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Progress in Physics (99)

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Introduction

When a strong magnetic field is applied to a crystalline solid, the laws of quantum mechanics confine the motion of band electrons into a set of discrete or quasi-discrete cyclotron orbits. This regime is usually referred to as the Landau quantization and the distinct cyclotron orbits are called Landau levels. Using light, one can then excite charge carriers from one Landau level into another. Tracing such excitations experimentally is known as Landau level spectroscopy and provides us with indispensable insights in the material's band structure. Since the early experiments in the 1950s, this technique has been widely employed as an extremely sensitive probe of semimetal and semiconductor band structures. The technique can also help assess the materials' quality. For example, one can obtain the scattering time or the carrier mobility, as well as the carrier density.

History of Landau level spectroscopy

Magneto-optical spectroscopy has a long history, intimately tied with important discoveries in solid state physics. Classically, magneto-optics dates to Michael Faraday and his 1845 experiments on magneto-optical rotation, today known as Faraday effect. However, quantum physics fundamentally changed this technique, how we understand it, and what we can learn from it.

The starting point for modern magneto-optics and Landau level spectroscopy are the first cyclotron resonance experiments from the early fifties. Arthur Kip, Gene Dresselhaus and Charles Kittel were the first to observe the cyclotron resonance in crystals of germanium [1]. Cyclotron resonance itself is a classical effect, where a charged particle which moves in a magnetic field follows a helicoidal path. An electron (or a hole) in a semiconductor can complete many cyclotron orbits before being scattered. It absorbs energy resonantly at its cyclotron frequency $\omega_c = eB/m_c^*$, where m_c^* is the cyclotron effective mass. Measuring cyclotron resonance then allows to measure effective masses.

Observing the cyclotron resonance in germanium was a key achievement for the history of solid state physics. It helped to experimentally establish the quasiparticle concept [2]. The cyclotron resonance experiment provided a mapping of the dependence of the cyclotron effective mass as a function of the angle between the magnetic field and the various crystallographic directions.

The first experiments on germanium were quickly followed by a series of experiments in many other semiconductors. Studies on InSb [3] showed extremely small effective mass carriers, with a large effective g factor, $g \sim -50$. Because InSb lacks spatial inversion, and has a large spin-orbit interaction, it has been discovered that it has linear k terms in $E(k)$, near the Brillouin zone center, where the band extrema occur.

