Laboratory Course

Time-Domain Terahertz Spectroscopy

Instruction Manual

Contents

1 Aim

- Measure the Terahertz transmission spectra of water vapour and assign the THz absorption lines from the transmittance spectra.
- Measure the thickness of a silicon wafer using THz time domain spectroscopy and calculate the transmittance of Silicon in the THz regime.
- Perform the THz transmission spectroscopy through an α-lactose pellet. Study the absorption lines of the α-lactose monohydrate from the THz transmission spectra.

2 Background

2.1 Terahertz Time Domain spectroscopy

Terahertz time-domain spectroscopy (THz-TDS) is a spectroscopic technique in which the properties of matter are probed with short pulses of terahertz radiation. Terahertz (THz) spectrum refers to the frequency domain ranging approximately from 100 GHz to 10 THz, corresponding to wavelengths from 3 mm to 30 μm.

The generation and detection scheme is sensitive to the sample's effect on both the amplitude and the phase of the terahertz radiation. By measuring in the time-domain, the technique can provide more information than conventional Fourier-transform spectroscopy, which is only sensitive to the amplitude.

THz-TDS requires generation of an ultrafast (thus, large bandwidth) terahertz pulse from an even faster femtosecond optical pulse, typically from a Ti-sapphire laser (Mai Tai laser). That optical pulse is first split to provide a probe pulse that undergoes an adjustable path length adjustment using an optical delay line. The probe pulse probes the detector that is sensitive to the electric field of the resulting terahertz signal. By varying the path length traversed by the probe pulse, the test signal is thereby measured as a function of time. The details of the THz spectroscopy can be seen in the reference [1].

2.2 THz Emission and Detection

III-V and II-VI semiconductors have served as reliable sources of THz radiation. Femtosecond optical excitation of semiconductors leads to the emission of THz radiation due to mostly two phenomenon 1) Transient photocurrents and 2) Nonlinear optical effects in the crystal. The nature and origin of the photocurrents and nonlinear phenomenon vary vastly across the

semiconductors. The structural and optoelectronic make-up of the semiconductor determines the strength of these emission mechanisms and emission amplitude.

2.2.1 Photoconductive antenna emitter

One way to enhance THz emission is to improve the strength of transient currents. Photoconductive antenna (PCA) is the perfect example for this. The schematic of a PCA is shown in Figure 1. The PCA has a semiconductor substrate with biasing electrodes often very closely spaced, in order to achieve a high electric field. On one side, the ultrafast, above-bandgap optical pulse creates photoexcited carriers that are accelerated by the application of a dc-bias and then recombine. The short duration of the excitation and the short lifetime of the photoexcited carriers generate a current transient, emitting a THz frequency pulse. The transient drift current, due to this artificially provided field, creates a much larger THz emission. PCAs offer a number of control parameters like bias voltage, antenna gap, antenna geometry, substrate material etc., which offer control over the THz amplitude and spectrum.

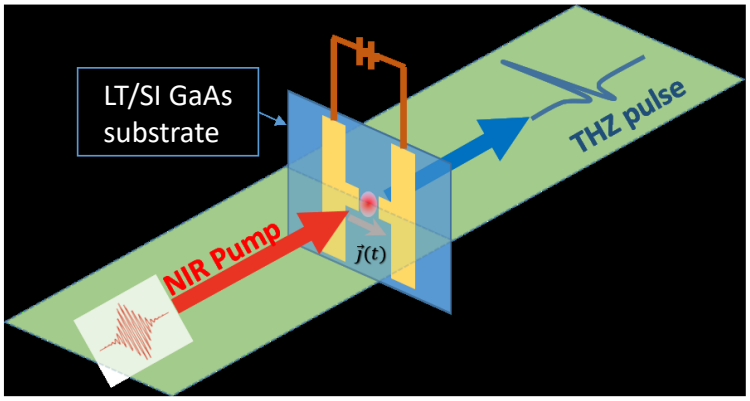

Figure 1. *Schematic of the THz photoconductive antenna emitter. Metallic antenna structure is fabricated on high resistivity substrate like LT- GaAs. The NIR pump generates the photo carriers at the antenna gap and the external bias drives the carriers to create ultrashort transient current j(t). This transient current j(t) acts as the source of THz radiation.*

Additionally, the current surge is oriented parallel to the surface, the favoured orientation for better outcoupling. The PCAs are designed to improve the generation and outcoupling of THz radiation. They offer far superior THz amplitude compared to ordinary unbiased semiconductor emitters.

