exchange-interaction, which can be magnitudes larger than the spin-orbit coupling. This is why time-reversal broken Weyl semimetals can potentially give rise to large and magnetically tunable Weyl cones, unlike inversion broken materials. Though so

![Figure 9.3: The Brillouin zone of GdPtBi with high-symmetry point labels and the triple point intersection on the high symmetry line $\Gamma - L$ [118].](image)

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far, however, only magneto-transport measurements on GdPtBi exist. In this Chapter, we are going discuss our contribution to the literature with our magneto-optical data.

9.1 Measurements

Given the strong sample dependency concerning GdPtBi [60], we should point out that the measurements discussed in the present Chapter were done on the same sample that was used in a previous study to obtain the zero-field broadband optical data [118]. Temperature-dependent resistivity of GdPtBi has been remeasured and is in accordance with the previous study [118], see 9.4, indicating that the sample remained intact. The sample becomes less resistive as the temperature increases, which is consistent with a semimetal and is in line with a low carrier density. Further, a kink at low temperatures can be observed, consistent with the antiferromagnetic phase transition at a Néel temperature of $T_N = 8.3$ K.

![Resistivity of GdPtBi measured over temperature. The low temperature kink marks the antiferromagnetic transition at $T_N = 8.3$ K.](image)

Figure 9.4: Resistivity of GdPtBi measured over temperature. The low temperature kink marks the antiferromagnetic transition at $T_N = 8.3$ K.
Infrared spectroscopy measurements of GdPtBi in magnetic fields are shown in Fig. 9.5. Unlike in the previous cases of TaP and NbAs, the spectrum does not have the characteristic of Landau level fans. The magneto-optical data was collected in a reflectivity setup from a (111)-oriented facet with the area of roughly 2 by 1.1 mm$^2$. The sample was kept at $T = 4.2$ K in helium exchange gas during the in-

Figure 9.5: Magneto optical measurements in Faraday configuration with $B || [111]$. Stacked plot of the relative reflectivity $R(B)/R(0)$ 1 T steps to the left and color plot of $R(B)/R(0)$ to the right. The measurement has been performed at liquid Helium temperature $T = 4.2$ K [1].
9.1. MEASUREMENTS

Figure 9.6: Relative reflectivity $R_B/R_0$ for two temperature points above the Néel temperature of $T_N = 8.3$ K.

field measurements, which is below the Néel temperature, $T_N = 9$ K. The sample holder was placed in a superconducting coil, which provided magnetic fields up to 16 T. We employed the Faraday configuration, with the magnetic field aligned along the crystallographic direction [111], meaning that the measurement probes the crystallographic plane (111). The experimental results of Fig. 9.15 (a) show the stacked plot of the in-field reflectivity curves normalized to the zero-field spectrum, $R(B)/R(0)$, in 1 T steps. Just above the lower limit of our measurement range ($\approx 4$ meV), a strong peak emerges, increasing in intensity and width roughly linearly propor-
tional to $B$. Another broadening, though less intense feature emerges at higher energies, marked by the transparent orange overlay. Two sharp constant-in-energy feature around 14 meV and 18 meV mark the presence of two phonons, which will not be topic of the main discussion. At 45 meV, a small discontinuity indicates the merge of two spectral ranges in our measurements. Wedged between the broad peaks, another sharp peak disperses just below the orange overlay. The Faraday configuration is typically employed to measure Landau levels, but unlike the previous measurements on TaP and NbAs, the spectrum does not bear the characteristics of typical Landau-level transitions. These, as we saw, express themselves in rich series of lines [34, 35, 123, 124], whereas here we mainly observe two large broadening features. We find it is more apt to describe the spectrum via as the result of an effective Zeeman splitting with a large $g$-factor. This splitting occurs due to large band shifts caused by the exchange interaction, as the free spins of Gd start to align under the application of an external magnetic field.

Under the application of a magnetic field, the antiferromagnetically aligned spins smoothly cant into a ferromagnetic order [61, 125]. Additional measurements were taken above $T_N$, shown in Fig. 9.6. To elevate the temperatures to $T = 20$ K and $T = 50$ K, a heating element was attached to the sample holder. Note that for this configuration, the measurement was taken only up to 45 meV. The spectra are not qualitatively much different from the $T = 4.2$ K measurement in Fig. 9.5, indicating that the antiferromagnetic spin alignment at $T = 4.2$ K does not change the interpretation of the results. This is consistent with the smoothly linear magnetization curve of [61] below the $T_N$, indicating “soft” spins that begin canting out of the antiferromagnetic alignment for low magnetic fields.

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9.2 Discussion - Field-Tunable Exchange Gap

Before interpreting our data, we should first expand on existing literature here. In the publication of Hirschberger et al. [60] magneto-transport data of GdPtBi along the [111] crystallographic axis is presented. Along this axis, GdPtBi hosts large Weyl cones if the spins align along [111], which can be achieved by applying an external magnetic field in parallel to [111]. The occurrence of these Weyl cones, it is argued, explains the occurrence of a large negative magneto-resistance measured along the applied magnetic field. This result is in line with the presence of the chiral anomaly, which leads to a current proportional to \( j \propto E||B \), as discussed in Section 3.2.1. A simplified band structure with purely parabolic bands is presented, as reproduced in Fig. 9.7, to explain the behavior of the band structure in magnetic fields. The exchange interaction, which splits the bands near the \( \Gamma \)-point acts as an effective Zeeman splitting. Since the four bands of \( \Gamma_8 \) belong to a \( J = 3/2 \) representation, the Zeeman splitting proportional to a term \( H_{\text{Zeeman}} = -g^* \mu_B 3/2 \cdot B \) is introduced, where the two hole bands split proportional to \( m_J = \pm 3/2 \) and the electron bands with \( m_J = \pm 1/2 \).

Except for a bump at the \( \Gamma \)-point, this agrees well with the DFT band structure, shown in Fig. 9.8. In this calculation the spins have been aligned along [111] and the 4f-electrons have been localized by introducing an on-site Coulomb parameter of \( U = 10 \) eV [126–128] in accordance with existing literature [61]. The bands at the \( \Gamma \)-point, which are degenerate if no field is applied, are separated along the \( L - \Gamma - L \)-line, apt for a description with an effective Zeeman splitting. The band-structure cut including two \( k \)-space dimensions on the right of Fig. 9.8 illustrates that the band intersections indeed occur in the form Weyl cones.

We retain that their idea is mostly in agreement with our results.
and will illustrate how we can replicate the character of the measured spectra using parabolic models with Zeeman terms. To keep the complexity low at this point, we consider a simple two-by-two Hamiltonian of the form

$$H = H_0 + (m_{s,↑} \sigma_{↑}^Ω + m_{s,↓} \sigma_{↓}^Ω) \Delta,$$

(9.1)

$$H_0 = -\frac{\hbar^2}{2m} (q_x^2 + q_y^2) \sigma_z + \frac{\hbar^2}{2m_x} q_x^2 \sigma_x.$$

(9.2)

Here, the Hamiltonian $H_0$ describes two isotropic parabolic bands of mass $m$ coupled by a term $\propto q_x^2$ (we remind that $q_i = k_i/a_i$, where $a_i$ are the lattice). The term $(m_{s,↑} \sigma_{↑}^Ω + m_{s,↓} \sigma_{↓}^Ω) \Delta$ in $H$ displaces the bands relative to each other, representing the Zeeman term. The notations $\sigma_{↑}^Ω = \frac{1}{2}(\sigma_0 + \sigma_z)$ and $\sigma_{↓}^Ω = \frac{1}{2}(\sigma_0 - \sigma_z)$ are used to express shifts of the bands in the separate spin subspaces of the model. The terms $m_{s,↑} = \frac{1}{2}, \frac{3}{2}$ and $m_{s,↓} = -\frac{1}{2}, -\frac{3}{2}$ will refer to the effective spin eigenvalues of the bands. The band with positive spin has negative

![Figure 9.7: Simplified band structure description of GdPtBi along the [111] direction without magnetic field on the left, and with magnetic field along [111]. The band separation in fields can be described effectively as a Zeeman splitting with magnetic quantum numbers $m_j = \pm 1/2, \pm 3/2$.](image)
9.2. DISCUSSION - FIELD-TUNABLE EXCHANGE GAP

Figure 9.8: Band structure of GdPtBi with spins aligned along [111]. The left cut traverses along the high-symmetry points $L - \Gamma - L$, which is parallel to $k_{[111]}$. The right cut is extended by $k_{[1\bar{T}]}$ [129, 130].

