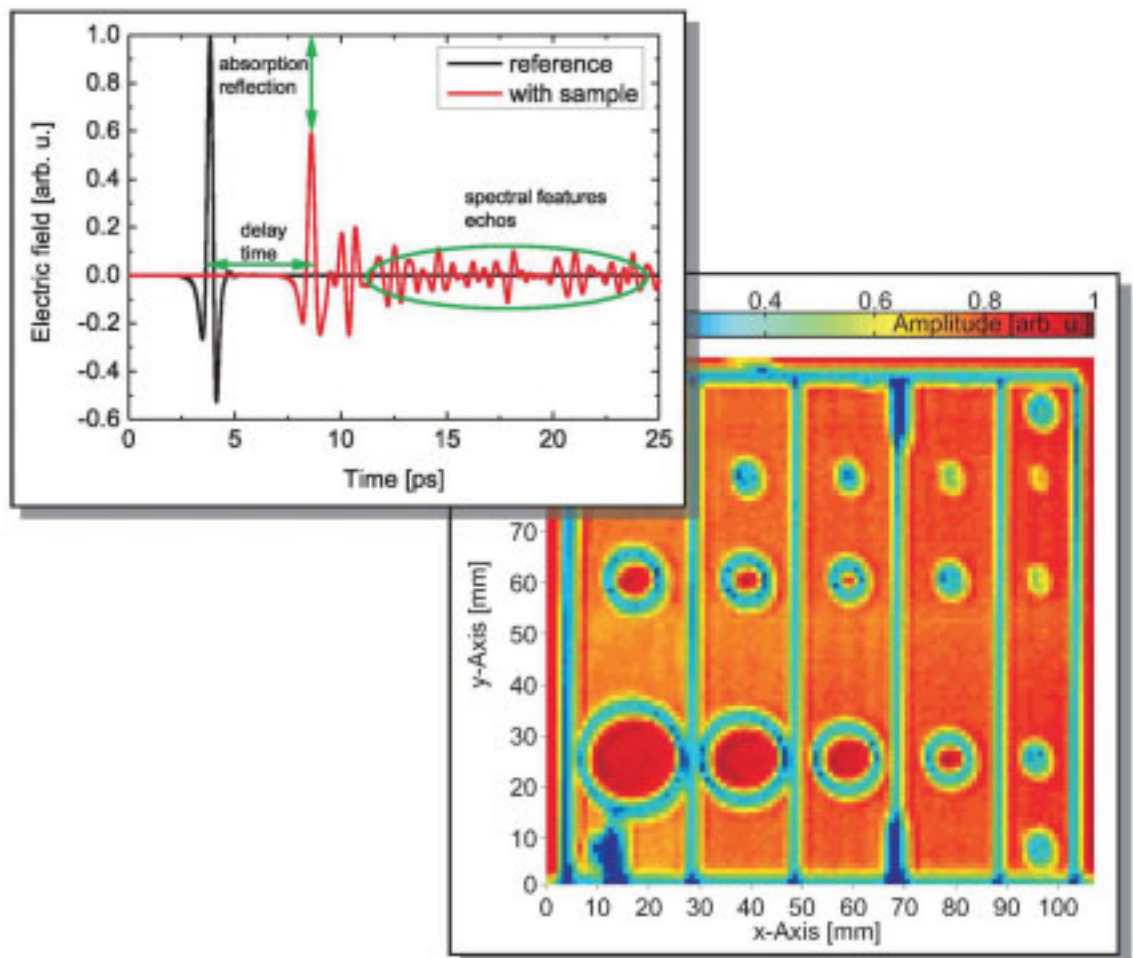


Michael Theuer

Terahertz Time-Domain Spectroscopy Systems for Fundamental and Industrial Applications



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Terahertz Time-Domain Spectroscopy Systems for Fundamental and Industrial Applications

Dissertation

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1 Introduction

The terahertz (THz) band between 100 GHz and 10 THz is particularly interesting for a wide range of applications since it unifies the properties and advantages of the adjoining spectral ranges, the infrared and the millimeter waves. But unfortunately in this spectral range the optical as well as electronic systems can not offer a sufficient performance in terms of power and sensitivity. So in both approaches, new sources and detectors are developed to expand the accessible electromagnetic spectrum into the THz range.

