Step-scan time-domain terahertz magneto-spectroscopy

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Abstract: We present a novel approach for terahertz time-domain spectroscopy of magneto-optic phenomena. The setup used in this work combines a tabletop pulsed magnet and a standard terahertz time-domain spectroscopy system. The approach is based on repetitive operation of the pulsed magnet and step-wise increment of the delay time of the time-domain spectroscopy system. The method is demonstrated by plotting the magneto-transmission spectra of linearly polarized THz pulses through the hole gas of a Ge sample and the electron gas of GaAs, InSb and InAs samples. Cyclotron resonance spectra are displayed in the frequency range from 200 GHz to 2 THz and for a magnetic field up to 6 T. The GaAs spectra are analyzed in more detail using simulations based on the Drude model.

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References and links
1. Introduction

Terahertz (THz) time-domain spectroscopy (TDS) is becoming most attractive for measuring optical properties of condensed matter systems at THz frequencies and particularly under exposure to high magnetic fields [1–7]. Such measurements are of fundamental importance since they ultimately provide the possibility to determine the magneto-optic tensors that govern dynamical properties of metallic, semiconducting, dielectric and magnetic systems [8]. Recently, we have measured a set of cyclotron resonance spectra in p-doped germanium (p-Ge) in the time domain and at very high speed during a single sweep of a long-duration pulsed magnet [9]. For this experiment, a portable, fiber-coupled THz TDS system [10] employing a high-speed delay line was operated at a pulsed magnet facility. In the present paper, we present a novel approach to measure magneto-optical phenomena, that is based on the repetitive operation of a portable, short-duration (FWHM pulse duration of about 2.5 ms) pulsed magnet placed in a standard THz TDS system [11]. Instead of high-speed TDS measurements, the transmitted electric field is measured step-wise with increments of the delay time after each pulse of the magnet. In the following, we first compare the previous fast-scan with the present step-scan approach. We then give a detailed presentation of the step-scan method, describe the experimental setup and display cyclotron resonance spectra of electrons in n-GaAs, n-InSb, n-InAs and holes in p-Ge. Two-dimensional plots representing the relative change of THz transmittance versus frequency and magnetic field are presented for a frequency range from 200 GHz to 2 THz and a magnetic field up to 6 T.

2. Step-scan approach

A side by side methodology of the step-scan approach introduced in this paper and the fast-scan approach used in [9] is presented in Fig. 1.

In the fast-scan method, a high-speed delay line was used, enabling repetitive measurement of the THz pulses during a single and long-duration shot of the pulsed magnet. Magnetic field pulses of durations in the order of some hundreds of milliseconds were used. In this situation, the scan speed ensures a quasi-constant magnetic field during the measurement of each individual THz time trace. In contrast, the new method bases on the repetitive operation of a short-duration pulsed magnet. As the pulse duration of the used miniature magnet is in the order of a few milliseconds, single-shot or fast-scan measurements are not feasible with the available high-speed delay lines. So, a different approach is introduced here. After each shot (or a definable number of shots) of the magnet, the delay time is incremented. During the magnetic pulse, a voltage induced in a pickup coil providing magnetic field calibration data as well as the electric field detected by the photoconductive detector, amplified and filtered by a lock-in amplifier is recorded. With these data, the electric field of the THz pulse at a given delay time can

References

Fig. 1. Comparison of the step-scan approach with the fast-scan approach from [9]. Instead of repetitive measurements of the time-domain traces during a single shot of the magnet, the electric field of the THz pulse is measured during repetitive operation of a short-duration pulsed magnet. The traces presented here are real measurement data, but trimmed for better readability. Typical values for $\tau_{\text{FWHM}}$ and $T_{\text{rep}}$ are 2.5 ms and about 7 s respectively.

be assigned to the magnetic field strength by interpolation algorithms. By doing so repetitively with successively incrementing the delay time, the (delay) time dependent electric field of the THz pulse for a given magnetic field strength can be extracted.

3. Experimental setup

The experimental setup of the system is shown in Fig. 2.