dispersion and vice versa for the negative spin, due to the minus sign in $H_0$. The remaining term $\Delta = g\mu_B B_z$ captures the field-scaling of the Zeeman term. If we neglect the term $\propto q^2_\sigma x$ for now, then a positive magnetic field shifts the parabolic bands into one another, forming an uncoupled nodal line. By introducing the term $\propto q^2_\sigma x$, the bands are coupled and the nodal line gaps out with the exception of two points, as shown on the left of Fig. 9.9. As the magnetic field is raised and the overlap of the parabolas grows larger, the coupling between the bands increases. This is well in line with the idea put forth by Ref. [60] and produces field-tunable Weyl points as seen in Fig. 9.8, in a simplified fashion. Now, we want to investigate the model’s optical response using Eq. (4.43). If we take spin eigenvalues $m_{s=1/2,\uparrow} = \frac{1}{2}$ and $m_{s=1/2,\downarrow} = -\frac{1}{2}$, the electron and hole band shift in equal amounts. Pinning the Fermi level at $E_F = 0$, coinciding with the touching point of the parabolic cusps at $B = 0$, we get the optical response shown on the right of Fig. 9.9. The optical
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\[ H = H_0 + \left( \frac{1}{2} \sigma^\uparrow - \frac{1}{2} \sigma^\downarrow \right) \Delta \]

Figure 9.9: Optical conductivity calculated for the Hamiltonian in Eq. (9.1) with \( s^\uparrow = \frac{1}{2} \) and \( s^\downarrow = -\frac{1}{2} \) within a range \( \Delta(B) \in [0,30 \text{ meV}] \) relative to the optical conductivity at \( \Delta = 0 \). With the choice of \( g = 32.4 \) this corresponds to the range of measurement \( B \in [0,16 \text{ T}] \). The scattering rate was chosen as \( \gamma = 5 \text{ meV} \).

conductivity shows a feature that grows linearly in \( B \) as the Zeeman shift gradually overlaps the bands with an increasing field. Since the Fermi level rests steadily in between the emerging cones, this triangle-shaped feature of increased optical activity emerges from \( E = 0 \) for all fields. Conversely, if the spin eigenvalues are chosen asymmetrically as \( m_{s=3/2,\uparrow} = \frac{3}{2} \) and \( m_{s=1/2,\downarrow} = -\frac{1}{2} \), the intersection point of the two bands shifts relative to the Fermi level and an interband-transition gap opens up. The increased relative optical activity now emerges at a finite energy in an applied field. This situation is illustrated in Fig. 9.10. This approach seems promising, considering the shape of the measured features in Fig. 9.5 (b). We would now like to extend this model to account for four bands.
9.2. DISCUSSION - FIELD-TUNABLE EXCHANGE GAP

\[ H = H_0 + \left( \frac{3}{2} \sigma_\uparrow - \frac{1}{2} \sigma_\downarrow \right) \Delta \]

Figure 9.10: Optical conductivity calculated for the Hamiltonian in Eq. (9.1) with \( s_\uparrow = \frac{3}{2} \) and \( s_\downarrow = -\frac{1}{2} \) within a range \( \Delta(B) \in [0, 30 \text{ meV}] \) relative to the optical conductivity at \( \Delta = 0 \). With the choice of \( g = 32.4 \) this corresponds to the range of measurement \( B \in [0, 16 \text{ T}] \). The scattering rate was chosen as \( \gamma = 5 \text{ meV} \).

Here, we should note, however, that the band structure discussed so far included the [111] crystal direction, which corresponds to the \( L - \Gamma - L \) line. Our measurement on the other hand, was taken in the plane orthogonal to [111]. The respective plane in the Brillouin zone, not containing [111], is illustrated in Fig. 9.11 (a). In Fig. 9.11 (b) the associated two-dimensional band structure cut is illustrated for \( B = 0 \). This zero-field band structure is indeed convincingly parabolic, however, the in-field band structure with fully aligned spins is rather complex, see Fig. 9.12. Within the (111) plane, a gap between the electron and hole bands forms. Previously, these bands had intersected in Fig. 9.8 to give rise to Weyl cones along [111]. It is worth noting that even though the band shifts are
large, the change in the Fermi surface is negligible, see Fig. 9.13. This indicates that the amount of free charge carriers does not increase drastically and its contribution to the optical spectrum is expected to remain marginal. In the zero-field measurements [118], the screened plasma-frequency is found to be around 4 meV. Thus, we can assume that the measured spectra mainly reflect the interband contributions.

In order to model this band structure, we assume the parabolic bands of Fig. 9.11 to move as indicated with the dashed-red lines in Fig. 9.12(b), due to the effective Zeeman shift. The gaps between the electron and hole bands are produced by off-diagonal coupling terms in the model Hamiltonian. The coupling should be chosen such that at zero field, without any Zeeman displacement of the bands, the four-fold degeneracy at the $\Gamma$ point is reproduced, in line with Fig. 9.11. As before, this can be achieved by using $\mathbf{q}$-dependent cou-
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Figure 9.12: (a) One-dimensional band structure cut along $K-\Gamma-K$ (see Fig. 9.11) and (b) the two-dimensional band structure of the (111) plane with the spins fully aligned along [111]. Exchange interaction due to spin alignment splits the bands. Dark-red dashed schematic bands have been overlayed to indicate the effective Zeeman splittings that our toy model is based on [129, 130].

Linear couplings will skew the model unreasonably, so we will constrain ourselves to $q^2$ terms. The model has been constructed as

$$H = \begin{pmatrix}
\frac{\hbar^2 q^2}{2m_e} + \mu B & \frac{1}{2} Y_- & \frac{1}{2} Y_+ & 0 \\
\frac{1}{2} Y_- & \frac{\hbar^2 q^2}{2m_h} + \frac{1}{2} B & Y_- & -\frac{1}{2} Y_+ \\
\frac{1}{2} Y_+ & Y_- & \frac{\hbar^2 q^2}{2m_h} + \mu B & \frac{1}{2} Y_- \\
0 & -\frac{1}{2} Y_+ & \frac{1}{2} Y_- & \frac{\hbar^2 q^2}{2m_e} + \mu B
\end{pmatrix} + \Delta B. \quad (9.3)$$

On the diagonal, we find the two pairs of parabolic bands describing electrons and holes with effective masses $m_e$ and $m_h$. Effective Zeeman terms $\mu B$ split the degenerate bands, intersecting an electron band with both hole bands, forming nodal lines, see Fig. 9.14 (b). We adhered to the spin-$3/2$ picture, and thus $\mu_{m_s} = m_s g^* \mu B$ with $m_s = \pm \frac{3}{2}, \pm \frac{1}{2}$. The assignment is consistent with cuts along [111].
where the lowest and highest band at the $\Gamma$ point (center of Fig. 9.12) correspond to the spins $s_z = \pm 3/2$, though the role of hole and electron bands has interchanged. The off-diagonal terms
\begin{equation}
Y_\pm = -\frac{\hbar^2}{2m_{xy}} \left(q_x^2 \pm q_y^2\right),
\end{equation}

(9.4)
couple the bands, qualitatively reproducing the essential band structure features. Thereof $Y_+$ has cylindrical symmetry, gapping out the electron and hole bands of the model as shown in Fig. 9.14 (c). The terms $Y_-$ partially gap out the remaining nodal intersection between the two hole bands, with the exception of four points in the $k_x$-$k_y$-plane, where the coupling $Y_-$ is zero and leads to point like intersections. At the same time it reproduces the flanks around the band structure and the protrusions of the lower electron band encircling the central hole pocket, as well as the flower-like and star-like cross sections in the hole and electron bands, compare Fig. 9.12. A good agreement between theory and experiment was found for the parameters
\begin{align*}
m_e &= \frac{3}{2} m_0, \quad m_h = -\frac{1}{4} m_0, \quad m_{xy} = \frac{1}{2} m_0, \\
\Delta &= 1.4 \, g^* \mu_B, \quad \mu_{\pm \frac{3}{2}} = \frac{3}{2} \, g^* \mu_B, \quad \mu_{\pm \frac{1}{2}} = \frac{1}{2} \, g^* \mu_B
\end{align*}

where $m_0$ is the free electron mass and $\mu_B$ is the Bohr magneton. The magnetic momenta $\mu_{\pm \frac{1}{2}, \frac{3}{2}}$ lead to an effective $g^*$ of 95,
expressing the shift of the bands relative to another. The shift \(\Delta B\) displaces all of the bands simultaneously. Fig. 9.15 compares the measured reflectivity (a) with the reflectivity obtained from the model Hamiltonian (b). Note, that a free-carrier cyclotron response has been added to the conductivity of the modelled Hamiltonian, according to the standard optical formulas [70]. We chose a plasma-frequency of \(\omega_{\text{pl}} = 3.5\) meV, the free-carrier (Drude) scattering rate \(\gamma_D = 3\text{cm}^{-1} = 0.38\) meV and \(\varepsilon_\infty = 80\), in consistency with [118]. The interband scattering rate was set to \(\gamma = 2\) meV. We find that
Figure 9.15: (a): Magneto-optical reflectivity of GdPtBi in the (111) plane at $T = 4.2K$ divided by the zero field reference, showing relative changes in the reflectivity as the magnetic field $B$ varies. (b): Calculated relative magneto optical response using the four band Hamiltonian, Eq. (9.3), with $\varepsilon_\infty = 40$ and $\gamma = 2.0$ meV. (c) and (d): The bands of the four band Hamiltonian for the values $B=0$ T hosting a closed gap, and at $B=10$ T [129, 130].

