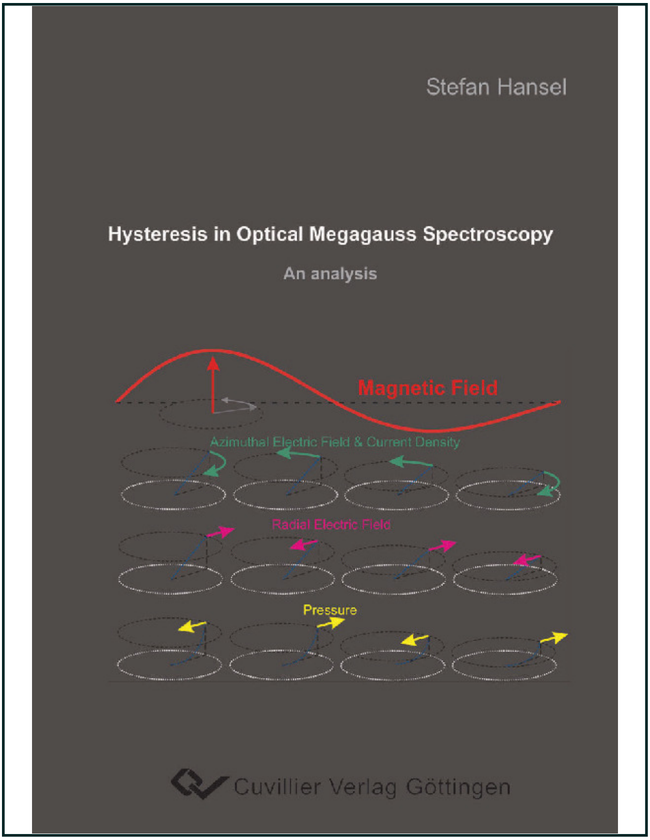
Stefan Hansel (Autor)

Hysteresis in Optical Megagauss Spectroscopy



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Hysteresis in Optical Megagauss Spectroscopy

An analysis

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Introduction

The cyclotron resonance is a very traditional tool in semiconductor physics for determining carrier effective masses. Materials with high effective masses, low mobility or high carrier concentration require high energies for the observation of cyclotron resonance. Therefore, Mid Infra-Red radiation energies are used in combination with very high magnetic fields.

The highest magnetic fields on a laboratory scale can only be generated at the expense of a very short pulse time of order μs. Whereas the field magnitude effects on semiconductors are well investigated, the transient character of the field generation has been assumed to be of minor influence.

Analytical methods developed for steady magnets with comparably low fields have been used in pulsed fields accordingly. In pulsed, sinusoidal fields it is possible to measure optical resonances in rising and falling field in one experiment. Intuitively one would expect identical behavior in this case. With improved experimental resolution and the resulting possibility of fully using available data on rising and falling field sides of a pulse the respective spectra showed discrepancies between each other.

In the last few years, it became apparent that observable cyclotron resonance spectra are not independent of the magnetic field generation process, that means, they depend on the sweep rate or pulse duration.

A spin hysteresis of the conduction band electrons of narrow gap semiconductors has been observed in transient magnetic fields [1–5]. The results could be readily explained with a spin lattice relaxation time of the conduction band electrons of order $1\,\mu s$. In this case the time dependence of the magnetic field enabled the detection of population dynamics for the spin split cyclotron resonances of electrons.

Recently, other hysteretic phenomena on the cyclotron resonances of electrons have been observed that could not be explained by a spin hysteresis, but showed a dependence rather on the field derivative than its temporal structure [6, 7]. Moreover, even well known materials, such as InSb, exhibited completely unexpected hysteretic behavior in transient magnetic fields. No previously known resonance was involved in the hysteresis, but a completely new phenomenon. This leads to the conclusion that the interaction of the transient magnetic field generation with a conducting sample is not negligible and must be systematically examined.

Although this problem has been known for many years, the influence of eddy currents and electric fields as induced by the transient character of the magnetic field pulse has been treated only by crude approximations. It was also incomplete regarding the effects connected with that such as a rise in temperature, a screened magnetic field and an exertion of pressure onto the conductor.