In this thesis new developments in the field of optical THz systems for THz time-domain spectroscopy (TDS) are investigated using femtosecond pulses for the coherent generation and detection. Starting from previous experiments, mostly carried out by C. Weiß in the group of Prof. R. Beigang at the University of Kaiserslautern, new schemes for THz emitters are elaborated. The main aim is to obtain a THz source with high output power in the frequency range between 100 GHz and 4 THz for femtosecond pumped systems. In a cooperation with the RIKEN Institute in Japan, in particular the group of Prof. K. Kawase and C. Otani, an enhancement cavity for the pump radiation is developed. This is an actively stabilized synchronously pumped ring resonator which recycles the unused pump radiation. As THz emitter a lithium niobate crystal in Cherenkov-type geometry is applied. Lithium niobate is particularly well suited as a THz emitter because of its high nonlinearity, high transparency in the near infrared and the well developed poling technique. To guarantee an efficient broadband surface emitting scheme also for longer crystals an attached silicon prism is used as THz output coupler. The enhancement cavity is characterized in terms of properties for the near infrared pump pulses as well as for the emitted THz radiation. Using coherent and incoherent detection the output is compared to other available emitters.

So far most of all optical THz systems are based on Ti:sapphire lasers with wavelengths around 800 nm. In order to apply readily available femtosecond fiber lasers at 1.5 μm wavelength, used e. g. in the telecommunication field, new materials for THz emitters have to be investigated. Here results of various emitters based on different semiconductors are presented. Wavelength dependent measurements up to 2 μm are possible using a CTA-OPO pumped by a tuneable Ti:sapphire laser. The theory of photoconductive surface emission is extended with respect to phase-coupled Hertzian dipoles. The experimental results are verified by numerical simulations.

For the application of THz radiation different TDS systems are realized. With respect to the experimental constraints the devices' layout has to be adapted. Limiting factors like laser power, pump wavelength, flexibility, space consumption and imaging optics are addressed. Portable systems and fiber based systems are constructed. A special emphasis is placed on a THz imaging system, which is used in feasibility studies for

industrial customers. This applied research and development takes place at the Fraunhofer Institute for Physical Measurement Techniques (IPM). Its recently established Dept. of THz Measurement and Systems is located at the Dept. of Physics at the University of Kaiserslautern.

Results of industrial relevant measurements are presented. The recorded THz electric field is typically evaluated in terms of amplitude and phase. This reduces the large amount of recorded data and shows a THz image underlining different properties of the device under test. Potential applications are in the field of non-destructive testing and final inspection (geometry, thickness, coatings, interfaces). Typical examples are layer thickness measurements and entirety checks in the production and packaging process. The detection limit in the time domain is demonstrated for thin layers and calibrated samples.

THz spectroscopy also can give the “spectral fingerprint”, an important decision criterion for the sample to be identified (content, substance distribution). Lactose is used as a substitute reference under laboratory conditions. Also real world explosives and poisons are measured. The advantages of THz time-domain spectroscopy compared to cw or passive THz systems are discussed.

THz-TDS can detect signals in reflection as well as in transmission. The used geometry depends on the sample and the experimental restrictions like metallic surfaces or absorbing substances. Therefore, the mentioned inspections are carried out either in direct transmission or in reflection into different angles. This is important for stand-off detection e.g. in safety and security applications. The aim here is to identify hazardous substances at the body of a person or within packages. In principle THz-TDS can spectroscopically identify substances. But real world applications are hampered by system and sample properties. The strongest limitation is given by water vapor absorption in ambient air. The accessible spectral bands are experimentally demonstrated and confirmed by simulations for different parameters like propagation length and humidity. Also the sample properties like surface roughness, interfaces and reflectivity are taken into account. A balanced discussion of the possibilities and limitations for THz stand-off detection is given.