A Ti:sapphire laser (MiTai from Spectra Physics) is employed as pump source for a standard THz TDS system. Only a small fraction of the available optical output of the laser is used to pump the emitter and detector photoconductive antennas with an average optical power of 20 mW each. In one of the arms, a linear translation stage is implemented as delay line. Two LT-GaAs photoconductive antennas with dipole metallization (dipole length of about 30 µm) are used as emitter and detector. The dipole structure results in a linear polarized emission of THz radiation as well as the detection of (mainly) only one polarization state. Standard Si lenses are utilized to couple the THz radiation from and to the antennas. The THz beam path is built by four off-axis parabolic mirrors providing a focus at which the center of the magnet coil is placed. The emitter antenna is biased by a 50 V RMS AC voltage with a modulation frequency of 57 kHz. The detector antenna is connected to a transimpedance amplifier (amplification of $10^7$ V/A, 3 dB bandwidth of 200 kHz), which is then connected to a lock-in amplifier (Stanford Research Systems SR830). Typical time-constants are set to 30 µs to be able to observe fast dynamics during a shot of the magnet. This is close to the modulation period of 17.5 µs, but feasible for stable modulation techniques (as it is in the case of using a frequency generator). The output of the lock-in amplifier as well as the signal detected by the pick-up coil is read by an A/D card with a sampling rate of 100 kHz and a resolution of 16 bit. For data acquisition,
Fig. 2. Experimental setup used in this work. The output of a mode-locked Ti:sapphire laser with a center emission wavelength of 780 nm is used. LT-GaAs photoconductive switches are used as emitter and detector and are pumped with an average optical power of 20 mW each. Four off-axis mirrors are used to guide the THz beam and to provide a focus, in which the magnet is placed.

measurement program was implemented in LabVIEW™. Each time a magnet pulse is detected, a 50 ms section of relevant data (10 ms section before and 40 ms section after the main peak of the magnetic field) is stored. After this, the delay line is incremented by a certain step size (0.1 ps in the experiments presented here). In the experiments presented in the following, only one shot of the magnet is used for each time delay, but SNR improvements are straightforward by averaging several shots before proceeding to the next time-delay step.

The assembled and disassembled magnet and cryostat are shown in Fig. 3.

Fig. 3. Photos of the assembled (a) and disassembled (b) magnet cryostat. The magnet coil is held by two glass fibre tubes, which are themselves attached to the walls of a small polycarbonate container. This transparent polycarbonate tube is intended to hold the liquid nitrogen during operation and has a slit in the top part for refilling and leakage of nitrogen gas. The outer polycarbonate tube serves as thermal isolation and is flooded by the evaporated nitrogen in the experiment.

The magnet is immersed in liquid nitrogen using a very crude double wall polycarbonate cryostat. The coil itself is 16 mm long and has an inner diameter of 6 mm. It is built of 400 turns of copper wire (with a diameter of 0.4 mm) wound around a fiber glass tube. It provides
a magnetic field up to 10 T with a field inhomogeneity less than 1% along a distance of 3 mm. The coil is held by a pair of wider fiber glass tubes with a diameter of 20 mm and a length of 70 mm, that are capped with a double layer foil window (plastic warp) for thermal isolation. This set of tubes ensures a wide enough optical access to the sample at the coil center. Nitrogen filling of the cryostat is achieved through an upper hole in the outer cylinder and a slit in the inner. Vaporized nitrogen is flooded around the inner vessel and evacuated through a hole at the bottom. This transparent cryogenic system actually provides an efficient cooling of the magnet coil with visual control of nitrogen level. The sample holder consists of a tube of 6 mm outer diameter on which an o-ring with an inner diameter of 3 mm is glued. On top of this o-ring, the semiconductor samples are glued with varnish. In the vicinity of the sample, a resistance thermometer (Pt100) is placed for monitoring of the temperature. This sample holder is inserted to the glass fibre tube supporting the magnet coil.

The magnet coil is electronically fed by a supply connected to a conventional wall plug. It delivers current pulses of several hundred amperes with a pulse duration of about 2.5 ms (FWHM pulse duration, depending on the peak current). Two capacitors of 1 mF each, store a maximum energy of 250 J at a peak voltage of 500 V. This energy is converted into magnetic energy by controlled discharge of the capacitors to provide pulses with peak magnetic fields of up to 10 T. At the same time, the energy ends up into Joule heating and then finally into nitrogen evaporation directly observed after each shot. Photos of the tabletop electric supply, refilling of the pulsed magnet during operation and the sample holder with pickup coil and sample are shown in Fig. 4.