The goal of this thesis is a fundamental expanding of the understanding of the effect of transient magnetic fields and semi-conducting samples with the focus on the implications for cyclotron resonance measurements, that is for magneto-transmission experiments. The magnitude and quality of eddy current related effects will be rigorously derived in unprecedented complexity. The experimental focus will be upon hysteretic phenomena in InSb that are analyzed with respect to the theoretical findings.

This thesis is divided into 6 chapters.

The first chapter summarizes hysteretic phenomena in cyclotron resonance experiments in single turn coils as have been reported prior to and at an early stage of this work. An analysis will be performed to draw conclusions upon the experimental requirements for the investigation of similar effects.

The second chapter will review the theory of cyclotron resonance and illuminate what sample properties can be deduced from experimental spectra. A large part of this chapter will deal with the interaction of a conducting sample with a transient magnetic field, that is a field with pulse durations of order μs. The effects of induced electric fields and eddy currents will be derived.

Chapters three and four are dedicated to the experimental setup and focus upon the required modifications imposed by the considerations in chapter one and two. Chapter three will focus upon the magnetic field generation, chapter four upon the optical setup for measurements of cyclotron resonance.

Chapter five is dealing with a hysteretic phenomenon that has not been previously reported in InSb. It will be shown how the transient magnetic field character interacts with the infrared relative transmission spectra of InSb. For this purpose various experimental techniques beyond the cyclotron resonance setup in a transient field will be used and extended. The influence of eddy currents on the spectra will be quantitatively determined and it will be demonstrated that a radial, HALL-like electrical field is built up inside the sample due to the transient magnetic field and causes the above mentioned hysteretic phenomena in InSb that have not been understood previously.

Chapter six will briefly report and investigate sweep rate dependent hysteretic phenomena on mercury based compounds. Measurements depending on the macroscopic sample dimensions will show that the size of the sample radius affects the observed spectra and macroscopic phenomena such as eddy currents become effective.

Finally a summary will be presented.

Chapter 1

Motivation

As of present day magnetic fields in the 100 T, or *Megagauss*, range and above can only be generated in a laboratory with various methods on a very short timescale of order microseconds. The time dependence of the magnetic field pulse enabled the observation of various hysteretic phenomena. Methods of data evaluation and drawing conclusions developed for DC magnetic fields can neither predict nor explain any of these results.

At first, this chapter reviews previous reports on hysteretic phenomena of non-magnetic semi-conducting systems on a μs timescale in transient magnetic fields as found in the literature.

1.1 General Features

A hysteretic phenomenon can generally be defined as a retardation of an effect behind its cause. Thus, hysteresis is a property of physical systems that do not instantly follow the forces applied to them, but react slowly, or do not return completely to their original state: that is, systems whose states are not in thermodynamical equilibrium and depend on their immediate history [8].

For magnetic materials this has been subject of many studies [9]. In the present investigation, semi-conducting materials shall be focused upon that do not contain magnetic dopants or components besides of the spin.

In this work hysteresis will be understood as discrepancies in the transmission of infrared electromagnetic radiation through a semiconductor during the rising and falling magnetic field, respectively. Two fundamentally different hysteretic phenomena can be observed, firstly, a discrepancy of the time relation of the magnetic field and the optical response of the semi-conducting system, secondly, differences in the absorption strength of optical transitions.

The first phenomenon is very difficult to be determined or observed conclusively, thus previous reports deal predominantly with the latter effect.

1.2 InAs/AlSb Single-Quantum Well

Historically the first publications and reports on hysteresis effects observed in infrared spectroscopy of semiconductors in transient magnetic fields investigated a single quantum well of InAs/AlSb [1–3] by the Megagauss Laboratory of the ISSP, Tokyo, Japan.