The model reproduces the experimental data very well. Estimates for the effective masses obtained for Shubnikov-de Haas oscillations range between $m^* = 0.23 \ m_0$ [60] to $m^* = 0.30 \ m_0$ [62] and are in line with our estimate for the hole masses $m_h$. Interestingly, a weak-$B$ estimate with a slight electron doping, $E_F = 3.1$ meV, yields an effective mass of $m^* = 1.8 \ m_0$ [60], which is consistent with our estimated electron band mass $m_e$. From the band-structure calculations we can furthermore extract an estimate for the $g$-factors. At full spin-alignment, the energy gap between the $m_s = \pm 3/2$ is roughly 400 meV, thus $E_{Zee, m_s=\pm \frac{3}{2}} \approx \pm 200$ meV. From [61] we know that full spin-alignment is reached at $B = 25$ T, which yields
Figure 9.16: From left to right: the evolution of the band gap between the electron and hole bands of the model Hamiltonian for $B = 10$ T along $k_z$. Beyond the band-touching point, the electron- and hole band pairs disperse $\propto k_z^2$.

$g^* = E_{Zee,m_s}/\mu_B m_s B \approx 92$, which is in good agreement with our model. Note that the optical interband response has been calculated over all three $k$-space dimensions. As the band structure progresses along $k_z$, the band coupling is effectively undone when going far from $k_z = 0$, see Fig. 9.16. As $k_z$ increases, the bands start to gap out with proportional to $k_z^2$, such that most of the optical conductivity is contributed near $k_z = 0$. The $k_z$ dispersion mainly smooths out the optical response. To understand the origin of the transitions in detail, the $k$-dependent optical conductivity

$$\sigma_{2D}^{\alpha,\beta,m,n} = \frac{1}{\sigma_{all}} \sum_{\omega,\{m,n\}} \frac{f_m - f_n}{E_{m,k} - E_{n,k}} \frac{\langle m|\hat{v}_{\alpha}|n\rangle \langle m|\hat{v}_{\beta}|n\rangle}{E_{n,k} - E_{m,k} - \hbar \omega + i\gamma}, \quad (9.5)$$

in two $k$-space dimensions for transitions bands $m$ to bands $n$, with $\sigma_{2D}^{\alpha,\beta,m,n} = \sigma_{2D}^{\alpha,\beta,m,n}(k_x, k_y, B)$. The notation $\{m, n\}$ below the sum should express that we may add any custom sets interband transitions. The result is then normalized by the $\sigma_{all}$ that sums over all possible transitions. In particular, we are interested in the added transitions to a specific band. Say, we want to investigate the op-
tical contributions to band 3. Then, we add over transitions from band 1 to 3 and band 2 to 3, and thus we would define \( m \in 1, 2 \) and \( n = 3 \) in the sum. We can map the resulting optical conductivity of every band onto its dispersion, as shown in Fig. 9.17. Here, every colored band displays the optical conductivity of all possible transitions to this same band. Since no transitions to the lowest band are possible, it is not depicted in color. The subfigures in Fig. 9.17 were calculated on a \( k_x-k_y \) mesh of size 400 \( \times \) 400 for \( k_z = 0 \) and \( B = 10 \) T.

We now see that the strongest transitions arise at the gapped nodal lines between the Weyl points and give rise to the strong low-energy feature in the relative reflectivity \( R_B/R_0 \) in Fig. 9.15. The broader high-energy feature originates from the remaining transitions, which are practically only to the lower electron band. At \( B = 10 \) T, the higher electron band is already too far removed to couple significantly to the hole bands.

9.3 Conclusions

Experimental studies of the magneto-optical reflectivity were performed on the triple-point semimetal GdPtBi. Unlike the cases of TaP and NbAs, in this compound we find no evidence for Landau levels. Instead, two broad maxima emerge in the relative reflectivity \( R(B)/R(0) \) under the application of an external magnetic field. In accord with our DFT calculations and with previous publications, we argue that we measure the optical signature of effective Zeeman shifts of the electronic bands in GdPtBi. These band shifts are mediated by the exchange-field as the soft spins of the Gd 4\( f^7 \) electrons align in applied magnetic fields. We propose a four-band Hamiltonian, built from two coupled pairs of parabolic hole and electron bands. Previous magneto-transport studies exist [60, 61], providing measurements along the [111] crystallographic direction in which the presence of large Weyl cones (>100 meV in size) is expected in mag-
Figure 9.17: The $\mathbf{k}$-resolved optical conductivity for the bands of the model Hamiltonian for $B = 10$ T. Each colored band contains all possible transitions to this band. Thus, a) contains transitions from band 0 to band 1, b) contains transitions from bands 0 and 1 to band 2, and c) contains transitions from band 0 and 1 to band 3.

The half-Heusler compound GdPtBi may prove useful in future studies as its large, field-tunable band shifts provide fine control over the size of Weyl cones. Further, since the band structure behaves qualitatively very different depending on the direction the external magnetic field is applied, the number, relative positions, and other characteristics may be tuned [122]. In conjunction with superconductors, interfaces with such exchange-driven Weyl semimetals may open up new avenues in the pursuit of creating braidable Majorana fermions.
With the recent interest in the novel type of physics harbored by topological materials, we set out to investigate the compounds TaP, NbAs and GdPtBi in this thesis. They belong to a subset of topological phases called the Weyl semimetals, which host not only characteristic features on the material surface, the Fermi arcs, but also show interesting bulk electron behavior. Their bulk electronic band structure is marked by the presence of linearly dispersing Weyl cones, hosting massless and spin-polarized electrons. This leads to a characteristic \( E \propto B \) Landau-level scaling and makes magneto-optical infrared spectroscopy an ideal tool to investigate this type of materials. Rather than these prototypical Landau-level spectra, however, the experiments performed in the course of this work had shown some unexpected behavior, requiring deeper analysis.

Starting with TaP, we have seen the footprint of the band inversion in the Landau-level transition spectrum. For Weyl cones to exist, it is necessary that two overlapping bands, which would form a nodal line in the spin-less band structure, are coupled via the spin-orbit interaction. If inversion symmetry is broken, this coupling can cause the nodal line to gap out, with the exception of a few nodal points, the Weyl nodes. A popular approach to describe these Weyl points, is a linear approximation of the bands, which then represent ideal Weyl cones. However, this research is an example case for
why the simple linear approximation is not always a valid approach. Rather than Landau levels with obvious $E \propto \sqrt{B}$ scaling, the measured spectrum most prominently shows a strong signature of the overlapping bands, expressed by the Landau level transitions scaling downwards in energy with increasing magnetic field. This moves the focus away from the Weyl points and requires a more macroscopic view of the band structure. To that end, we have proposed a model Hamiltonian, which describes two intersecting parabolic bands coupled appropriately to yield linear band intersections. The calculated Landau-level spectrum reproduces the experiment well and confirms the origin of the downwards scaling transition energies in the inverted band structure of the intersecting bands that give rise to the Weyl points. For an uncoupled nodal line, it is clear that the gaps between Landau levels of the oppositely dispersing bands reduces with magnetic fields, until they undergo a Lifshitz transition at high enough fields. Due to the coupling that gaps out the nodal line, optical transitions between the bands become allowed, and finally Landau-level transitions with decreasing energy can be observed.

Conversely, in the case of NbAs, which also belongs to the TaAs-family, the effect of the band inversion is not obvious in the measured Landau-level spectrum. On the contrary, mostly a dense set of curved transition lines is visible. It is immediately clear that the spectrum fits neither the $E \propto B$ scaling massive Landau levels, nor the $E \propto \sqrt{B}$ scaling of massless Landau levels. A previous study had measured a similar spectrum on the same compound, though asserted that it is the combined result of the two different types of Weyl cones present in the TaAs-family compounds, labelled $W_1$ and $W_2$, which would explain the large number of transitions. However, the band velocities along the $k$-direction parallel to the applied magnetic field, which we identify as $k_z$, are starkly different for the two types of Weyl cones, by more than an order of magnitude. With this, also the density of states for the two cones should be vastly different and thus, as we have shown explicitly, it is unlikely that both Weyl
cones can be observed simultaneously. The $W_1$ transitions should strongly dominate the spectrum and mask all transitions from $W_2$. Another point was the small extent of the Weyl bands due to the much smaller spin-orbit coupling in the Nb-containing compounds. DFT calculations estimate them to be on the order of $\approx 20 \text{ meV}$, whereas the measured transition lines reach beyond $100 \text{ meV}$, which seems to be a clear contradiction. Thus, we argued that the Weyl cones are negligible in size and do not contribute significantly to the measured Landau levels. Another way to obtain the required number of Landau levels is to use gapped Dirac cones, which are hyperbolic in shape. Alongside detailed band structure calculations, a model was built from intersecting hyperbolic bands, emulating the character of the electronic bands. The fitted model provides a very good match with the experimental results and estimates the size of the Weyl cones to be even smaller than those obtained in DFT calculations.