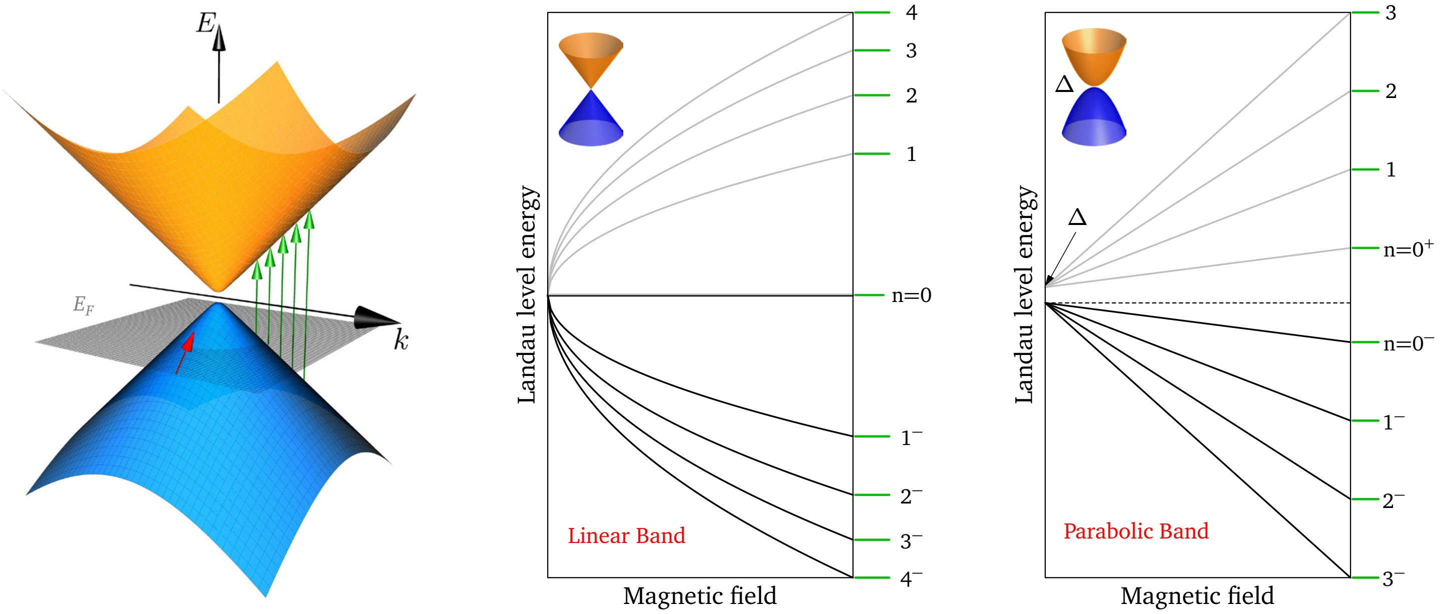

Figure 1. Left: Band structure described by a model of massive Dirac electrons. The Fermi level E_F is shown in gray. Short red arrow illustrates intraband excitations, while green arrows show interband optical excitations. Middle and right: Schematics of Landau levels for a conical band (e.g., in graphene) and two parabolic bands (e.g., in semiconductors). The field dependence of Landau levels differs profoundly. The $\sqrt{B}$ dependence is a hallmark of conical bands, while the linear in B dependence is typical of systems with a parabolic dispersion. The zero-field extrapolation of optical excitations between Landau levels provides us with a useful estimate of the band gap.

Even though some Landau level spectroscopy experiments can be successfully interpreted using a simple picture of classical cyclotron resonance, usually a full quantum-mechanical treatment of the data is necessary. This may be illustrated on cyclotron resonance in germanium. While cyclotron resonance of electrons in germanium can be explained semi-classically, the interpretation of hole cyclotron-resonance data requires a detailed knowledge of the Landau level spectrum. This is due to a more complex structure of the valence bands in germanium, comprising two anisotropic dispersion branches (referred to as light and heavy holes).

In three-dimensional materials, Landau quantization splits the electronic bands into one dimensional sub-bands. These sub-bands show up very distinctly in optical data, as their density of states can exhibit sharp peaks under favourable experimental conditions. Soon after the pioneering experiments, it became clear that the interband optical absorption in high magnetic fields was a method which could reveal a wealth of band-structure information [4].

Landau level dispersion

To illustrate how Landau level spectroscopy works, let us consider a pair of bands, in our example defined by a massive Dirac dispersion and let us show processes induced an incident photon. If the Fermi level is placed within the valence band, two processes may occur at $B = 0$. The first one is an intraband excitation of electrons – so-called Drude absorption, characteristic of all systems with free charge carriers – illustrated by the red arrow in Figure 1(left). Here, carriers are excited just across the Fermi energy. The interband excitation is the second possible process. It brings an electron from a partly occupied conduction band, across the band gap, into the empty conduction band. When a sufficiently strong magnetic field is applied, both type of processes are impacted by Landau quantization.

Here, we will consider two simple cases, see Fig. 1(middle-right). A band with a linear dispersion, *E(k)*, has the Landau level spectrum reading:

$$\varepsilon_n^{\pm} = \pm\hbar v\sqrt{k_z^2 + \frac{eB}{c}2n} \quad (1),$$

where the individual levels are labelled by the integer Landau level index n, v is the velocity parameter describing the steepness of the conical band, and B stands for the magnetic field. In turn, a parabolic band gives the following sequence of Landau levels:

$$\varepsilon_n = \frac{\hbar^2 k_z^2}{2m} + \hbar\omega_c\left(n+\frac{1}{2}\right) \quad (2).$$

In both cases, the k_z is the momentum along the applied magnetic field that is not quantized by the applied magnetic field. Importantly, a detailed quantum-mechanical analysis – so-called selection rules – is needed to know the probability of different excitations of electrons in the Landau level spectrum. Most often, the rule is that the Landau level index has to change by 1 or -1.

