3 Terahertz Time-Domain Spectroscopy

In contrast to the electronic approach the photonic THz techniques are based on lasers. Continuous wave lasers, pulsed lasers with nanosecond pulse length as well as ultrashort pulsed lasers in the sub 100 fs regime are used. A big progress has been made towards compact and reliable devices since the realization of the first femtosecond (fs) laser source. Thus, it is now possible to use this photonic high-end technique not only in research but also in real world applications.

The advantages of mode-locked femtosecond laser sources are high peak power and a broadband spectrum. Fourier limited pulses are delivered by solid state Ti:sapphire lasers. Most versions are also tunable in wavelength, sometimes even between 690 nm and 1020 nm. Depending on the particular application and THz setup, some specifications are more important than others. The relevant devices are introduced in this chapter.

3.1 TDS Principle

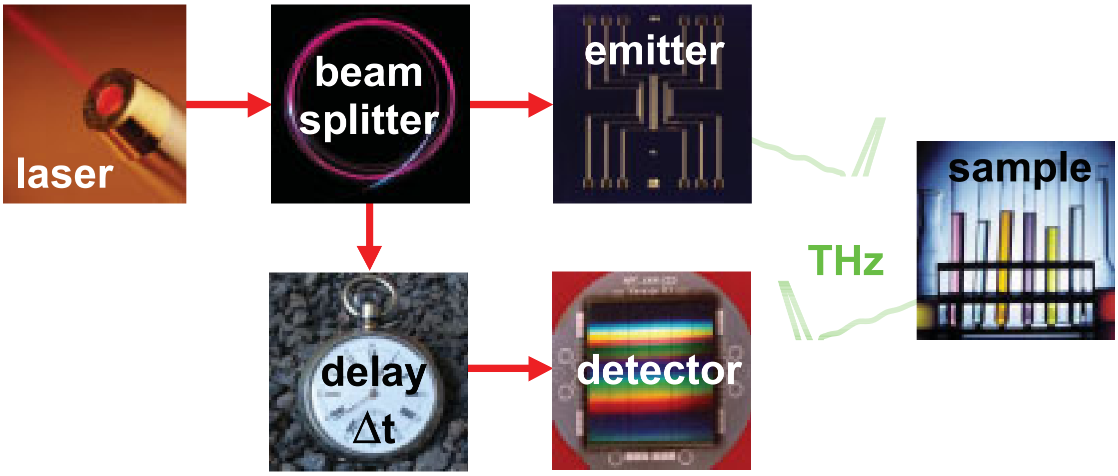

Figure 3.1: Principle of the THz time-domain spectroscopy system (THz-TDS). Major components: laser, emitter, detector and a delay producing device

The basic principle of terahertz time-domain spectroscopy (THz-TDS) is to sample a fast transient slowly by coherent detection (Fig. 3.1). Here the same laser pulse is split into two fractions which are used for the generation of THz radiation in the emitter and to gate the detector, respectively. The pump pulse length and the integration time of the detector are smaller than the THz pulse length. So the detector can sample the THz pulse with small temporal gates stepwise as a function of delay Δt. The temporal delay is typically caused by a spatial displacement of a mirror. So high frequencies can be sampled. By changing the runtime between pump and detector pulse, the electric field (not only the intensity) can be detected. A following Fourier transformation shows the spectral amplitude and phase. Further on, the repetition rate of the laser is much higher than the inverse pulse length, so the detector is insensitive most of the time. For typical experimental conditions it is only active for $10^{-3}\,\%$ of the time (duty cycle). If during that period the THz pulse is present, the detector gives a signal. Otherwise it only integrates over environmental electromagnetic noise. For the rest of the time it gives only a neutral signal, depending on the type of detector [5]. This reduces the noise of the system considerably.