The results of this thesis mark steps towards the acceptance of THz radiation in industrial applications: The introduction of an enhancement cavity together with a better understanding of coherent THz emitters gives rise to THz sources with higher output power. The progress on the components’ side allows the THz systems to be used in a real world environment. Finally, the shown imaging and spectroscopic results underline the possibilities of THz-TDS as a valuable technique in the field of non-destructive testing and stand-off detection.

Einleitung

Der Terahertz (THz) Spektralbereich zwischen 100 GHz und 10 THz ist für eine große Anzahl von Anwendungen interessant, da er die Eigenschaften und Vorteile angrenzender Spektralbereiche, nämlich der Infrarot-Strahlung und der Millimeter-Wellen, vereinigt. Jedoch zeigen gerade in diesem Spektralbereich sowohl die optischen als auch elektronischen Systeme unglücklicherweise keine ausreichende Leistungsfähigkeit hinsichtlich Ausgangsleistung und Empfindlichkeit. Von beiden Ansätzen her werden neue Quellen und Detektoren entwickelt, um das zugängliche elektromagnetische Spektrum um den THz Bereich zu erweitern.

In dieser Arbeit werden neue Entwicklungen im Gebiet der optischen THz Systeme untersucht. Dabei wird das Prinzip der Zeitbereichs-Spektroskopie, bei der Femtosekundenpulse zur kohärenten Erzeugung und zum Nachweis verwendet werden, eingesetzt. Aufbauend auf die vorausgehenden Experimente, die im Wesentlichen von C. Weiß in der Arbeitsgruppe von Prof. R. Beigang an der Technischen Universität Kaiserslautern durchgeführt wurden, werden neue Ansätze für THz Quellen ausgearbeitet. Das Hauptziel ist es, eine THz Quelle mit hoher Ausgangsleistung im Frequenzbereich zwischen 100 GHz und 4 THz für durch Femtosekunden-gepumpte Systeme zu erhalten.

In einer Kooperation mit der Arbeitsgruppe von Prof. K. Kawase und C. Otani des RIKEN Instituts in Japan wurde eine Überhöhungs-Kavität für die Pumpstrahlung entwickelt. Diese besteht aus einem aktiv stabilisierten, synchron gepumpten Ringresonator, der die ungenutzte Pumpstrahlung wieder verwertet. Als THz Sender wird ein Lithium Niobat Kristall in Cherenkov-Geometrie verwendet. Um einen effizienten, breitbandig emittierenden Sender auch für lange Kristalle zu erhalten, wurde ein Silizium Prisma als THz Auskoppler verwendet. Somit wird die THz Strahlung direkt unter der Oberfläche erzeugt und dadurch die Materialabsorption deutlich vermindert. Die Überhöhungs-Kavität wird auf ihre Spezifikationen hin untersucht, sowohl im Hinblick auf die Eigenschaften für die Pulse im nahen Infrarot als auch für die ausgesandte THz Strahlung. Diese wird mittels kohärentem und inkohärentem Nachweis detektiert und mit anderen verfügbaren Emittlern verglichen.

Bisher basierten die meisten THz Systeme auf einem Titan:Saphir Laser bei einer Wellenlänge von 800 nm. Um jedoch fertig verfügbare Femtosekunden-Faserlaser, wie sie bereits im Telekommunikationsbereich bei 1.5 μm Wellenlänge eingesetzt werden, verwenden zu können, müssen neue Materialien für THz Sender untersucht werden. Ergebnisse von Sendern aus verschiedenen Halbleitern werden präsentiert. Messungen bei variabler Wellenlänge bis zu 2 μm sind durch den Einsatz von durchstimbaren Titan:Saphir Lasern und eines CTA-OPOs möglich. Die Theorie für photoleitende Oberflächenemission wird um den Aspekt der phasenstarr gekoppelten Dipole erweitert. Die experimentellen Ergeb-

nisse werden durch numerische Simulationen bestätigt.