2.2.2 Photoconductive antenna Detector

The detection of THz pulses usually relies on photoconductive and nonlinear optical techniques, just as in the case of generation of THz radiation. Let us briefly discuss one of the most popular detection techniques for THz radiation namely photoconductive antenna.

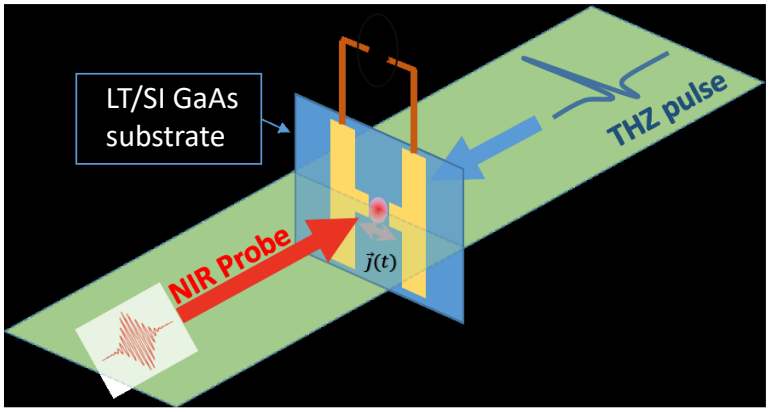

Figure 2. *Schematic of the THz photoconductive antenna detector. The construction is very similar to that of a PCA emitter except that there is no external bias. The NIR probe generates the photocarriers at the antenna gap and the incoming THz electric field drives the photocarriers. The resulting voltage/current across the antenna gap is measured.*

PCAs can function as THz detectors via the photoconductive mechanism. The construction of a detector PCA is similar to the emitter PCA in the sense that they have a pair of metal electrodes fabricated on a semiconducting substrate like LT-GaAs. The above-bandgap pumping of the semiconductor gap introduces new charge carriers in the antenna gap. The incoming THz electric field drives the new carriers and creates a charge separation or a current across the electrodes. Measuring the THz induced current enables us to measure the THz field. By varying the time delay between the THz pulse and NIR probe, one could map out the THz pulse shape. The Figure 2 shows the schematic of THz detection in PCAs.

Further details can be found in the following link, PCA – photoconductive antenna.

2.3 Advantage of THz-TDS

The THz time domain spectroscopy has many advantages to offer. Let us discuss some of the advantages of this scheme compared to other modes of spectroscopy and DC measurements.

- **Escape Kramer Kronig analysis**

THz TDS directly measures the complex valued transmission/reflection of materials, which lets us calculate the complex valued conductivity, dielectric function and refractive index. Spectroscopy in other branches of optics measures the intensity and hence has to rely on tedious Kramer Kronig analysis to determine these complex quantities. Easy access to these functions using THz-TDS helps easily understand the nature of conductivity in the materials.

- **Nanoscopic probing**

A common assumption is that the length scale probed by a radiation of particular frequency is comparable to the distance carriers diffuse during the period of one oscillation. According to Einstein's diffusion theory (1), one could roughly estimate the length scale probed by a radiation (ω=$2\pi\nu$) by the following the equation:

$$(1)\quad l(\omega) \approx \sqrt{\frac{D}{\omega}}$$

Where D is the Einstein's diffusion coefficient of the carriers in the medium. For Instance, in the silicon, with a diffusion coefficient of nearly 36 cm^2/s , investigation with a bandwidth of roughly 0.4 to 2.5 THz probes the length scales in the range 10 to 20 nm. The THz response in this case reflects the conductivity dynamics in this length scale. The THz spectroscopy hence becomes particularly useful in the cases where the nanocrystalline dimensions are in this length scale. The usual THz spot sizes are of the order of millimeters. In such cases, the THz response indicates the space averaged nanoscopic conductivity of the material. Carrier localization and backscattering effects are also prevalent at these dimensions.

- **Problems with DC measurements**

Standard DC measurements like Hall Effect, van der Pauw, FET measurements etc. do offer conductivity parameters. However, these schemes have other parasitic effects that arise from contact resistance, Schottky junctions etc. In addition, there could be decay and damage to the sample at the metal contact etc. THz technique is immune to these issues. Moreover, many studies have shown the consistency of THz-TDS results with the standard DC measurements.