3.2 Pulse Propagation and Data Processing

Since in THz-TDS systems electric fields are detected, a Fourier transformation can bring the spectral amplitude including intensity and phase. To calculate this with a computer algorithm like the Fast-Fourier-Transform (FFT) procedure it is important to have equally spaced supporting data points in the time domain. This can be assured either by moving the delay line to the appropriate discrete positions (stepwise) or by synchronized readout of the lock-in amplifier and a continuously running linear stage (on-the-flight).

The mathematic algorithm expects 2^n data points. If the data set does not fulfill this condition one can add zeros at the end of the array (zero-filling). But this can bring residuals and changes in the recorded line shape. If the FFT is calculated using Origin® the software will automatically fill up zeros pretending to have a higher spectral resolution.

The mathematical relation of the temporal distance between the data points Δt and the frequency resolution $\Delta\nu$ is given by the number of data points in the time domain N

$$\Delta\nu = \frac{1}{N\Delta t} \tag{3.1}$$

This is the reason for scanning long delay times in the TDS. The longer the scan, the more independent data points can be recorded and thereby the spectral resolution is increased.

According to the Nyquist-Shannon sampling theorem the maximum detectable frequency $\Delta\nu_{\mathrm{max}}$ is

$$\Delta\nu_{\mathrm{max}} = \frac{1}{2\Delta t} \tag{3.2}$$

Higher frequencies can not be detected, they are canceled out by the sampling technique. In the experimental realization this is not a strict cut-off relation but more a steady

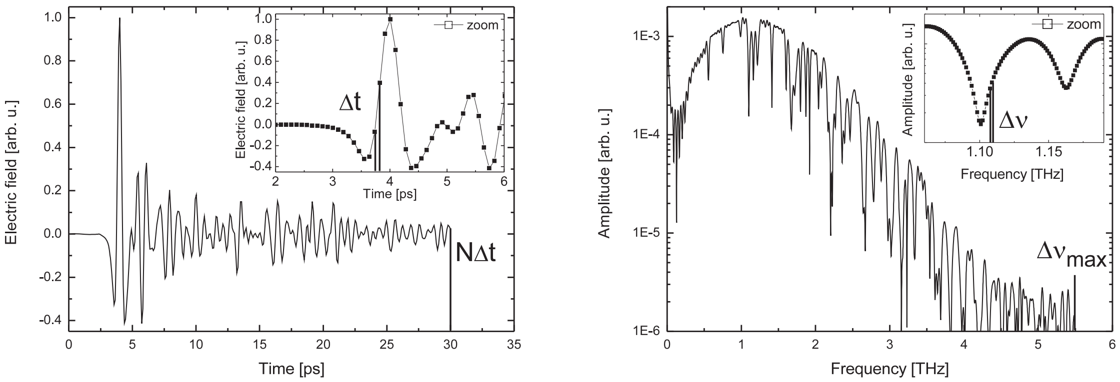

Figure 3.2: Relation between the time and frequency domain: Fourier transformation. The spectral resolution $\Delta\nu$ and the maximum detectable frequency $\Delta\nu_{\max}$ is defined in the time domain by data point spacing Δt and the amount of data points N.

limiting factor for the high frequency limit. If the time constant of the lock-in amplifier is too long or the velocity of the delay line is too fast, the high frequencies are not detectable, limiting the accessible bandwidth despite dense sampling.

If we record the electric field $E_{sample}(\omega)$ with a sample in the THz beam then the complex wave vector $k(\omega)$ will influence the transmitted pulse. But all the characteristics of the combination of emitter and detector as well as the properties of the imaging optics like mirrors and lenses are included in the measured electric field. Also the humidity will show a strong absorption at discrete lines (see chapter 8.6).

Therefore typically a reference $E_{ref}(\omega)$ is recorded without a sample to get rid of all system parameters besides the changes induced by the device under test. A division of these two fields (see eq. 2.10) brings the sample's transmission:

$$T(\omega) = \frac{E_{sample}(\omega)}{E_{ref}(\omega)} = e^{-\frac{\alpha z}{2}} e^{i[k_0 + \Delta k(\omega)]z} \tag{3.3}$$

So for a known propagation length or sample thickness z the material constants refractive index n and absorption coefficient α can be calculated. Further details on the evaluation of the particular parameters with respect to imaging applications are discussed in chapter 8.