Unterschiedliche THz Systeme werden zur Anwendung der THz Strahlung entwickelt. Unter den gegebenen Randbedingungen muss der Aufbau jeweils angepasst werden. Die begrenzenden Faktoren wie etwa Laserleistung, Pumpwellenlänge, Flexibilität, Platzbedarf und bildgebende Optiken werden ausgeführt. Tragbare und faserbasierte Systeme werden konzipiert. Ein Schwerpunkt liegt auf einem abbildenden System, welches in Machbarkeitsstudien für industrielle Kunden zum Einsatz kommt. Diese anwendungsnahe Forschung und Entwicklung findet am Fraunhofer-Institut für Physikalische Messtechnik (IPM) statt. Die kürzlich eingerichtete Abteilung THz Messtechnik und Systeme befindet sich im Fachbereich Physik an der Technischen Universität Kaiserslautern.

Ergebnisse industriell relevanter Messungen werden gezeigt. Die aufgenommen THz elektrischen Felder werden üblicherweise auf Amplitude und Phase hin ausgewertet. Dies reduziert die große Datenmenge und ermöglicht ein THz Bild, welches verschiedene Eigenschaften des Probekörpers hervorhebt. Potentielle Anwendungen liegen im Gebiet der zerstörungsfreien Materialprüfung und Endkontrolle (Ausmaße, Dicke, Beschichtungen, Grenzflächen). Typische Beispiele sind Schichtdickenmessungen und Vollzähligkeitsprüfungen im Produktions- und Verpackungsprozess. Die Nachweisschwelle im Zeitbereich wird für dünne Schichten anhand kalibrierter Probenkörper bestimmt.

Die THz Spektroskopie kann auch den "spektralen Fingerabdruck" liefern, ein wichtiges Entscheidungskriterium für die vermessene Probe (Inhalt, Wirkstoffverteilung). Milchkucker wird dabei als Austauschstoff unter Laborbedingungen verwendet. Aber auch echte Sprengstoffe und Gifte werden gemessen. Die Vorteile der Zeitbereichs-Spektroskopie im Vergleich zu Dauerstrich oder passiven THz Systemen werden diskutiert.

Die Signale der THz Zeitbereichs-Spektroskopie können sowohl in Reflexion als auch in Transmission detektiert werden. Die verwendete Geometrie hängt von der Probe und den experimentellen Randbedingungen wie etwa metallischen Oberflächen oder absorbierenden Substanzen ab. Deshalb werden die Untersuchungen entweder in direkter Transmission oder Reflexion in unterschiedliche Winkel durchgeführt. Dies ist wichtig für die Ferndetektion, wie sie z. B. in Sicherheitsanwendungen erforderlich ist. Das Ziel dabei ist es, gefährliche Stoffe am Körper einer Person oder in Behältnissen zu erkennen. Grundsätzlich kann die THz Zeitbereichs-Spektroskopie Stoffe durch spektrale Auswertung nachweisen. Aber echte Anwendungen werden durch die System- und Probeneigenschaften beeinträchtigt. Die stärkste Begrenzung stellt die Absorption in Wasserdampf der Umgebungsluft dar. Die zugänglichen spektralen Bänder werden experimentell bestimmt und durch Simulationen mittels Parametern wie Propagationslänge und Luftfeuchte bestätigt. Auch die Probeneigenschaften wie etwa Oberflächenrauigkeit, Grenzflächen und Reflektivitäten werden berücksichtigt. Eine ausgewogene Diskussion der Möglichkeiten und Grenzen der THz Ferndetektion wird gegeben.