Finally, we investigated the triple-point semimetal GdPtBi. The compound is known to be antiferromagnetic below $T = 9 \text{ K}$ and the transition can be observed in temperature-dependent measurements of the electrical resistance. However, magnetization measurements suggest that the antiferromagnetic order is broken at small fields and the compound behaves essentially paramagnetic. Band structure calculations indicate that the triple points, which GdPtBi hosts, split into Weyl points if a magnetic field is applied. For a magnetic field along the [111] crystallographic direction, the electronic bands along the magnetic field move in energy in a manner that reminds of a Zeeman effect for an effective Hamiltonian of a spin $S = 3/2$ fermion. In reality, this is of course caused by the exchange interaction. Though, we can make use of an effective Zeeman term as a means to simplify the mathematical description. We set out to measure the Landau levels of such magnetically tunable Weyl cones, however, the magneto-optical infrared spectrum instead shows mainly two peaks strongly broadening with $B$. The spectrum
can be reproduced with a model Hamiltonian, which is based on coupled parabolic bands, which are tuned by the effective Zeeman term. As the bands move and cross one another in the applied field, the $k$-dependent off-diagonal terms lead to a stronger coupling between some of the bands. In turn, the optical transitions between the crossing bands become more intense, reproducing the measured features well. With this approach, we can associate the bands with an effective $g$-factor of 92, well in line with previous studies, and provide the first magneto-optically recorded trace of this effect. Due to its magnetically tunable topology, GdPtBi holds great potential for future research and technical applications.
Part IV

Appendix
Appendix

A

LANDAU LEVEL WAVEFUNCTIONS FOR WEYL FERMIONS

In Section 3.2 we derived the Landau level spectrum for Weyl electrons. Here, the calculation of the wave functions is provided. We had set up the auxiliary equation

\[ \hat{H}_{\text{aux}} \phi_n(x, y, z) = E^2 \phi_n(x, y, z), \quad (A.1) \]

which is solved by the wave function

\[ \phi_n(x, y, z) = \varphi_n(w) e^{ik_y y + ik_z z} \frac{\sqrt{L_y L_z}}{\chi_{m_s}}. \quad (A.2) \]

where we remind that

\[ w = \frac{x - x_0}{l_B}, \quad x_0 = -\frac{\hbar k_y}{eB}. \quad (A.3) \]

The eigenenergies were given by

\[ E = \pm \sqrt{\hbar^2 \omega_D^2 (n + \frac{1}{2} + m_s) + v^2 \hbar^2 k_z^2}, \quad (A.4) \]

where \( m_s = \pm 1/2 \) and

\[ \omega_D^2 = 2v^2 eB/\hbar. \quad (A.5) \]
The $\varphi_n(w)$ are the Hermite functions
\[ \varphi_n = \frac{1}{\sqrt{2^n n!}} \pi \frac{1}{4} e^{-\frac{w^2}{2}} H_n(w) \tag{A.6} \]
which fulfill
\[ \partial_w \varphi_n(w) = \sqrt{\frac{n}{2}} \varphi_{n-1}(w) - \sqrt{\frac{n+1}{2}} \varphi_{n+1}(w) \tag{A.7} \]
\[ w \varphi_n(w) = \sqrt{\frac{n}{2}} \varphi_{n-1}(w) + \sqrt{\frac{n+1}{2}} \varphi_{n+1}(w), \tag{A.8} \]
where $\varphi_{-1} = 0$. We had established that the wave functions $\psi_R$ and $\psi_L$ of the Weyl electrons can be found by calculating
\[ \psi_{R,n,m_s} = (E + c\sigma(\hat{p} + e\hat{A}))\phi_{n,m_s} \tag{A.9} \]
\[ \psi_{L,n,m_s} = (E - c\sigma(\hat{p} + e\hat{A}))\phi_{n,m_s}. \tag{A.10} \]
In Eqs. (A.9) and (A.10) we have terms of the form $\partial_x \varphi_n(w)$ and $x \varphi_n(w)$. With Eqs. (A.7) and (A.8) we find
\[ \partial_x \varphi_n(w) = \frac{1}{l_B} \partial_w \varphi_n(w) = \frac{1}{l_B} \left( \sqrt{\frac{n}{2}} \varphi_{n-1}(w) - \sqrt{\frac{n+1}{2}} \varphi_{n+1}(w) \right) \tag{A.11} \]
\[ x \varphi_n(w) = (x_0 + l_B w) \varphi_n(w) \tag{A.12} \]
\[ = x_0 \varphi_n(w) + l_B w \left( \sqrt{\frac{n}{2}} \varphi_{n-1}(w) + \sqrt{\frac{n+1}{2}} \varphi_{n+1}(w) \right) \tag{A.13} \]
Plugging these relations back into Eq. (A.9), we determine
\[ \psi_{R,n,m_s} = \mathcal{N}_R \left[ \sigma_x \frac{-i\hbar v}{l_B} \left( \sqrt{\frac{n}{2}} \varphi_{n-1}(w) - \sqrt{\frac{n+1}{2}} \varphi_{n+1}(w) \right) \right. \]
\[ - \sigma_y (\hbar k_y) \varphi_n(w) + \sigma_y \frac{\hbar v}{l_B} \left( \sqrt{\frac{n}{2}} \varphi_{n-1}(w) + \sqrt{\frac{n+1}{2}} \varphi_{n+1}(w) \right) \]
\[ + (E + \hbar k_y \sigma_y + \hbar k_z \sigma_z) \varphi_n(w) \left] \phi_{yz} \chi_{m_s}. \right. \]
APPENDIX A. LANDAU LEVEL WAVEFUNCTIONS FOR WEYL FERMIONS

The Pauli matrices act on the spinors as follows

\[ \sigma_x \chi_{m_s} = \chi_{-m_s}, \quad \sigma_y \chi_{m_s} = \pm i \chi_{-m_s}, \quad \sigma_z \chi_{m_s} = \pm \chi_{m_s}. \quad (A.14) \]

With that, we evaluate the normalization of \( \psi_{R,n,m_s} \), yielding

\[ |\psi_{R,n,m_s}|^2 = \left[ \left[ \chi_{-m_s} \cdot (\pm i \chi_{-m_s}) \right] \frac{v^2 \hbar^2}{l_B^2} \left( \frac{n}{2} |\varphi_{n-1}|^2 + \frac{n+1}{2} |\varphi_{n+1}|^2 \right) \right. \]

\[ + \left[ \chi_{-m_s} \cdot (\mp i \chi_{-m_s}) \right] \frac{v^2 \hbar^2}{l_B^2} \left( \frac{n}{2} |\varphi_{n-1}|^2 - \frac{n+1}{2} |\varphi_{n+1}|^2 \right) \]

\[ + \chi_{m_s} \cdot \chi_{m_s} E^2 + (\pm \chi_{m_s}) \left( \pm \chi_{m_s} \right) \left( E v_h k_z + (\pm \chi_{m_s}) \cdot \chi_{m_s} E v_h k_z \right) \]

\[ \left. + (\pm \chi_{m_s}) \cdot (\pm \chi_{m_s}) \left( \pm \chi_{m_s} \right) \right] \frac{v^2}{l_B^2} \left( \frac{n}{2} |\varphi_{n-1}|^2 - \frac{n+1}{2} |\varphi_{n+1}|^2 \right) \]

\[ + \left[ \left[ \chi_{-m_s} \cdot \chi_{-m_s} \right] \frac{v^2}{l_B^2} \left( \frac{n}{2} |\varphi_{n-1}|^2 + \frac{n+1}{2} |\varphi_{n+1}|^2 \right) \right. \]

\[ + \left[ \chi_{-m_s} \cdot (\mp i \chi_{-m_s}) \right] \frac{v^2 \hbar^2}{l_B^2} \left( \frac{n}{2} |\varphi_{n-1}|^2 - \frac{n+1}{2} |\varphi_{n+1}|^2 \right) \]

\[ \left. + \chi_{m_s} \cdot \chi_{m_s} E^2 + (\pm \chi_{m_s}) \left( \pm \chi_{m_s} \right) \right] \frac{v^2}{l_B^2} \left( \frac{n}{2} |\varphi_{n-1}|^2 + \frac{n+1}{2} |\varphi_{n+1}|^2 \right) \]

\[ \left. + (\pm \chi_{m_s}) \cdot (\pm \chi_{m_s}) \left( \pm \chi_{m_s} \right) \left( E v_h k_z \right) \right] \frac{v^2}{l_B^2} \left( \frac{n}{2} |\varphi_{n-1}|^2 - \frac{n+1}{2} |\varphi_{n+1}|^2 \right) \]

Thus the normalization constant \( N_R \) for the right-hand Weyl fermion is

\[ N_R = \left[ 2 \left( n + m_s + \frac{1}{2} \right) \frac{\hbar^2 v^2}{l_B^2} + (E - 2m_s v_h k_z)^2 \right]^{-\frac{1}{2}}. \quad (A.15) \]

For the left-handed wave function \( \psi_{L,n,m_s} \) we find a similar expression

\[ \psi_{L,n,m_s} = N_L \left[ \sigma_x \frac{i \hbar v}{l_B} \left( \sqrt{\frac{n}{2}} \varphi_{n-1}(w) - \sqrt{\frac{n+1}{2}} \varphi_{n+1}(w) \right) \right. \]

\[ + \left. \sigma_y \left( v_h k_y \right) \varphi_n(w) - \sigma_y \frac{\hbar v}{l_B} \left( \sqrt{\frac{n}{2}} \varphi_{n-1}(w) + \sqrt{\frac{n+1}{2}} \varphi_{n+1}(w) \right) \right] \]

\[ + \left( E - v_h k_y \sigma_y - v_h k_z \sigma_z \right) \varphi_n(w) \phi_{yz} \chi_{m_s}, \]

and the corresponding normalization constant \( N_L \) is

\[ N_L = \left[ 2 \left( n + m_s + \frac{1}{2} \right) \frac{\hbar^2 v^2}{l_B^2} + (E + 2m_s v_h k_z)^2 \right]^{-\frac{1}{2}}. \quad (A.16) \]

Note, that two chiral solutions with linear dispersions exist, which are \( \psi_{R,0,-\frac{1}{2}} = 0 \) with \( E = v_h k_z \) and \( \psi_{L,0,-\frac{1}{2}} = 0 \) with \( E = -v_h k_z \), see Ref. [23].