![Fig. 4. (a) Tabletop power supply of the pulsed magnet employing two 500 V, 1 mF capacitors. (b) Liquid nitrogen refilling of the pulsed magnet during operation. (c) Sample holder with pickup coil.](image)

4. Results

The change in the detected THz electric field amplitude for a n-doped GaAs sample and a p-doped Ge sample are shown in Fig. 5 and Fig. 6, respectively. In these intensity plots the difference \( \Delta E(t_{\text{meas}}, \tau_{\text{delay}}) = E(t_{\text{meas}}, \tau_{\text{delay}}) - E(0, \tau_{\text{delay}}) \) is displayed together with the magnetic field \( B(t_{\text{meas}}) \). The temperature measured by the resistance thermometer was about 120 K in both cases.

The change of the THz waveform induced by the magnetic field is clearly visible and in general follows the trend of the magnetic field. For both samples, the first etalon echo (which is of course also influenced by cyclotron resonance) is observed. Even already in this time-domain data it is obvious, that p-Ge possesses a more complex behavior. This can be deducted most clearly from the down-sweep (of the magnetic field, starting from 5 ms magnet time) characteristics. In the p-Ge data, a low frequency component (heavy hole resonance as will be
Fig. 5. Change of the detected THz electric field amplitude as a function of delay time and magnet time for n-GaAs displayed as an intensity plot as well as conventional plots extracted thereof. The THz amplitude values for no magnetic field are subtracted from the measured data to obtain these differential plots. In addition, the magnetic field pulse is shown.

shown later) is present at the highest field and is accompanied by a high frequency component in the down-sweep, starting from about 7 ms magnet time. In the n-GaAs sample data, the frequency of the resonance seems to change up to a magnetic field of 4 T and then builds a plateau. This feature is not caused by cyclotron resonance, it occurs at frequencies lower than the collision frequency, where featureless absorption decreases at increasing magnetic fields.

To obtain the change of the THz transmittance induced by the magnetic field, the THz waveforms \( E(\text{meas}, \tau_{\text{delay}}) \) were numerically transformed to waveforms dependent on the magnetic field \( E(B, \tau_{\text{delay}}) \). Afterwards, they were Fourier-transformed and referenced to the data at no magnetic field.

For magneto-optical transmission spectroscopy measurements of dilute plasma systems, the relevant information can be displayed by two-dimensional plots representing the relative change of the transmitted intensity dependent on the frequency and the magnetic field

\[
\left( \frac{E(\omega, B)}{E(\omega, 0)} \right)^2 - 1. \tag{1}
\]

In the limit of low-density plasma, where the sample reflectance does not vary significantly in the measured THz frequency range, such transmittance spectra are proportional to the change of absorbed power in the sample under exposure to the magnetic field [12]. In these systems the absorbed power is dissipated by the current \( J_x \) driven by the linearly incident polarized wave \( E_{\text{inc}} = E_0 e^{-i\omega t} \). In this case, the power absorbed per unit volume is

\[
P = \frac{1}{2} Re(J E^*) = \frac{1}{2} Re(\sigma_{xx}) E_0^2. \tag{2}
\]
Fig. 6. Change of the detected THz electric field amplitude as a function of delay time and magnet time for p-Ge displayed as an intensity plot as well as conventional plots extracted thereof. The THz amplitude values for no magnetic field are subtracted from the measured data to obtain these differential plots. In addition, the magnetic field pulse is shown.

It turns out, that the transmittance spectra of a wave polarized along the x-axis directly display the real part of the diagonal conductivity tensor \( \text{Re}(\sigma_{xx}) \). In the following, we present THz transmittance spectra for bulk n-GaAs, p-Ge, n-InSb and n-InAs quantum well. The results for GaAs will be analyzed using the low-density limit approximation, while the ones of the other samples are only briefly commented.

Figure 7 shows experimental and numerical results of the relative change of THz transmittance dependent on the frequency and magnetic field for a bulk n-type GaAs thick layer. This sample was grown by liquid phase epitaxial technique for implementing sensitive photoconductive detectors with high THz absorption from 1s to 2p impurity transition at liquid-helium temperature [13]. The layer thickness was 60 µm with an electron concentration of \( 1.5 \times 10^{15} \) cm\(^{-3} \). Measurement was performed at 120 K. Obviously, two striking features appear in both plots of Fig. 7. A broad line of decreased transmission that saturates at 50% of the zero field transmission value and a green area below a threshold frequency that displays increased transmission. The average slope of the absorption line and the resonance equation

\[
m^* = \frac{eB}{\omega_c}
\]