Die Ergebnisse dieser Arbeit markieren Schritte in Richtung der Akzeptanz der THz Strahlung in industriellen Anwendungen. Die Verwendung einer Überhöhungs-Kavität zusammen mit einem weitergehenden Verständnis von kohärenten THz Sendern führt zu THz Quellen mit höherer Ausgangsleistung. Die Entwicklung auf der Seite der Komponenten ermöglicht den Einsatz von THz Systemen in realistischen Umgebungen. Schließlich

unterstreichen die abbildenden und spektroskopischen Ergebnisse die Einsatzmöglichkeiten der THz Zeitbereichs-Spektroskopie als eine nützliche Technik im Bereich der zerstörungsfreien Materialprüfung und Ferndetektion.

2 Fundamentals

Since years most of the electromagnetic spectrum is accessible due to advanced optics and sophisticated electronic sources. For the infrared (IR) and the visible range strong lasers or parametric sources are available since the invention of the laser [1, 2].

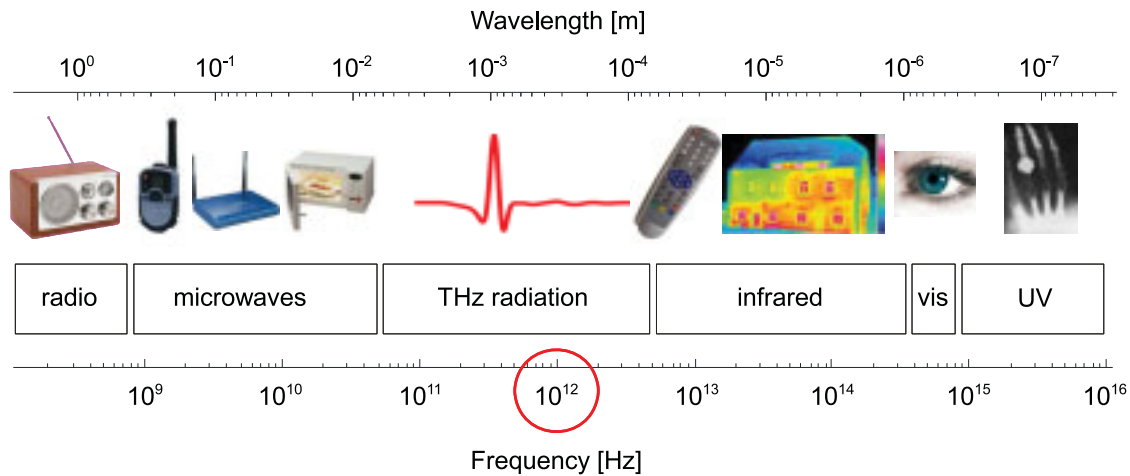


Figure 2.1: Electromagnetic spectrum in units of wavelength and frequency

But the optically accessible wavelength range has been limited towards the long wavelengths, except of some frequency-fixed gas lasers in the far infrared (FIR). On the other side of the electromagnetic spectrum the electronic sources are quite elaborated. But for the frequency range above 100 GHz the output power is rather low. Altogether there was a gap in the accessible electromagnetic spectrum, the so-called terahertz gap in the frequency range between 100 GHz and 10 THz. While both approaches, the photonic and the electronic, expand towards the THz range, for fundamental research at least for linear investigations, the available sensitivity is quite sufficient. For university purposes where cost, energy consumption, size and 24/7-reliability play a minor role compared to industrial requirements, a standard terahertz time-domain spectroscopy (THz-TDS) system is a very good device to address the frequency range between 100 GHz and 4 THz.

However, for real world industrial applications current THz systems have to be improved in terms of power, the detectors in terms of sensitivity and the overall system have to become smaller, cheaper and more reliable. From that point of view also big efforts in fundamental research have to be made to bring the devices in an appropriate condition that a company can sell systems at the free market. So the aim here is to show a

considerable progress towards a running prototype or demonstrator which is state-of-the-art and applicable in industrial processes. So the THz gap can be filled also with applications. It would be very strange if this spectral range would be the only one which is not used for applications.