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Here, the Fourier transform of the conductivity tensor $\sigma$ into frequency space is provided. We consider a general auto-correlation function $\chi^{\hat{A}_\alpha \hat{A}_\beta}$ in terms of energy eigenstates $|n, k\rangle$. We suppress the $k$ in the following notation for brevity. Further, without loss of generality, we set $t_0 = 0$. Then

$$
\chi = \chi^{\hat{A}_\alpha \hat{A}_\beta}(t, t', k) = -\frac{i}{\hbar} \theta(t') \langle (\hat{A}_\alpha^I(t) \hat{A}_\beta^I(t') - \hat{A}_\beta^I(t') \hat{A}_\alpha^I(t)) \rangle_0
$$

$$
= -\frac{i \theta_t}{\hbar Z} \sum_{n, m} \langle n | e^{-\beta H_0} [\hat{A}_\alpha^I(t) | m \rangle_0 \langle m | \hat{A}_\beta^I(t') - \hat{A}_\beta^I(t') | m \rangle_0 \langle m | \hat{A}_\alpha^I(t) | n \rangle_0
$$

$$
= -\frac{i \theta_t}{\hbar Z} \sum_{n, m} \langle n | e^{-\beta H_0} \hat{A}_\alpha^I(t) | m \rangle_0 \langle m | \hat{A}_\beta^I(t') | n \rangle_0
$$

$$
+ + \frac{i \theta_t}{\hbar Z} \sum_{n, m} \langle n | e^{-\beta H_0} \hat{A}_\beta^I(t') | m \rangle_0 \langle m | \hat{A}_\alpha^I(t) | n \rangle_0
$$

$$
\overset{(4.5)}{=} -\frac{i \theta_t}{\hbar Z} \sum_{n, m} \langle n | e^{-\beta E_n} e^{-\frac{i}{\hbar} E_n t} A^S_\alpha e^{\frac{i}{\hbar} E_m t} | m \rangle_0 \langle m | e^{-\frac{i}{\hbar} E_m t} A^S_\beta e^{-\frac{i}{\hbar} E_n t'} | n \rangle_0
$$

$$
+ + \frac{i \theta_t}{\hbar Z} \sum_{n, m} \langle n | e^{-\beta E_n} e^{-\frac{i}{\hbar} E_n t'} A^S_\alpha e^{\frac{i}{\hbar} E_m t'} | m \rangle_0 \langle m | e^{-\frac{i}{\hbar} E_m t'} A^S_\beta e^{-\frac{i}{\hbar} E_n t} | n \rangle_0
$$
\[ \chi = -\frac{i\theta_{t'}}{\hbar Z} \sum_{n,m} e^{-\beta E_n} e^{\frac{i}{\hbar}(E_m-E_n)(t-t')} \langle n | A^S_\alpha | m \rangle_0 \langle m | A^S_\beta | n \rangle_0 
+ \frac{i\theta_{t'}}{\hbar Z} \sum_{n,m} e^{-\beta E_m} e^{\frac{i}{\hbar}(E_n-E_m)(t-t')} \langle n | A^S_\beta | m \rangle_0 \langle m | A^S_\alpha | n \rangle_0 \]
\[ = -\frac{i\theta_{t'}}{\hbar Z} \sum_{n,m} e^{-\beta E_n} e^{\frac{i}{\hbar}(E_m-E_n)(t-t')} \langle n | A^S_\alpha | m \rangle_0 \langle m | A^S_\beta | n \rangle_0 
+ \frac{i\theta_{t'}}{\hbar Z} \sum_{n,m} e^{-\beta E_m} e^{\frac{i}{\hbar}(E_n-E_m)(t-t')} \langle m | A^S_\beta | n \rangle_0 \langle n | A^S_\alpha | m \rangle_0 \]
\[ = -\frac{i\theta_{t'}}{\hbar Z} \sum_{n,m} (e^{-\beta E_n} - e^{-\beta E_m}) e^{\frac{i}{\hbar}(E_m-E_n)(t-t')} \langle n | A^S_\alpha | m \rangle_0 \langle m | A^S_\beta | n \rangle_0 \]

where the shorthand notation \( \theta(t') = \theta_{t'} \) was used. The time-dependent operator \( \hat{A}^I_\alpha(t) \) is defined in the interaction picture, whereas \( \hat{A}^S_\alpha \) is defined in the Schrödinger picture. Between these pictures, the following relation holds

\[ \hat{A}_\alpha(t) = e^{iH_0 t/\hbar} \hat{A}^S_\alpha e^{-iH_0 t/\hbar}, \] (B.1)

with the time-independent unperturbed Hamiltonian \( H_0 \) and where we assume the operators \( \hat{A}^S_{\alpha,\beta} \) to have no explicit time-dependency. Note, that the expectation values \( \langle ... \rangle_0 \) are explicitly taken at time \( t_0 = 0 \), which is equivalent in any picture. The operators \( \hat{A}_{\alpha,\beta} \) however, are evolved by Eq. (B.1). At the equal sign marked with \( (*) \), we insert Eq. (B.1) such that we can write all operators and states uniformly at \( t_0 = 0 \). This gives rise to the extra factors \( e^{\pm iE_{n,m}t/\hbar} \).

We write \( \omega_{n,m} = E_{n,m}/\hbar \) and define \( \omega_{nm} = \omega_n - \omega_m = -\omega_{mn} \). In Eq. (4.39) we wrote the modified Fourier transform as

\[ h(t) = \frac{1}{2\pi} \int_0^\infty H(\omega^+) e^{-i\omega^+ t} d\omega^+, \] (B.2)

\[ H(\omega^+) = \int_0^\infty h(t) e^{i\omega^+ t} dt, \] (B.3)

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containing a term \( \omega^+ = \omega + i\gamma \) to account for a decay of the response to the perturbation. We adjust the integration limits according to our choice \( t_0 = 0 \) by accounting for the action of the Heaviside function \( \theta(t' - t_0) \). Since

\[
\int_0^\infty ie^{it\omega^+} e^{-it\omega_{nm}} dt = \frac{-1}{\omega^+ - \omega_{nm}},
\]

(B.4)

it is easy to see that

\[
\chi_{\hat{A}_\alpha, \hat{A}_\beta}(\omega^+, \mathbf{k}) = \frac{1}{\hbar Z} \sum_{n,m} (e^{-\beta E_n} - e^{-\beta E_m}) \frac{\langle n|\hat{A}_\alpha|m\rangle \langle m|\hat{A}_\beta|n\rangle}{\omega^+ - \omega_{nm}}.
\]

(B.5)

Here, we suppress the notation \( \langle ... \rangle_0 \) and indices \( S \), but states and operators are taken at time \( t_0 = 0 \). This suppressed notation is common in the literature. We use Eq. (4.6) and insert the Fermi-Dirac distributions \( f_n = e^{-\beta(E_n - \mu)}/Z \), defining \( f_{nm} = f_n - f_m \) to obtain

\[
\chi_{\alpha, \beta}(\omega^+, \mathbf{k}) = -\frac{1}{\hbar} \sum_{n,m} f_{nm} \frac{\langle n|\hat{A}_\alpha|m\rangle \langle m|\hat{A}_\beta|n\rangle}{\omega^+ - \omega_{nm}}.
\]

(B.6)

Inserting the current operators \( \hat{J}_\alpha = -e\hat{v}_\alpha \) and summing over \( \mathbf{k} \), we get

\[
\sigma_{\alpha, \beta}(\omega^+) = -i\frac{\sigma_0}{\omega} \delta_{\alpha, \beta} - \frac{ie^2}{\hbar V} \sum_{n,m,\mathbf{k}} f_{nm} \frac{\langle n|\hat{v}_\alpha|m\rangle \langle m|\hat{v}_\beta|n\rangle}{\omega^+ - \omega_{nm}}.
\]

(B.7)

For the origin of the first term, which is called the diamagnetic response, see the discussion above Eq. (4.36). We can decompose the denominator in the second term, the paramagnetic response, as

\[
\frac{1}{\omega_{nm} - \omega^+} = \frac{1}{\omega_{nm}} + \frac{\omega^+}{\omega_{nm}(\omega_{nm} - \omega^+)}.
\]

(B.8)
Figure B.1: The graph shows the $\omega$-dependent paramagnetic part of $\sigma$ prior to the replacement $\omega \rightarrow (\omega_n - \omega_m)$ (blue) and after the replacement leading to Eq. (4.43) (orange), plotted with respect to energy $E = \hbar \omega$. This is for a transition between two states, from $E_m$ to $E_n$ at a temperature of $T = 0$, thus $\mu = E_F$.