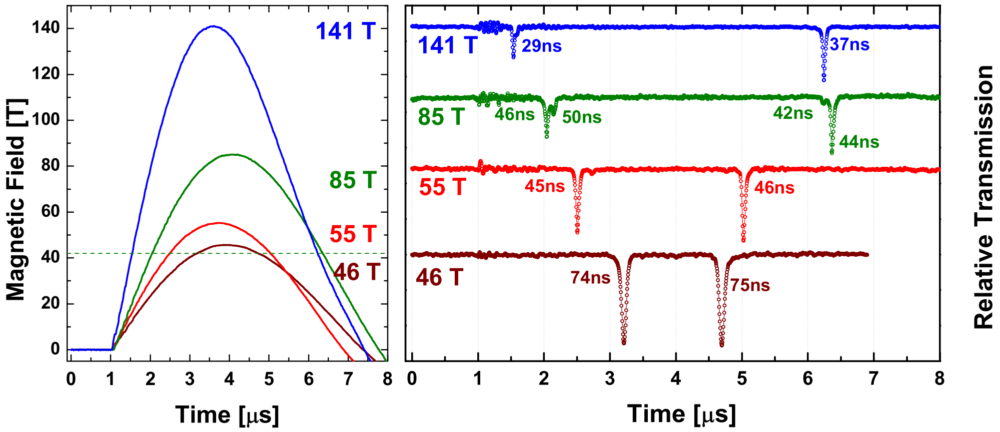

Figure 1.1: *Comparison of relative transmission through samples at 16 K and 10.59 μm at various magnetic fields with significantly different sweep rates [1–3]. It can clearly be seen that the sweep rate does have an effect on the relative population of the spin split electron level. The FWHM values of time are given at the respective resonances. The corresponding magnetic field traces are given on the left side. The horizontal line indicates the spin down resonance position.*

The investigations have been carried out on samples with 15 nm InAs quantum wells in 15 nm and 20 nm barriers of AlSb with InSb like interfaces, carrier concentration was determined to be $3-5\times10^{11}\,cm^{-2}$ at 4 K and $9-12\times10^{11}\,cm^{-2}$ at 300 K and mobilities of $26\,m^2/Vs$ and $10\,m^2/Vs$, respectively [3].

Using a cyclotron resonance setup with a single turn coil magnetic field generator for a wavelength of 10.6 μm and a temperature of 16 K a dependence of the spin split cyclotron resonance on the maximum field was observed. The raw data is given in fig. 1.1 which can be transferred into the relative transmission spectrum vs. magnetic field of fig. 1.2.

While in the falling field side only the spin down state is observed, the spin up state becomes increasingly prominent in the rising field side with increasing maximum field. It is however, important to note that the total absorption intensity for spin up + spin down absorptions is conserved for all fields.

For detailed analysis of the optical traces it is important to estimate the effects of the recording equipment. There is no report on the parameters of the detector system used to obtain the data in [1–3]. For a cyclotron resonance measurement increasing the maximum field means a higher field derivative and thus a shorter time a resonance can be recorded by the detector system.

It can be clearly observed in fig. 1.1 that the resonance strength increases with decreasing maximum field, that is, increases with increasing recording time. This is a phenomenon that can occur when the resonance FWHM is short in time, shorter than the time defined by the bandwidth of the detector system. To obtain the signal from the sample the optical trace must be convolved with the response function of the detector system. It is approximately constant from DC to the detector bandwidth, but decreasing rapidly beyond that value. The FWHM time during which the resonance is recorded