The within this thesis described research topics use ultrashort femtosecond (fs, 10^{-15} s) pulses in the near infrared (NIR) to generate and detect THz pulses coherently. Because of the substantial progress in the field of nonlinear optics, pulsed fs lasers are accessible. The high peak power, good beam profile and low phase fluctuations at a high repetition rate are the main reasons for the widespread use of ultrashort pulsed lasers. Fs lasers can be operated not only under laboratory conditions but already in industrial turn-key systems in a rough environment.

2.1 Pulse Propagation

Electromagnetic waves are completely characterized by the space and time-dependent electric field $E(x, y, z, t)$. Its sources and temporal evolution are described by the linear Maxwell equations. Therefore the solutions can be applied to all wavelength ranges from the ultraviolet to the far infrared. The physics in this thesis is closely related to the NIR and the THz spectral range. So all notations introduced in the chapter remain valid for both frequency ranges despite showing a large difference in energy per photon by a factor of about 200.

A description of the electric field $E(x, y, z, t) = E(t)$ in the time or frequency domain is equivalent and can be translated using a Fourier transformation [3]

$$E(\omega) = \mathcal{F}\{E(t)\} = \int_{-\infty}^{\infty} E(t) e^{-i\omega t} dt \quad (2.1)$$

For convenience reasons also a complex representation of the electric field is common

$$E(t) = \mathcal{E}(t) e^{i\Phi(t)} \quad (2.2)$$

with an envelope of the electric field $\mathcal{E}(t)$ and an oscillating phase $\Phi(t)$ in the time domain. The phase $\Phi(t)$ can be expressed as

$$\Phi(t) = \varphi_0 + \varphi(t) + \omega_\ell t \quad (2.3)$$

This segmentation makes sense because when it comes to wave packets or pulses in most cases only a slowly varying phase $\varphi(t)$ is superimposed to a high frequency oscillating term ω_ℓ (mean or carrier frequency). Then the instantaneous frequency $\omega(t)$ of the pulse is

$$\omega(t) = \frac{d\Phi(t)}{dt} = \omega_\ell + \frac{d\varphi(t)}{dt} \quad (2.4)$$

Therefore a discussion of the slowly varying phase $\varphi(t)$ is sufficient to describe phenomena like chirp or frequency-shifts occurring due to self-phase modulation (SPM). The constant phase φ_0 is the carrier envelope offset [4] and of no concerns for our kind of experiments. Please note that the concept of envelope and carrier frequency is only valid for pulses which have a bandwidth which is small against the carrier frequency. That may not be the case for ultrashort THz pulses where the bandwidth often exceeds the carrier frequency.

Starting again with the Maxwell equations up to now the spatial coordinates and the vector character were neglected. The Maxwell equations in Cartesian coordinates give for the electric field $\mathbf{E}(x,y,z,t)$:

$$\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \mathbf{E}(x,y,z,t) = \mu_0 \frac{\partial^2}{\partial t^2} \mathbf{P}(x,y,z,t) \quad (2.5)$$

with μ_0 the magnetic permeability of free space and $\mathbf{P}$ the polarization of the medium. This source term $\mathbf{P}$ is the response of the medium and can be decomposed into two parts

$$\mathbf{P}(t) = \mathbf{P}_L(t) + \mathbf{P}_{NL}(t) \quad (2.6)$$

where $\mathbf{P}_L(t)$ varies linearly with the electric field while $\mathbf{P}_{NL}(t)$ shows a nonlinear behavior. The first term is responsible for linear losses and gain, diffraction as well as dispersion, while the latter one stands for e.g. sum and difference frequency generation. The nonlinear source term $\mathbf{P}_{NL}$ will be focussed especially in chapter 4.1 as the reason for THz emission. Restricting eq. 2.5 further to linear polarization and propagation in z-direction brings the reduced wave equation

$$\left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) E(z,t) = \mu_0 \frac{\partial^2}{\partial t^2} P_L(z,t) \quad (2.7)$$