We use the $f$-sum rule for the free carriers on the Fermi surface [67, 131]

$$\sum_{n,m} f_{nm} \frac{\langle n|\hat{v}_\alpha|m\rangle \langle m|\hat{v}_\beta|n\rangle}{\omega_{nm}} = -\frac{N_f}{m} \delta_{\alpha,\beta}. \quad (B.9)$$

With this, the first term of Eq. (B.8) cancels precisely with the diamagnetic term and we obtain a familiar expression

$$\sigma_{\alpha,\beta}(\omega^+) = -\frac{ie^2}{\hbar V} \sum_{n,m,k} f_{nm} \frac{\langle n|\hat{v}_\alpha|m\rangle \langle m|\hat{v}_\beta|n\rangle}{\omega_{nm}} \frac{1}{\omega^+ - \omega_{nm}}. \quad (B.10)$$

Fig. B.1 shows the qualitative energy dependency of the paramagnetic term with (blue line) and without (orange line) the canceling term. The qualitative change is that a feature centered around $\omega = 0$ is eliminated by the cancellation.
Appendix

C

THE QUANTUM HALL EFFECT

With regards to topological band structures, the importance of the quantum Hall effect cannot be understated. For a plethora of topological band structures, a type of quantum Hall effect can be associated, such as the quantum Hall effect (QHE), the QSHE (spin), the QAHE (anomalous), the QTHE (topological), the QSkHE (skyrmions). Moreover, the measurement of a quantized conductance provides a smoking-gun experiment to the associated topologies. With the Kubo formalism of Chapter 4, we are shortly covering the quantum Hall conductance in the present section. We start by taking the limit $\omega \to 0$ in Eq. (4.54) and further discarding the $k_z$ integral, reducing the problem to two dimensions. We arrive at the following equation for $\sigma_{\text{Hall}} = \sigma_{x,y}^{\text{LL}}(\omega \to 0)$

$$\sigma_{\text{Hall}} = -\frac{iG_0}{l_B^2} \sum_{n,m} \frac{f_m - f_n}{\hbar^2} \frac{\langle n|\hat{v}_x|m\rangle \langle m|\hat{v}_y|n\rangle}{(\omega_n - \omega_m)^2}, \quad (C.1)$$

where we take the scattering $\gamma = 0$ for simplicity. For low temperatures, the Fermi-functions can be approximated as Heaviside functions $f(E) = \Theta(E_F - E)$, where $E_F$ is the Fermi energy. With this, the sum can be modified to only sum over relevant states such that $f(E_n) - f(E_m) = 1$ for all summands, yielding
\[ \sigma_{\text{Hall}} = \frac{iG_0}{l_B^2} \sum_{E_n > E_F} \sum_{E_m < E_F} \frac{\langle n|\hat{v}_x|m\rangle \langle m|\hat{v}_y|n\rangle - \langle n|\hat{v}_y|m\rangle \langle m|\hat{v}_x|n\rangle}{(E_n - E_m)^2}. \] (C.2)

This conductivity we may identify with the one given in the famous TKNN paper [5], but multiplied with the degeneracy imposed by the Landau levels. Unlike [5], we assume electrons to be quasi-free in this derivation, rather than in the context of a periodic lattice potential. This is a valid assumption and avoids the trouble of dealing with Harper’s equations and Hofstadter’s butterfly.

The free electrons are described by the Hamiltonian

\[ \hat{H} = \frac{1}{2m}(\hat{p}_x^2 + \hat{p}_y^2) \rightarrow \hat{H} = \hbar \omega_c (\hat{n} + \frac{1}{2}), \] (C.3)

where on the right-hand side we write the harmonic oscillator equation we get from performing Peierl’s substitution, with the cyclotron frequency \( \omega_c = eB/m \). Within the sum \( \sum_{E_n > E_F} \sum_{E_m < E_F} \), the only transition elements that contribute to the Hall conductivity are those for which \( n = m \pm 1 \), since the ladder operators occur to linear order in the velocity operators

\[ \hat{v}_x = \frac{\hbar}{\sqrt{2ml_B}} (\hat{a} + \hat{a}^\dagger), \quad \hat{v}_x = \frac{i\hbar}{\sqrt{2ml_B}} (\hat{a} - \hat{a}^\dagger). \] (C.4)

It is easy to show that for \( m = n - 1 \) we obtain

\[ \frac{\langle n|\hat{v}_x|n-1\rangle \langle n-1|\hat{v}_y|n\rangle - \langle n|\hat{v}_y|n-1\rangle \langle n-1|\hat{v}_x|n\rangle}{(E_n - E_m)^2} = \frac{i\hbar eBn}{m}. \] (C.5)
APPENDIX C. THE QUANTUM HALL EFFECT

The only bands for which $E_n > E_F$ and $E_m < E_F$ are simultaneously fulfilled while also respecting the transition rules imposed by the velocity operators, are the levels just above and below $E_F$, depicted in Fig. C.1. We then find

\[
\sigma^\text{Hall} = -G_0 n. \tag{C.6}
\]

This is in fact identical with Eq. (2.30) of Chapter 2

\[
\sigma^\text{TKNN}_{x,y} = \frac{e^2}{h} C_n, \tag{C.7}
\]

where $C_n$ is the Chern number, or equivalently the TKNN-invariant $N$ of [5]. The quantum-Hall conductivity is plotted in Fig. C.3. Though, if the conductivity is only contributed by the Landau levels closest to $E_F$, one can wonder how it carries $n$ conductance quanta $G_0$. For a finite sized sample, one finds that each Landau level contributes a conduction channel at the boundary of the sample [132], see Fig. C.2. Each Landau level that intersects with the Fermi energy at the sample boundary adds one quantum of conduction to the overall conductivity. As the distances between Landau levels increase with the field $B$, Landau levels pass through the Fermi energy and stop contributing to the Hall conductivity, and thereby increasing the quantum Hall resistivity,

\[
\rho^\text{Hall} = \rho_{x,y} = \frac{\sigma_{x,y}}{\sigma_{x,x}^2 + \sigma_{x,y}^2}, \tag{C.8}
\]

also plotted in Fig. C.3. Here, we assume $\sigma_{x,x} = \sigma_{y,y}$ and $\sigma_{x,y} = -\sigma_{y,x}$. Following the same procedure as for Eq. (C.2), we see that

\[
\sigma_{x,x} = \frac{-iG_0}{\ell_B} \sum_{E_n > E_F} \sum_{E_m < E_F} \frac{\langle n|\hat{v}_x|m\rangle \langle m|\hat{v}_x|n\rangle - \langle n|\hat{v}_x|m\rangle \langle m|\hat{v}_x|n\rangle}{(E_n - E_m)^2} = 0, \tag{C.9}
\]
Figure C.3: Numerically calculated quantum-Hall conductivity (left) and resistivity (right) for a free electron of mass $m = m_0/20$ and a finite Fermi energy $E_F = 50$ meV.

such that we obtain

$$\rho^{\text{Hall}} = \frac{1}{\sigma^{\text{Hall}}}$$  \hspace{1cm} (C.10)

As a side note, this assumes Landau levels that are infinitely sharp in energy. With a finite broadening $\rho_{x,x}$ peaks whenever a Landau level crosses $E_F$. Note that electron-electron interactions may lift the degeneracy of the Landau levels and give rise to the fractional quantum Hall effect [133, 134].