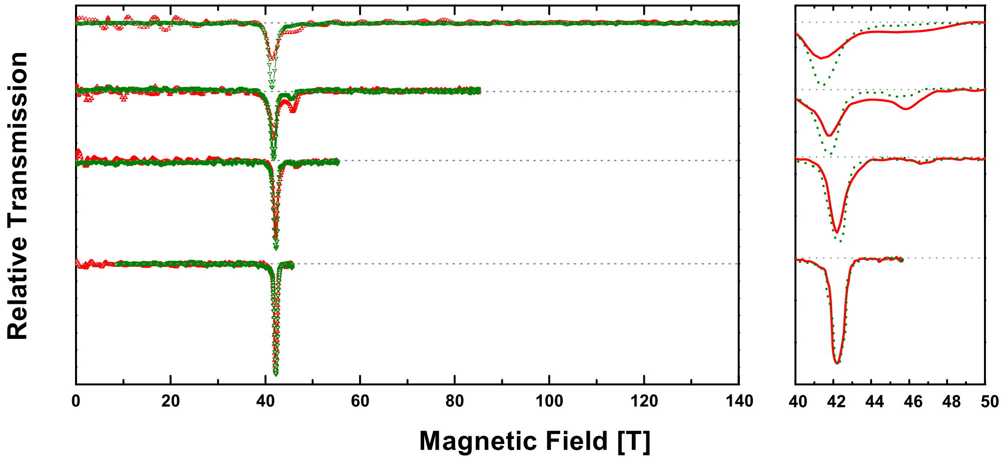

Figure 1.2: *Hysteresis in the relative transmission spectra of a InAs/AlSb single quantum well at 10.59 μm wavelength and 16 K Temperature. [1–3] The graph on the right shows details of the hysteresis affected spin split CR. The dashed lines are the down sweeps.*

increase from 29 ns at the fastest rising field to 74 ns at the slowest field rise. There is a time comparable to that value recorded in falling field side except for the highest field due to a much smaller value for the field derivative.

A band width of 20 MHz of the detection system could explain the trace for the highest field and the increase of resonance strength with increasing FWHM time of the resonance in all spectra. The quantitative data evaluation of these traces is questionable without detailed information on the detector system. However, the falling field resonance has approximately the same FWHM as the rising field resonance for the other fields. The response function will be similar for both resonances so that differences in relative resonance strength will be sample related. The discussion by [1–3] attributes all phenomena to the sample response to the magnetic field.

The pulse duration for all fields was $\sim 6\,\mu s$, much longer than the experimentally determined momentum scattering times of typically 0.1 ns at 12.5 T . Thus thermal equilibrium distributions for carriers can be expected at fields at this value, both spin split levels should be equally populated for a slowly varying field.

For a transient magnetic field the situation changes. With increasing field electrons depopulate the spin up level and relax into the spin down level. Thus the lower level will be populated by more electrons than expected from equilibrium statistics. Using a rate equation similar to equation (1.2) a spin relaxation time of $\sim 1\,\mu s$ was obtained.

The LANDAU level wave functions are a mixture between spin up and down states, thus relaxation processes between different spin states by a electric type perturbation are possible [10]. Because of the discrete density of states, the spin flip relaxation is an energy loss process and can happen through an inelastic emission of phonons. It was found that single phonon processes are unlikely due to the LO phonon energy of 29.5 meV which is larger than the ZEEMAN energy and scattering by single acoustic phonons cannot satisfy energy and momentum conservation at the same time. Yet, it was shown that multi-phonon scattering can bypass these processes and account for the long relaxation