- **Non-destructive testing**

Measuring the THz response is a non-destructive way of probing the samples. Additional processing like metallization or contact formation (which are necessary in the DC conductivity measurements) are not required for THz measurements. This leaves the sample intact and reusable in other applications.

3 Experiment

3.1 THz-TDS experimental setup

The schematic of the THz setup is shown in figure 3. We use a femtosecond laser source (Mai Tai Spectra-Physics) with a central wavelength of around 800 nm for the optical excitation of the THz emitters. The 100 fs pulses arrive at a rate of 80 MHz. Lenses are used to control the fluence and excitation spot size. Attenuators achieve additional control over the excitation density. A fraction of pulse energy is redirected to gate the detector PCA. ND filters are used to dial down the probe power reaching the PCA detector.

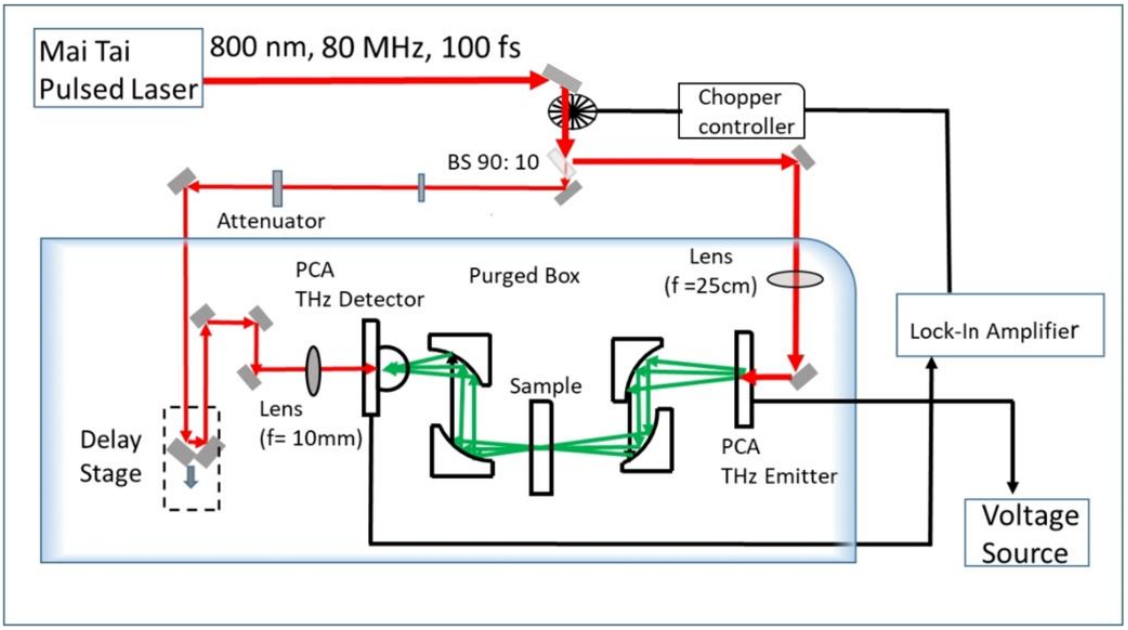

Figure 3. *The schematic of the experimental system for performing THz time domain spectroscopy. The system uses interdigitated PCA (iPCA) as the source of THz radiation and a bowtie PCA as detector. The system measures the THz transmission of the system under study.*

3.2 Measurement Procedure

The signals detected by PCA are very weak. The extraction of such tiny signals is usually done with a lock-in amplifier (LIA) and a mechanical chopper, which periodically blocks the optical beam. The LIA locks the internal signal amplification to the frequency, at which the laser beam is chopped. Instead of using a mechanical chopper, here the reference of the LIA is given from the waveform generator, which is shown in the figure 4. This suppresses the background as well as the noise. We apply an alternate bias at 415Hz to the THz emitter (Fig. 4). The fundamentals of the waveform generator are given in the appendix 4.2. Fundamentals of LIAs can be found in appendix 4.3.

The PCA, with the aid of Lock-in technique, reads the electric field at that part of the pulse that temporally overlaps with the arrival of gating pulse. By varying the relative delay between the THz pulse and the NIR gating pulse, one can map the entire THz pulse. The delay between them is adjusted by using a linear translational stage. The apparatus consists of mirrors mounted on a motorized translational stage whose motion alters the THz-NIR delay. For instance, the movement of the delay stage by 10 μm would create a delay of $\approx 66 fs$.