3.3 Optics

The most obvious difference between the NIR and THz waves is that typically optical devices like lenses or mirrors have extents larger than the used wavelength for the NIR while in the THz range the wavelength reaches the same order of magnitude like the geometrical dimensions of the devices itself. So much higher precautions have to be taken into account for the THz waves when it comes to coherence, diffraction or scattering.

As for all optical sources also the THz emitters have an inherent divergency while the detectors show a limited viewing angle. The THz beam path between these two, the more precise: the transfer function, has to be optimized in terms of amplitude, focus diameter, astigmatism, frequency distribution, additional echo pulses and so on.

A surface emitter (chapter 4.2) itself does not need additional optics. The pump beam defines the divergency of the THz beam because it is a diffraction limited source. So if placed in the focus of a mirror, a good beam quality is expected. But for a photoconductive switch (PCS) collimating optics are essential due to the high divergency of the point source and small angle of total internal reflection. Silicon (Si) is very well suited to be used as a substrate for THz transmission optics. The refractive index is quite high ($n_{\mathrm{Si}}^{\mathrm{THz}} = 3.41$ [6]) and it shows only very low dispersion and small absorption. The design of a Si lens is important to increase the angle of view of the emitter and to adjust the THz beam to the application. Different lens layouts are proposed in literature depending on the particular system. Various calculations and simulations as well es experiments have proven this [7–9]. So a good spectral density and a collimated beam with a reduced divergency can be reached.

There is also the possibility to work without a Si lens. But then there are always strong echoes due to multi Fresnel reflections in the substrate. Typically Si has nearly the same refractive index as GaAs ($n_{\mathrm{GaAs}}^{\mathrm{THz}} = 3.59$ [10]) and so only a negligible reflection at the interface occurs. Also sapphire [11] or germanium optics are in use. Anti reflection coatings are difficult to be fabricated for the THz range. The coating layer thicknesses have to be in the wavelength range which is some tens of microns. This is much more than for visible optics. So different techniques are elaborated to produce appropriate optics also for the THz range (Si: [12], Ge: [13]).

Another possibility is to operate the PCS in reflection. This means one illuminates the PCS on its metallized side and detects not the THz radiation that is generated in forward direction but to look at the THz radiation which is emitted back towards the exciting laser beam. Here the dispersion in the PCS is minimized because the THz radiation does not have to propagate through the substrate by the expense that no Si lens can be attached and echoes and divergency problems occur unless otherwise compensated.

The design of all optics is influenced by the large bandwidth of the THz pulse with respect to the center frequency. So the difference in divergency within the THz spectral contributions can be observed, especially for a focussed system [14]. Due to diffraction theory the high frequency part of the THz spectrum can be focussed to a smaller focus than the long wavelengths.

This fact can also be used to restrict the broadband spectrum to a higher center frequency. A pinhole next to the emitter or next to the focus can bring a better spacial resolution [15, 16]. Then the pinhole acts as a frequency filter. The same result can be obtained by evaluation of the THz data in the frequency domain. This would be a kind of software-pinhole allowing the measurement at particular frequency bands. Various attempts have been done to transmit THz pulses on metallic [17] or plastic wave guides [18, 19] to control the propagation. But high losses, dispersion, diffraction and bad coupling efficiencies limit the possibilities for this approach up to now.

For the THz range only a few techniques are known to produce anti-reflection coatings for optics. Therefore, reflective optics are preferable to transmissive optics if available. The optics used within this thesis are off-axis parabolic mirrors to guide the THz beam. Their advantages are:

- no echo pulses, refractive index steps
- high reflectivity, protected gold coating
- large area, angle of view

The particular used models depend on the device and the required focus. If space is not a problem and a long Rayleigh length is needed, the particular mirror (Melles Griot 02POA017) is an off-axis parabolic with 63.5 mm diameter at a focal distance of 120 mm. The protected gold coating has a reflectivity of better than 95 % in entire THz range.