Landau level spectroscopy of topological materials

A number of topological materials have their band structures marked by low energy scales. Together with their low carrier densities and high carrier mobilities, this makes them well suited for experiments using infrared light and mag-

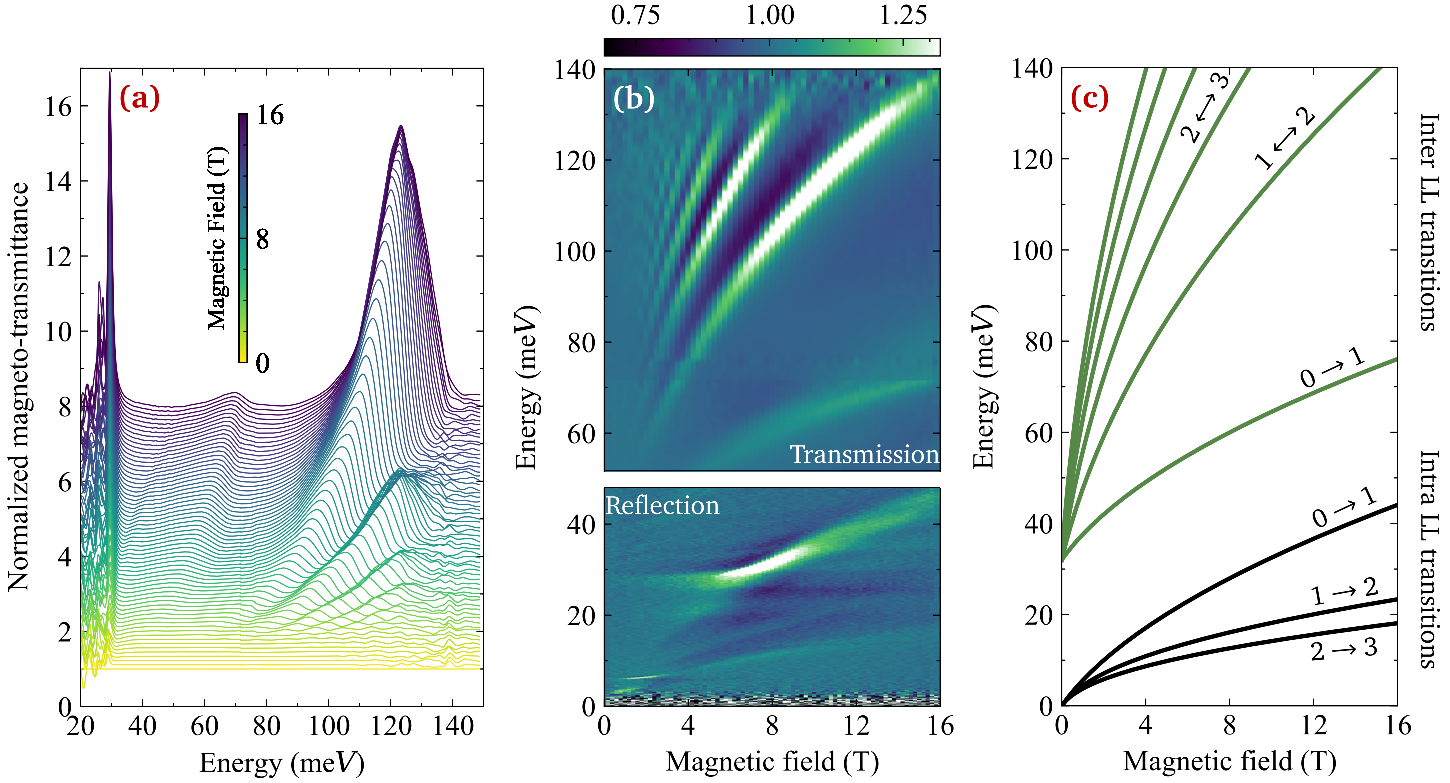

Figure 2. Magneto-optical spectra measured on a crystal of TlBiSSe. This material is a good illustration of a Dirac semimetal. (a) Stacked photon-energy-dependent spectra of relative magneto-transmission, measured up to 16 T. (b) Colorplot containing both magneto-transmission and magneto-reflection data. Lighter parts of the colorplot correspond to inter- and intra-Landau level transitions. (c) Using a massive Dirac model, we can assign the optical transitions to different combinations of Landau levels. The extrapolation of the 0 → 1 line into zero field gives us the value of the band gap.