yield an effective mass \( m^* \) of \((0.075 \pm 0.005) m_0 \). This value is higher than the accepted value of 0.067 \( m_0 \) for electrons at the bottom of the conduction band, but nevertheless consistent with recent temperature-dependent measurements [14]. The above spectral features can be explained by numerical simulations based on the Drude model. The absorbed power per unit volume is given by

\[
P = \frac{1}{4} \sigma_0 E^2 \left( \frac{1}{1 + (\omega_c - \omega)^2 \tau^2} + \frac{1}{1 + (\omega_c + \omega)^2 \tau^2} \right).
\]
This expression includes two contributions, which correspond to the two circularly polarized waves of equal amplitude that combine in the linearly polarized wave. For the case $\omega \tau > 1$, according to Eq. (4), the wave mode that rotates in the same sense as the electrons gets resonantly absorbed and therefore called cyclotron resonance active (CRA) mode, while the other mode, denoted cyclotron resonance inactive (CRI), is practically unchanged by the magnetic field. This is shown in Fig. 7, where, however, the CR line develops a saturated plateau. Indeed, the numerical simulation demonstrates that, after crossing the 60 µm thick layer, the transmitted power of the CRA wave vanishes in a wide frequency range around the resonance peak. In this case, because the CRI is not changed by the magnetic field, the transmitted THz intensity drops to 50 % near the resonance peak and a saturated plateau is observed around the CR line. On the other hand, at low frequencies ($\omega \tau < 1$), the absorption of both CRA and CRI waves decreases with magnetic field according to Eq. (4) and gives rise to the green area. The threshold frequency gives a fair estimate of the reciprocal collision time $\tau$, which is about 4 ps here, in agreement with the measured mobility value at this temperature of about $8 \cdot 10^4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.

Figure 8 shows experimental results of the measured relative change in the THz transmittance for p-Ge, n-InAs and n-InSb (with the same scale for the magnetic field for comparison). The free holes concentration is $2 \cdot 10^{14}$ cm$^{-3}$ for the p-Ge sample, free electrons concentration $3 \cdot 10^{14}$ cm$^{-3}$ for n-InSb and $5 \cdot 10^{11}$ cm$^{-2}$ for the InAs quantum well. As before, the intensity at a given frequency is normalized to its zero magnetic field value.

For the p-Ge data, two hole cyclotron resonances (light and heavy) and a weak resonance in between due to quantum mechanical effects [15] are observed. The lines slopes give light and heavy hole effective masses of 0.04 $m_0$ and 0.3 $m_0$, respectively. The absence of green area
indicates a lower value of collision frequency that falls in the grayed out area of poor SNR. The obtained results agree very well with the results from the fast-scan measurements with a long-duration pulsed magnetic field up to 12 T [9], but provide a broader frequency range as well as an improved SNR. Again, only single shots of the magnet between step-wise increments of the delay line were used. Due to the use of lock-in technique and a longer effective measurement time, the results presented here have a higher SNR (in the order of $10^2$) and provide access to a broader spectrum compared to the fast-scan technique (SNR in the order of 10).

The plots for InSb and InAs are limited to fields below 1 T and 2.8 T and they are only presented here for illustrating the step-scan technique. The average slope effective masses of $0.02 \, m_0$ and $0.035 \, m_0$ for InSb and InAs, respectively, deviate significantly from accepted values of $0.014 \, m_0$ and $0.026 \, m_0$ [16]. However, detailed analysis explaining the deviations, including magneto-plasma effects [4, 12], is outside the scope of this paper.

5. Conclusion

We have demonstrated a novel and simple approach for THz magneto-optical spectroscopy, which is based on a portable, repetitive pulsed magnet combined with a standard THz time-domain spectroscopy system. Magneto-transmission spectra of linearly polarized THz pulses through semiconductor layers of GaAs, Ge, InSb and InAs were displayed in the frequency range from 200 GHz to 2 THz and for magnetic field up to 6 T. The data were obtained with a single magnet shot between step-wise increments of the TDS delay line. It is straightforward to improve the signal to noise ratio of the measured THz electric field amplitude by averaging multiple shots between delay line increments or by increasing the magnet duty cycle. This
tabletop step-scan time-domain THz magneto-spectrometer based on a liquid-nitrogen cooled resistive mini-coil can be implemented at higher magnetic fields and broadly used for further magneto-optics experiments.

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