3.4 Electronic Devices

Typical THz pulses show a spectral distribution between 100 GHz and 4 THz. Compared to commercially available electronic devices and wirings like high-speed network adapters or network cables THz frequencies are a much higher . Today's network standards have stated working frequencies of 100 MHz (Cat-5 standard). A plug like a SMA connector has a bandwidth with a cut-off frequency of 34 GHz. So by using nowadays wires it is impossible to transport signals with THz frequencies. It should be mentioned that there is serious progress in guiding THz pulses on free-space wires [20] or fibers [21] and attempts to use a complete free-space solution for THz communication [22]. Here one further advantage of TDS can be seen: If there is no movement in the time domain (the linear stage is standing still), it is nearly a DC case for the electronics like the preamplifier. So to sample THz frequencies already slow components with bandwidths in the kHz range are sufficient. Of course, to increase the signal-to-noise ratio and to reduce the data acquisition time, high frequency devices are advantageous. The measured signals can be guided in standard wires and BNC cables.

Lock-in Detection

Most of the presented THz signals in this thesis are recorded using a lock-in amplifier. The used model is the Stanford Research SR830. It offers a sensitivity range between 2 nV and 1 V at a frequency response of 100 kHz. The function is as follows: The interested signal is chopped (periodically modulated) and the lock-in amplifier provides at the output the fraction of signal that is modulated with the same frequency canceling out most other frequencies and DC parts. In the experiment this is realized by using a mechanical (optical) chopper for the laser pulses or a modulated bias for the emitter antenna. Only for the enhancement cavity the chopper was used in the THz beam because of electronic reasons (control circuit). So also weak signals on a high background noise level can be detected. Offset signals and periodic modulations at other frequencies (like the 50 Hz emission from power supplies) are suppressed very accurately. THz antennas are sensitive to the whole electromagnetic spectrum (with a reduced sensitivity) even without illumination. Therefore also the electromagnetic pollution at 50 Hz or the high frequencies of e. g. a fluorescent lamp at up to 100 kHz can also bring a strong signal. That is why the chopping frequency must not be a multiple of the noise frequency.

Amplifier

Despite the high sensitivity of the lock-in amplifier it appeared that an additional preamplifier can increase the signal-to-noise ratio and make the system more stable to environmental influences. The explanation is that it is advantageous to amplify the small currents in the detector spatially very close to the antenna itself. Then afterwards the amplified voltage is transmitted in the longer cable when the additional noise is present. So less environmental noise can be collected because the antenna effect of the cable is minimized.

3.5 Mechanical Components

The most important mechanical device in a time-domain spectroscopy system is the optical delay unit. Like in a Fourier spectrometer it is necessary to scan a signal as a function of delay. Here it is realized using a retro reflector on a moveable stage. A more elegant method would be to use two fs lasers with a slightly detuned repetition rate. Then one laser produces the THz pulses while the other one samples it. This technique is called asynchronous optical sampling (ASOPS) [23]. But here, because of cost and rigidity, a mechanical delay unit producing a time difference by adding a path length was applied.

Linear Stages

The used linear stage is a stepper motor driven device manufactured by Physik Instrumente (PI) in Karlsruhe, Germany. It has the advantage of a high mechanical stability and a good positioning accuracy (bidirectional reproducibility and origin better 1 µm, step size 0.1 µm). So it is possible to measure small changes of time differences which will be of importance later on for timing critical measurements. The maximum scanning range was, depending on the particular model, between 10 cm and 30 cm. This is much more than the typically scanned value of 2 cm, but important for the first alignment of the system and for thick samples. The velocity is limited to a maximum of 20 mm/sec which has to be considered also for the data acquisition time.