The Fourier analysis of the pulses contains amplitude as well as phase information of the frequency components. We have used a Lab-View program to automate the entire process of data collection. All the parameters of the lab view program are given in the appendix 4.4. The program controls the precise movement of the delay stage and reads the lock-in readings at every step of the delay stage motion. This way a signal of the desired length and temporal resolution can be recorded for the chosen lock-in parameters. The emission amplitude determines the lock-in time constant and delay stage wait-time. A weak signal requires a longer averaging time on the lock-in amplifier. The data acquisition of the THz-TDS is shown in figure 4.

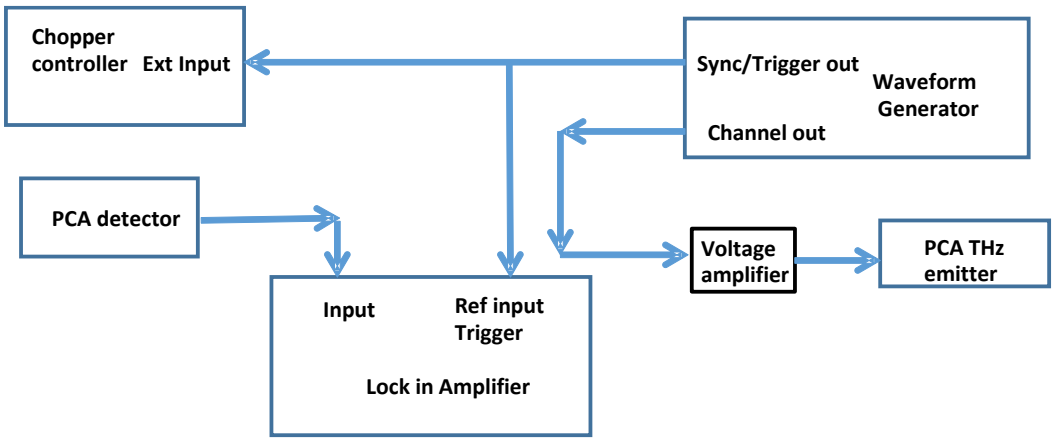

Figure 4. *THz-TDS signal acquisition method*

3.3 Measurement Plans

Task 1: Measuring the signal to noise ratio of the THz spectrum.

- Perform the THz transient scan in the purged chamber, at least three times.
- Perform the THz transient scan after blocking the THz beam, at least three times.

How do you calculate the S/N?

Task 2: Understanding the water vapour absorption lines in the THz regime using THz-TDS spectroscopy.

- Record two THz transients, one with purging and one without purging with dry air. Both scans should have the same initial position and total number of points.
- Each of the scans should be taken at least three times for averaging for better signal/noise ratio. How do you interpret the results?

Task 3: Measure the THz transmission of a Silicon substrate and find the thickness of the substrate from the time domain signals.

- Measure two THz transients while the chamber is purged with dry air. One without the Si sample and one with. The signal obtained with the sample will have a reduced field and will be shifted in the time domain. Why?
- Each of the scan should be taken at least three times for averaging for better signal/noise ratio. How do you interpret these observations?

Task 4: Understanding the THz absorption lines of an α-lactose monohydrate (α-LM) pellet using THz-TDS spectroscopy.

- The experiment will be conducted with an α-LM pellet with a thickness of ~2mm. The pellet was prepared by pressing 0.8 grams of α-LM powder for 20 minutes under pressure of 220 kN.
- Measure two THz transients while the chamber is purged with dry air. One without the α-LM pellet sample and one with. The signal obtained with the sample will have a reduced field and will be shifted in the time domain. Why?
- Each of the scans should be taken at least three times for averaging for better signal/noise ratio. How do you interpret these observations?

3.4 Data Analysis

Data analysis will be done in MATLAb.

1) Plot the S/N on the same plot as the signal for both the time domain and the frequency domain. Calculate the value of the S/N for both. Note that you can use the MATLAB function **snr** (snr MATLAB documentation).

2) The time domain sample signal is denoted as $E_s(t)$. The time domain reference signal is denoted as $E_R(t)$, which is taken without the sample.

3) Make sure to average all measurements and plot the time domain spectra of the sample and the reference signals on the same plot.

4) Perform the Fourier transform on the time domain sample and reference signal and obtain $E_S(\omega)$ and $E_R(\omega)$ respectively. The Fourier transform can be done using the **FFT** MATLAB function (fft MATLAB documentation). Plot the frequency domain spectra of the sample and the reference signals on the same plot.