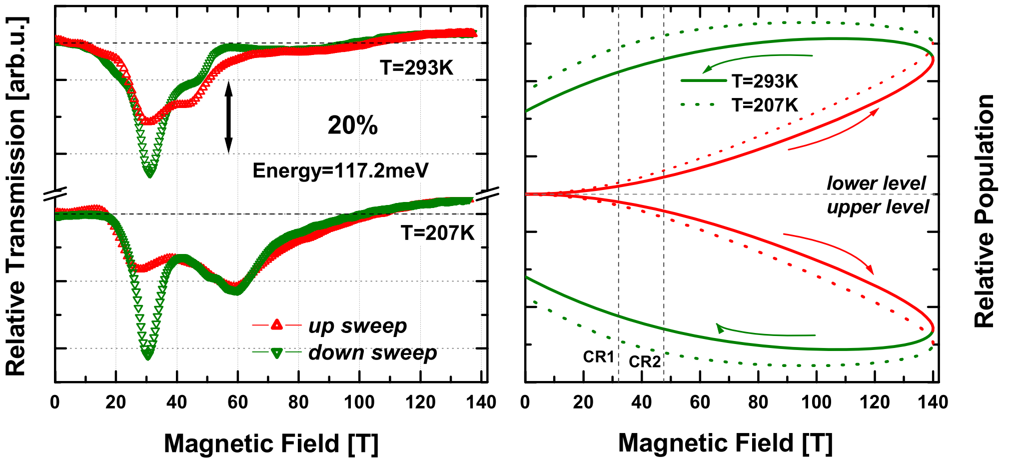

Figure 1.3: *Left: the relative transmission of infrared radiation of 117.2 meV through a HgSe sample at room temperature and 207 K in a magnetic field of up to 140 T. The population changes for the spin split electron cyclotron resonance can clearly be observed [4,5]. Right: The non-equilibrium distribution for the upper and lower spin-state of the spin split cyclotron transition.*

times [2,3,11].

Non-equilibrium populations of electron spins created by a transient magnetic field can be probed by CR for characteristic times of 0.1 - few μs. The authors of [3] suggest that InSb-related 2 DEGs should be the most promising candidates for the observation of that effect as the g-factor is large, therefore a large Zeeman splitting results in long spin flip relaxation time.

The observations could only be made for the higher carrier concentration sample and have been obscured by other effects for the lower concentration sample [3].

1.3 Quasi Bulk HgSe

A pronounced hysteresis of the population of the magnetic field dependent electronic energy levels in a quasi bulk sample of $2\,\mu m$ MBE grown HgSe has been reported on several occasions [4,5]. The phenomenon affecting the spin split electron cyclotron resonance was observed at elevated temperatures $T \geq 207\,K$ in a single turn coil generated magnetic field of $6\,\mu s$ pulse length and 140 T magnitude.

The data of [4,5] are shown in figure 1.3. The relative transmission spectra of the up and down sweep have identical resonance positions but the respective intensity of absorption is higher in the down sweep for the spin down resonance at cost of the intensity of the spin up resonance line. The total absorption intensity $I_{tot} = I(\uparrow) + I(\downarrow)$ is conserved in up and down sweep. Earlier reports of a similar observation (section 1.2) have tentatively been explained by a delayed population adjustment of the energy levels involved due to the finite spin-lattice relaxation time in pulsed transient magnetic fields.

Applying a two level system for spin states equally populated at zero magnetic field the relative population was simulated. The non-equilibrium distribution functions $f_1(t)$

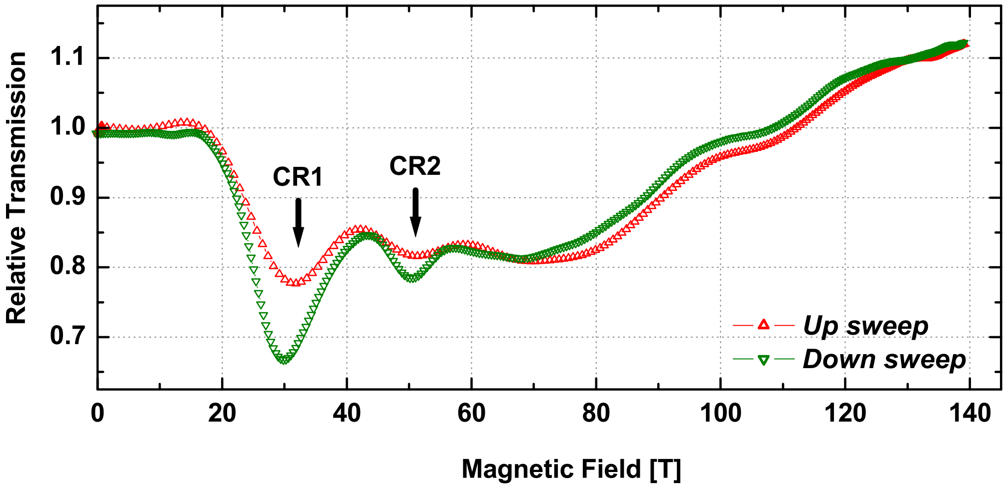

Figure 1.4: *The relative transmission of infrared radiation of 117.2 meV through HgTe at room temperature in magnetic fields up to 140 T. The population changes for the spin split electron cyclotron resonance can clearly be observed [6, 7].*