We have seen now that two fundamentally different effects, the Landau quantization of Bloch electrons in magnetic fields, and the influence of the Berry curvature in topological band structures, can both be characterized by the same quantity, the quantum Hall conductance. This seems perhaps odd, but as stated before, the Berry curvature hosted by topological bands acts similar to a magnetic field in $k$-space. This analogy translates also to the semiclassical equations of motion giving rise to an *anomalous velocity* term [42]

\[
\dot{r} = \partial_k E - \hat{k} \times \Omega, \quad \text{(C.11)}
\]

\[
\hbar \dot{k} = -\partial_r E - e \dot{r} \times \mathbf{B}. \quad \text{(C.12)}
\]
In this context, both $\mathbf{B}$ and $\mathbf{\Omega}$ should give rise to non-quantized measurable quantities. Indeed, the equations above describe the regular Hall effect for the magnetic field $\mathbf{B}$, whereas a corresponding measurement related to the Berry curvature $\mathbf{\Omega}$ is the anomalous Hall effect in magnetized conductors. This anomalous velocity was formulated as early as 1954 [135, 136], long before topological band theory took roots.
Diese Arbeit umfasst theoretische und optische Studien zu einer Auswahl topologischer Materialien, nämlich den Verbindungen TaP, NbAs und GdPtBi. Die Topologie in der Physik der kondensierten Materie ist ein junges und sich schnell entwickelndes Gebiet und befasst sich mit Materialien, die sich dadurch auszeichnen, dass die physikalischen Eigenschaften derer Oberflächen sich qualitativ von denen ihres Inneren unterscheiden. Topologische Isolatoren beispielsweise, ermöglichen keine oder nur eine geringe elektrische Leitung durch das Material, aber dafür über topologische Oberflächenzustände, welche leitende Kanäle an den Rändern des Materials bereitstellen. Im Idealfall können diese Zustände spindpolarisiert, masselos und rückstreuungslos durch nichtmagnetische Verunreinigungen leiten, was einen verlustfreien Ladungstransport ermöglicht. Darüber hinaus sind topologische Phasen, die entweder dem Material innewohnen oder durch externe Parameter, wie beispielsweise einem Magnetfeld, verursacht werden können, durch physikalische Observablen gekennzeichnet, welche quantisierte Werte annehmen, die wiederum proportional zu topologischen Invarianten sind. Diese Invarianten haben die seltsame Eigenschaft, dass sie aus Wellenfunktionen des Materialinneren berechnet werden können, wenn sie doch das Verhalten der Elektronen auf der Materialoberfläche bestimmen. Wichtig ist, dass
topologische Invarianten nicht fein abgestimmt werden müssen, son- 
dern einen endlichen Bereich im Parameterraum abdecken. Da-
her kann die Topologie einiger Systeme ziemlich widerstandsähnig 
gegenüber externen Parametern wie Druck, Temperatur oder Verun-
reinigungen sein, bevor sie durch einen topologischen Übergang 
in das sogenannte triviale Regime gezwungen werden. Mit diesen kom-
bierten Eigenschaften wecken topologische Phasen das Interesse 
an potenziellen Computeranwendungen, wie etwa durch Skyrmionen 
betriebene Racetrack Memorys oder topologische Majorana-Qubits 
für Quantencomputer, die mittels Schnittstellen zwischen topologis-
cher Materie und Supraleitern realisiert werden können.

Eine Untergruppe topologischer Phasen, auf die sich diese Arbeit 
konzentriert, sind die Weyl-Halbmetalle. Dabei handelt es sich um 
Scharen elektronischer Bänder, die konische Schnittpunkte bilden, 
nämlich die Weyl-Kegel, welche durch masselosen Weyl-Fermionen 
ausgezeichnet sind. Weyl-Kegel kommen in chiralen Paaren vor, die 
durch spezielle Arten von Oberflächenzuständen, sogenannte Fermi-
Arcs, miteinander verbunden sind. Weyl-Halbmetalle weisen eine 
Vielzahl interessanter physikalischer Eigenschaften auf, darunter vor 
allem die chirale Anomalie, einen anomalen Strom zwischen Weyl-
Kegeln entgegengesetzter Chiralität. Weyl-Fermionen und die damit 
verbundene chirale Anomalie haben ihren Entstehungspunkt ursprüng-
lich in der Teilchenphysik. Die Entdeckung von Weyl-Fermionen in 
kondensierter Materie im Jahr 2015 birgt nicht nur grosses Poten-
zial für physikalische Anwendungen, sondern hat auch eine Flut an 
Grundlagenforschung mit sich gebracht.

Im Rahmen dieser Arbeit werden die Volumeneigenschaften der Weyl-
Halbmetalle TaP, NbAs und der potenziell magnetfeldabhängige Vari-
ante GdPtBi mittels Infrarotspektroskopie untersucht. Ziel ist es, 
eine Eigenschaft konischer Bänder auszunutzen: Wenn sie Magnet-
feldern ausgesetzt werden, sind die Energien ihrer Landau-Niveaus 
proportional zu $\sqrt{B}$. Dadurch unterscheiden sie sich deutlich von 
den Landau-Niveaus massiver Elektronen, welche linear in $B$ sind,


\[ R_K = \frac{h}{e^2} \approx 25813 \, \Omega, \]


Einige Zeit zuvor zeichnete sich auf Seiten der Teilchenphysik eine andere Entwicklung ab. Im Jahr 1969 arbeiteten S. L. Adler, J. S.

![Figure C.5: Feynman-diagram der ABJ-Anomalie beim Zerfall des Pions $\pi^0 \rightarrow \gamma \gamma$.](image)
Bell und R. Jackiw am Zerfall des Pions $\pi^0 \rightarrow \gamma\gamma$ in zwei Photonen. Die theoretische Abklingzeit des Prozesses war viel länger als experimentelle Werte angaben. Die vorhergenannten Autoren fanden eine Lösung des Rätsels in der chiralen Anomalie oder der ABJ-Anomalie [8, 9]. Das berühmte Feynman-Diagramm des Pionenzerfalls ist in Abb. C.5 dargestellt. Wie wir gleich sehen werden, hat die chirale Anomalie Konsequenzen für die Chiralitätserhaltungsgesetze von Weyl-Fermionen. Um dies zu erklären, betrachten wir den Hamilton-Operator eines masselosen Dirac-Fermions [10]

$$\hat{H} = c \alpha \hat{p} = \begin{pmatrix} -c \sigma \cdot \hat{p} & 0 \\ 0 & c \sigma \cdot \hat{p} \end{pmatrix} = \begin{pmatrix} \hat{H}_L & 0 \\ 0 & \hat{H}_R \end{pmatrix}, \quad \text{(C.13)}$$

welcher hier in der sogenannten chiralen Basis aufgeführt ist. Hier ist $c$ die Lichtgeschwindigkeit, $\hat{p} = -i\hbar \nabla$ ist der Impulsoperator, $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ ist der Vektor der Pauli-Matrizen und $\alpha = -(\alpha_x, \alpha_y, \alpha_z)$ sind die $\alpha$-Matrizen welche in der chiralen Basis als $\alpha_i = -\sigma_z \otimes \sigma_i$ gegeben sind, mit $i \in \{x, y, z\}$. In dieser masselosen Dirac-Gleichung trennt sich der Spinor $\psi$ in zwei chirale Sektoren $\psi = (\psi_L, \psi_R)$. Die Chiralität ist eine Eigenschaft welche beschreibt, dass die Spin- und Impulsvektoren für $\psi_L$ (linkshändig) antiparallel und für $\psi_R$ (rechtshändig) parallel ausgerichtet sind. Die $2 \times 2$-Hamiltons $\hat{H}_{L,R}$ sind als Weyl-Hamilton bekannt und die chiralen Wellenfunktionen $\psi_{L,R}$ beschreiben die Weyl-Fermionen. Der zugehörige relativistische Viererstrom der Dirac-Fermionen ist $J^\mu = \psi^\dagger \gamma^0 \gamma^\mu \psi = (c\hat{\rho}, \hat{j})$, mit $\mu \in \{0, 1, 2, 3\}$. Die $\gamma$-Matrizen in der chiralen Basis sind
\( \gamma^0 = \sigma_x \otimes 1, \quad \gamma^i = i\sigma_y \otimes \sigma_i. \) Der Viererstrom enthält in der Nullkomponente die Teilchendichte \( \hat{\rho} = \psi^\dagger \psi, \) während die restlichen Einträge den Teilchenstrom \( \hat{j} = c\psi^\dagger \alpha \psi, \) wobei \( \alpha_i = \gamma^0 \gamma^i. \)

Dieser Teilchenstrom erfüllt die Kontinuitätsgleichung

\[
\begin{align*}
\partial_\mu J^\mu &= 0 \quad \text{(C.14)} \\
\partial_t \hat{\rho} + \nabla \cdot \hat{j} &= 0 \quad \text{(C.15)}
\end{align*}
\]

Integralform: \( \partial_t \hat{N} + \int_S dS \cdot \hat{j} = \Sigma. \) (C.16)