netic field. In fact, many important insights on topological materials have been gleaned from Landau level spectroscopy. High-quality materials, where carriers can complete many cyclotron orbits before scattering, let us observe inter-Landau level transitions already at low magnetic fields. For example, in ZrTe$_5$ which has conduction and valence bands dispersing nearly linearly in two spatial directions, inter-Landau level transitions are observed, at low photon energies, at magnetic fields as low as a few milliteslas [5]. This allowed for a precise determination of a small band gap (5 meV) that increases monotonically with temperature [6].

Figure 2 shows another example of a topological material, TlBiSSe, which may be referred to as a “text-book Dirac semimetal” [7]. We show it here to illustrate the power of Landau level spectroscopy when applied to a topological semimetal. Through detailed measurements of magneto-transmission and magneto-reflection (Fig. 2(a)), one can construct a colormap (Fig. 2(b)) containing a series of transitions. Each of these lines is then attributed to a certain transition between a pair of Landau levels, such as shown in Fig. 2(c). This allows us to determine precisely the band structure parameters: the velocity parameters and also a small band gap, reaching in this case 4.0×10^5 m/s and 32 meV, respectively. Moreover, one may notice in Fig. 2(b) that there is an anti-crossing of the 0 → 1 line with an infrared-active phonon, suggesting an electron-phonon coupling.

Almost as a rule, Landau level spectroscopy provides deep and novel insight into any topological material it touches. Let us list a few relevant examples, in which authors of this text were involved. For the topological insulator Bi$_2$Se$_3$, this technique showed that the conduction and valence bands are both parabolic-like, and firmly established the band gap value [8]. In the related topological insulator Bi$_2$Te$_3$, Landau level spectroscopy shows that the fundamental band gap is not at the Γ point, contrary to popular belief [9]. As a first candidate of a Dirac semimetal, Cd$_3$As$_2$ incited a lot of excitement. However, magneto-optical experiments showed that the linear energy dispersion in this system is largely unrelated to its topological properties [10,11]. Weyl semimetals TaP and TaAs were also carefully studied using magneto-optical experiments. In TaP, one can obtain the exact scale of the band inversion, leading to Weyl nodes, and verify detailed theoretical models [12]. In TaAs, similar experiments showed that the number of Weyl nodes is smaller than previously believed [13]. In nodal line semimetals, Landau level spectroscopy uncovered a previously unseen relativistic-like effect, Lorentz boost, which can effectively mask the band gap [14].

Conclusions

Landau level spectroscopy is an established technique with a long and distinguished past. Presently it is employed by a handful of experimental solid-state physics groups around the world. Despite being much less represented than, for example, photoemission spectroscopy, this technique has provided us with a wealth of information about semiconductors and semimetals, be they topological or not, and will hopefully continue to do so in the years to come.

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Landau level spectroscopy is strongly tied to the high magnetic field technology development, and to our capabilities of generating sufficiently high magnetic fields. Until the 1950s, experiments were performed using electromagnets which could only reach about 2 T. In the early sixties, the available magnetic field range was extended to 15 – 20 T. These magnetic fields were achievable in several high magnetic-field facilities. In the late sixties, reasonably affordable superconducting coils made it possible to generate magnetic fields in the 10 T range and perform sophisticated magneto-optical experiments in one's own laboratory.

Modern experiments in high magnetic fields can be done in pulsed magnetic fields reaching several hundreds of Tesla. For the infrared experiments, however, we need static magnetic fields, which are limited to below 50 T. For example, at the Laboratoire National des Champs Magnétiques Intenses (LNCMI) in Grenoble, one can currently achieve magnetic fields up to 36 T using resistive magnets that are water-cooled and based on copper alloys. Their internal structures have special geometries to ensure very rapid thermal exchange with the cooling water, and to resist the Lorentz forces. Very soon, the static high field limit at LNCMI will be increased to 45 T in a hybrid magnet, combining a resistive insert magnet in a superconducting outer magnet.