and $f_2(t)$ for the two magnetic field dependent energy levels $E_1(B(t))$ and $E_2(B(t))$ can be characterized starting with the equilibrium distributions $f_{o1/2}(t)$ of the Boltzmann type

$$f_{oi}(t) = \frac{\mathbf{e}^{-E_i(B(t))/kT}}{\mathbf{e}^{-E_1(B(t))/kT} + \mathbf{e}^{-E_2(B(t))/kT}} \tag{1.1}$$

with i=1,2. The non-equilibrium distribution functions $f_i(t)$ then obey the equation

$$\frac{df_i(t)}{dt} = -\frac{f_i(t) - f_{oi}(t)}{\tau} \tag{1.2}$$

assuming that the relaxation time τ is equal for both levels.

Considering the time dependence of the magnetic field pulse and combining it with a $\vec{\mathbf{k}} \cdot \vec{\mathbf{p}}$ calculation of the energy levels involved the non-equilibrium distribution functions have been simulated. The result is given in figure 1.3. A spin lattice relaxation time $\tau = 1.2\mu s$ is in complete agreement with the experimental findings.

The actual physical mechanism responsible for this spin lattice relaxation has not been deduced [4,5]. For higher carrier concentration samples the effect was not observed. [12]

1.4 Quasi Bulk HgTe

Being similar to HgSe in most important electronic properties a study of HgTe was performed and compared with the results of HgSe [6,7]. The most important experimental finding is reproduced in figure 1.4. The sample was $2\,\mu m$ MBE grown quasi bulk HgTe. A change in the absorption strength in up and down sweep could be detected, yet the increase in the absorption strength of the spin up resonance was not on cost of the magnitude of the spin down resonance, the total absorption intensity is not conserved

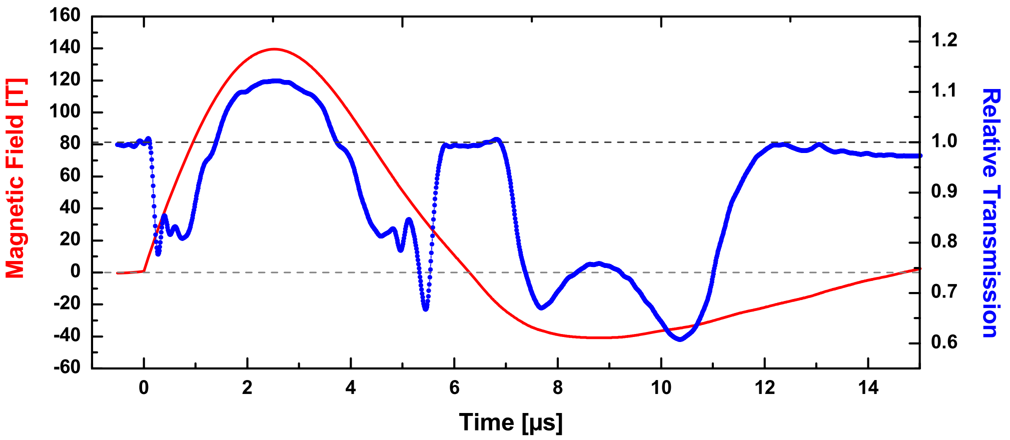

Figure 1.5: *The relative transmission and magnetic field trace for HgTe at 10.59 μm and room temperature. It is clearly seen that the resonance strength continues to increase after the first zero crossing. Moreover, the FWHM of the resonances are larger than 100 ns for all resonances.*

during up and down sweep. Tentatively, this suggests two competing physical processes or a completely different one.

Moreover, a further increase in absorption strength was observed even after the zero crossing when the resonance field was met in opposite field direction as depicted in fig. 1.5. This could be observed for various temperatures in the range from 6 K to 300 K and was found to be sweep rate dependent [6, 7].

Temperature dependent studies have been carried out to deduce a set of high field parameters for the $\vec{\mathbf{k}}\cdot\vec{\mathbf{p}}$ - model but the respective spectra have not contributed to further insights on relaxation phenomena [6, 7].