Die Kontinuitätsgleichung impliziert, dass die Änderung der Teilchenzahl \( \hat{N} \) innerhalb eines Volumens \( V \) mit einem Strom durch die Oberfläche \( S \) des Volumens verbunden ist, dargestellt in Abb. C.6. Wenn innerhalb von \( V \) eine Teilchenquelle vorhanden ist, dann ist \( \Sigma \) eine positive Zahl, wenn eine Teilchensenke vorhanden ist, ist \( \Sigma \) negativ, und wenn die Teilchenzahl erhalten bleibt, dann ist \( \Sigma = 0. \) Wir können chirale Ströme \( \hat{j}_{L,R} = c\psi_{L,R}^\dagger \alpha \psi_{L,R} \) definieren, die ihre eigenen Kontinuitätsgleichungen erfüllen. Insbesondere für die Differenz zwischen links- und rechtshändigen Teilchen,

\[
\begin{align*}
\partial_\mu J^\mu_5 &= 0 \quad \text{(C.17)} \\
\partial_t (\hat{\rho}_L - \hat{\rho}_R) + \nabla \cdot (\hat{j}_L - \hat{j}_R) &= 0, \quad \text{(C.18)}
\end{align*}
\]

wobei \( J^\mu_5 = \psi^\dagger \gamma^0 \gamma^\mu \gamma_5 \psi \) der chirale Teilchen-Viererstrom ist, mit \( \hat{\rho}_{L,R} = \psi_{L,R}^\dagger \psi_{L,R} \) und \( \gamma_5 = i\gamma_0 \gamma_1 \gamma_2 \gamma_3 = -\sigma_z \otimes 1. \) Die chirale Teilchendichte \( \hat{\rho}_L - \hat{\rho}_R \) quantifiziert die Differenz zwischen links- und rechtshändigen Teilchendichten, und \( \hat{j}_L - \hat{j}_R \) definiert eine chirale Stromdichte. Diese chirale Kontinuitätsgleichung sollte die Menge an links- und rechtshändigen Fermionen innerhalb eines Volumens bewahren, wenn keine Ströme durch die Oberfläche ausgetauscht werden. Doch 1969 kam etwas Interessantes ans Licht, als Adler, Bell und Jackiw soeben die erste Quantenanomalie entdeckten: Es stellte sich heraus, dass bei der Quantisierung einer "klassischen" Feldtheorie Symmetrien zusammenbrechen können. In diesem
Fall wird die chirale Symmetrie gebrochen und ein anomaler chiraler Teilchenstrom erzeugt. Der chirale Stromoperator erfüllt dann

\[ \partial_\mu J_5^\mu = \frac{e^2}{16\pi^2\hbar^2} F_{\mu,\nu} \tilde{F}^{\mu,\nu} = \frac{e^2}{4\pi^2\hbar^2} \mathbf{E} \cdot \mathbf{B} \neq 0 \]  

mit \( \tilde{F}^{\mu,\nu} = \frac{1}{2} \epsilon^{\mu,\nu,\rho,\lambda} F_{\rho,\lambda} \), wobei \( F_{\mu,\nu} \) der Feldstärketensor und \( \epsilon^{\mu,\nu,\rho,\lambda} \) der Levi-Civita-Tensor ist. Dies impliziert, dass während in einer klassischen Feldtheorie die Chiralität eines Systems erhalten bleibt, die parallele Anwendung eines elektrischen Feldes \( \mathbf{E} \) und eines magnetischen Feldes \( \mathbf{B} \) stattdessen die Chiralität des Gesamtsystems verändern kann, wenn Quanteneffekte berücksichtigt werden. Dies führt zu einem Ungleichgewicht in der Anzahl chiraler Partikel, wobei wir jedoch feststellen sollten, dass die Gesamtpartikeldichte richtigerweise erhalten bleibt. Im Fall der masselosen Weyl-Fermionen verändert die Anwendung paralleler Felder dauerhaft die Chiralität des Systems. Das Vorhandensein eines Massenterms auf den Nebendiagonalen Einträgen im Hamilton-Operator von Gl. (C.13) würde die chiralen Unterräume vermischen und eine Relaxation eines solchen chiralen Ungleichgewichts ermöglichen. Damals ging man davon aus, dass Weyl-Fermionen in Form von Neutrinos [11, 12] realisiert werden. Heute wissen wir durch Messungen der Neutrino-Flavour-Oszillationen [13], welche einen solchen Relaxationsprozess darstellen, dass Neutrinos endliche Massen haben, und es ist bisher nicht bekannt, dass ein freies Weyl-Fermion existiert.

Werfen wir einen Blick auf eine besondere Anwendung der chiralen Anomalie, nämlich das Axion [14–16]. Der anomale Strom modifiziert den Lagrange-Operator der Quantenfeldtheorie um einen Term [14]

\[ \mathcal{L} = \frac{\theta g^2}{32\pi^2} F_{\mu,\nu} \tilde{F}^{\mu,\nu}, \]  

und bricht die chirale Symmetrie der Feldtheorie. Der Parameter \( g \) beschreibt die Kopplungsstärke zwischen dem elektromagnetischen Feld und dem Fermion, während der Parameter \( \theta \) ein freier Parameter ist, der jeden Wert zwischen 0 und \( 2\pi \) annehmen kann. Die
chirale Anomalie existiert in äquivalenter Weise für Gluonenfelder \( G_{\mu\nu} \) (man ersetze \( F_{\mu\nu} \) durch \( G_{\mu\nu} \) in der obigen Gleichung) in der Quantenchromodynamik (QCD) \([17]\). Die Gluonenfelder sind für die Bindung der Quarks verantwortlich, aus denen Neutronen und Protonen bestehen. Eine Observable, für die der Effekt der chiralen Anomalie relevant werden soll, ist das elektrische Dipolmoment des Neutrons. Jenes ändert sich, wenn der \( \theta \)-Term in die Theorie einbezogen wird. Theoretische Schätzungen ergeben Werte in der Größenordnung \( d_n^{\text{theo}} = 4.5 \times 10^{-16} \theta \ e \cdot m \), wobei \( e \) die Elektronenladung ist, wohingegen Messungen eine Obergrenze von \( d_n^{\text{exp}} < 2.9 \times 10^{-26} \ e \cdot m \) feststellen. Dies impliziert, dass \( \theta \) in der Größenordnung von \( \theta < 10^{-11} \) liegen muss. Da \( \theta \) jeden Wert zwischen 0 und \( 2\pi \) annehmen darf, ist es seltsam, dass es ohne konkreten Grund so klein ist. Dies ist das sogenannte \textit{starke CP-Problem} (Charge-Parity). Die bisher vielversprechendste Lösung besteht darin, \( \theta \) zu einem (periodischen) Feld \( \theta(\mathbf{r}) \) zu machen, sodass die potentielle Energie minimiert wird, wenn sich die Teilchenfelder der QCD in den Minima von \( \theta(\mathbf{r}) \) einpendeln. Dies ist die Peccei-Quinn-Theorie und sie führt zu einem bosonischen Teilchen namens Axion \([14–16]\).

Winkel $\theta$ auf $\pi$ festgesetzt, während an der Oberfläche der Probe, die an das Vakuum gebunden ist, $\theta$ schlagartig auf 0 sinkt. Bereiche, in denen $\theta$ konstant ist, erzeugen keine neue Physik. Somit ist das Volumen einer Probe mit einem konstanten $\theta = \pi$ physikalisch identisch mit demselben System mit $\theta = 0$, beziehungsweise ohne $\theta$-Term. Die Axion-Elektrodynamik findet ausschließlich auf der Oberfläche der Probe statt, wo eine plötzliche Änderung von $\theta = \pi$ zu $\theta = 0$ auftritt, wodurch ein leitender Oberflächenkanal entsteht. Damit war der Axion-Isolator geboren, der erste topologische Isolator [21, 22].

zten Dispersionen der Energien \( E(k) \) der linearen Landau-Niveaus (\( k \) ist die Wellenzahl) drückt das elektrische Feld die Elektronen in einem Kegel nach unten unter \( E_F \), wodurch dieser effektiv entleert wird, und im anderen Kegel nach oben, welcher ihn effektiv füllt und insgesamt chirales Ungleichgewicht erzeugt wird.


Diese Abhandlung ist wie folgt aufgebaut:

- In Kapitel 1 skizziert die Einleitung kurz den Zusammenhang zwischen topologischen Phasen und Teilchenphysik, wobei der Schwerpunkt auf den Weyl-Fermionen und der chiralen Anomalie liegt.
• In Kapitel 2 führen wir zunächst an geometrischen Beispielen in die Topologie ein und arbeiten uns dann an topologischen Beschreibung von Weyl-Kegeln im $k$-Raum unter Verwendung von Berrys Grössen heran.

• In Kapitel 3 leiten wir die Landau-Quantisierung von Bandelektronen ein, die häufig bei Niederenergienäherungen elektronischer Bandstrukturen angewendet wird. Darüber hinaus enthält das Kapitel eine kurze Beschreibung der chiralnen Anomalie von Weyl-Halbmetallen in Magnetfeldern.

• In Kapitel 4 liefern wir eine Herleitung der Kubo-Formel für die optische Leitfähigkeit. Die resultierende, wohlbekannte Gleichung kann zur Auswertung der Reflexivitätsspektren in den kommenden experimentellen Kapiteln verwendet werden.

• In Kapitel 5 wird der Versuchsaufbau beschrieben und die Details der Experimente skizziert.

• In Kapitel 6 wird das Landau-Niveauspektrum des Weyl-Halbmetalls TaP diskutiert. Am auffälligsten und unerwartetsten sind Spuren der Bandinversion, die ein wesentlicher Bestandteil für das Vorhandensein von Weyl-Kegeln ist. Die Messungen und vorläufige Analyse wurden in der Master-These des Autors präsentiert [1].


• In Kapitel 8 wird das magnetooptische Spektrum des mag-


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