5) The transmittance can be calculated by taking the ratio of the FFT magnitude of the sample and the FFT magnitude of the Reference signal, $T=\frac{E_S(\omega)}{E_R(\omega)}$. Plot the transmittance.

   NOTE: Adjust your frequency axes to fit the appropriate THz range.

6) Compare the absorption lines in the transmittance of the water vapour with the previous literature. You can follow the reference [2].

7) The calculated Thickness of the Si wafer from the THz measurement should be near ~500µm. The thickness of the silicon wafer can be measured from the time domain as well as frequency domain THz spectra. Show the results of both calculations.

   - **Thickness measurement from the THz Peak position shift in the Time domain signal.**
     A. Utilize THz signal in air as a reference.
     B. If the peak position shift is Δt, then the thickness can be calculated using the following equation:

$$L = \frac{c\Delta t}{n_1 - 1}$$

Where c is the speed of light in air, and n_1 is the refractive index of Si.

- **Thickness measurement from the frequency domain spectra.**

    A. Perform the Fourier transform of the time domain THz signal for the sample and reference measurements.
    B. If the distance between peaks in the FFT magnitude is Δν, then the thickness can be calculated from the following equation:

$$L = \frac{c}{2\Delta\nu n_1}$$

8) The percent transmittance of the silicon wafer in the THz regime should be 50-55%. Calculate the transmittance of your measurements using the time-domain spectra using the following equation, $\%T = \frac{E_{PP_S}(t)}{E_{PP_R}(t)} \cdot 100\%$, where $E_{PP}(t)$ is the peak-peak value of the THz time-domain signal. Why can we calculate the percent transmittance this way?

9) Compare the THz transmittance spectra and the absorption line of α-Lactose pellets with the previous literatures [3].

4 Appendix

4.1 Safety Instructions

1. Improper use of instrumentation may cause damage or may be even dangerous. It is absolutely necessary that you follow these safety instructions.

2. Use only the equipment, which was given to you by the lab instructor.

3. Due to the high laser power, wearing laser goggles is mandatory. Ask the instructor for appropriate laser goggles.

5. Do not change or touch the mirror optics on the optical table. Slight misalignments are fatal!

6. Do not change electrical wiring or voltages of the devices unless instructed by the instructor.

We understand that you are familiar with these safety instructions and that you accept them.

4.2 Settings of Waveform Generator EDU33212A

Caution:

- There is no need adjusting the settings of the EDU33212A. The instructor has done this for you.
- In case you want to change the settings: call the instructor. He will supervise your work.
- **Careful:** The output of the waveform generator is fed to the PCA THz emitter. In case if the voltage exceeds +5V, then the THz emitter will be damaged

The waveform generator is giving a square wave electrical output with a duty cycle ~8% and frequency of 415 Hz. All the parameters are shown below:

Frequency: 415 Hz

High level: +5V

Low level: 0V

Phase: 23^0

Duty Cycle: 8%

4.3 Settings of Lock in Amplifier SR830

Caution:

- There is no need to adjusting the settings of the EDU33212A. The instructor has done this for you.
- Wrong settings will give erroneous measurements.

The parameter of this instrument is given below:

Time constant: 100ms

Sensitivity: 2 mV/nA for reference measurement, 100μV/pA for sample measurement

Slope: 12 dB/oct

Sync filter: off

Reserve: Normal

Signal Input: A, **Coupling:** AC, **Ground:** Ground

Trigger: Ref. in

4.4 Setting of Lab View Programme:

Caution:

- There is no need adjusting the settings of the EDU33212A. The instructor has done this for you.
- In case you want to change the settings: call the instructor. He will supervise your work. The parameters of the lab view programme is given below

Step size: 5μm

THz axis: 1

Sample axis: no need to change.

Right limit: 1000

Lock in GPIB: 8

EPS300: 1

5 References

[1] J. B. Baxter and G. W. Guglietta, *Terahertz Spectroscopy*, Anal. Chem. 83, 4342 (2011).

[2] X. Xin, H. Altan, A. Saint, D. Matten, and R. R. Alfano, *Terahertz Absorption Spectrum of Para and Ortho Water Vapors at Different Humidities at Room Temperature*, J. Appl. Phys. 100, 094905 (2006).

[3] Euna Jung, et al. *Terahertz time domain spectroscopy of crystalline α-lactose monohydrate*, Biochip Journal 2.4 (2008).