However, the data were insufficient for detailed analysis and conclusions regarding exact derivation of relaxation times and physical mechanisms.

1.5 Bulk InSb

At the beginning of this study, a new and very remarkable observation was made in the infrared transmission of bulk InSb samples. The experimental data have been reported at several occasions, e.g. [13]. An example optical trace is given in fig. 1.6.

There is a transmission drop at very low fields that can be observed only *after* zero crossings of the magnetic field while there is no feature at the respective position before the zero crossing. The magnitude of the drop is proportional to the absolute value of the field derivative. The authenticity of the spectra is given by the observation of the cyclotron resonance of electrons that is in excellent agreement with theory and literature.

The cyclotron resonance of InSb has been previously investigated in STC generated fields [14–16] as well as extensively in non-destructive pulsed magnets on a 100 ms time scale e.g. [17]. The phenomenon described here could not be observed in those publications because of the trigger noise [14], the generally poorer signal to noise ratio and

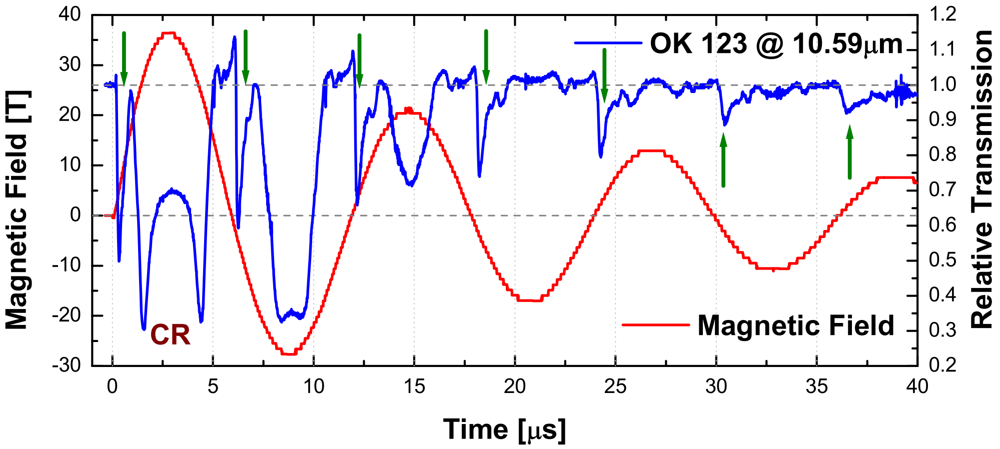

Figure 1.6: *Relative transmission of InSb, sample OK 123, at room temperature with* $\lambda = 10.59\mu m$*. The optical trace shows the electron cyclotron resonance marked CR and an additional feature after each zero crossing of the magnetic field marked by arrows. Further discussion in the text.*

slower sweep rate [15–17].

At this point it must be pointed out that for all three, InSb, HgTe and HgSe the FWHMs of all resonances are larger than 100 ns and the detector bandwidth of 60 MHz there were no contributions from the response function to the resonance strength.

1.6 Conclusions

Three types of hysteretic phenomena reported so far can be distinguished.

- **Type I**: (observed in InAs/AlSb SQWs and quasi bulk HgSe) A relaxation of charge carriers from one energetic level into another. That manifests itself in a characteristic spectrum. In a material showing two resonant features in the rising field trace we find the absorption intensity of one resonant feature increasing vs. the rising field trace at the expense of the other decreasing in the falling field trace while the overall integrated intensity is preserved. This process is reasonably well understood if the two resonances involved are the spin split cyclotron resonances.

- **Type II**: (observed in quasi bulk HgTe) An increase of absorption intensity in the falling field trace for resonant features without decreasing the intensity of any other resonant feature or the overall transmission. Type I and II hysteretic phenomena are modifications to well described resonant features and are explainable with variations of energy level population.

- **Type III**: (observed in bulk InSb) Additional transmission drops, possibly resonant structures, can be observed depending on the sweep rate of the magnetic field generation. These features do only occur in fields generated on a μs-timescale. These phenomena have no slow field or DC field analogon.