A linear response of the medium is related to the dielectric susceptibility $\chi(\omega)$ in the frequency domain by

$$P_L(z,\omega) = \epsilon_0 \chi(\omega) E(z,\omega) \quad (2.8)$$

This together with a Fourier transformed eq. 2.7 gives a differential equation for the electric field in the frequency domain

$$\left(\frac{\partial^2}{\partial z^2} + \omega^2 \epsilon(\omega) \mu_0\right) E(z, \omega) = 0 \quad (2.9)$$

where $\epsilon(\omega)$ is the dielectric constant defined by $\epsilon(\omega) = [1 + \chi(\omega)] \epsilon_0$. A solution of eq. 2.9 is

$$E(z, \omega) = E(0, \omega) e^{-ik(\omega)z} \quad (2.10)$$

with $k(\omega)$ the complex wave vector defined by the refractive index $n(\omega)$

$$k^2(\omega) = \omega^2 \epsilon(\omega) \mu_0 = \frac{\omega^2}{c^2} n^2(\omega) \quad (2.11)$$

Comparable to the decomposition in eq. 2.3 also here $k(\omega)$ is expanded around the carrier frequency ω_ℓ

$$k(\omega) = k(\omega_\ell) + \left.\frac{dk}{d\omega}\right|_{\omega_\ell} (\omega - \omega_\ell) + \frac{1}{2} \left.\frac{d^2k}{d\omega^2}\right|_{\omega_\ell} (\omega - \omega_\ell)^2 + \dots \quad (2.12)$$

The inverse first derivation of the k-vector at the center frequency is also known as the group velocity v_g . It describes the propagation velocity of the envelope of the wave packet

$$v_g = \left(\left.\frac{dk}{d\omega}\right|_{\omega_\ell}\right)^{-1} \quad (2.13)$$

The second derivation of k is called the group velocity dispersion (GVD)

$$k''_\ell = \left.\frac{d^2k}{d\omega^2}\right|_{\omega_\ell} = -\frac{1}{v_g^2} \left.\frac{dv_g}{d\omega}\right|_{\omega_\ell} \quad (2.14)$$

This term is responsible for the broadening of a pulse. Typical optical components made of glasses show a positive GVD in the visible and near infrared range. The propagation of an initially bandwidth limited pulse in a dispersive medium extends the pulse duration. Different frequencies travel with unequal phase velocities so that a linear spectral distribution called chirp under the pulse envelope is present. This effect may be compensated by angular dispersion in prisms or grating.

For transmission of pulses in fibers, a special focus has to be set on dispersion. The first because even in low dispersive media the dispersion can add up to large values because the distances are much longer than the extents of lenses or crystals. Even if the linear dispersion is compensated, the third order dispersion (TOD) causes pulse broadening which limits the quality of the transmission. Besides the effects of dispersion, also other

nonlinear effects in fibers have to be accounted for.

If the wave vector $k(\omega)$ contains also an imaginary part, the linear losses α are also accounted for

$$k(\omega) = k_0 + \Delta k(\omega) + i \frac{\alpha(\omega)}{2} \quad (2.15)$$

and the initial electric field $E(\omega, 0)$ will suffer losses and phase changes for a given propagation length z .

2.2 Nonlinear Effects

In cases of strong electric fields $E(t)$ which are provided by lasers or focused beams the external electric fields come into the same order of magnitude as the inherent electric fields in the material. Then also nonlinear contributions of the polarization have to be taken into account in eq. 2.5. This is due to the response of the electrons (here assumed to be instantaneous) and their nonlinear potential curve for high energies.

$$P_{\text{NL}}(t) = \epsilon_0 \chi^{(2)} E(t) E^*(t) + \epsilon_0 \chi^{(3)} E(t) E^*(t) E(t) + \dots \quad (2.16)$$

The first approximation to quantify the material parameter of the nonlinear response is $\chi^{(2)}$. This term is sufficient for physical phenomena like second harmonic generation (SHG) and difference frequency generation (DFG).

$$P_2(t) = \epsilon_0 \chi^{(2)} E(t) E^*(t) \quad (2.17)$$

The simple ansatz $E(t) = E_0 e^{i\omega t}$ leads to

$$P_2(t) = \frac{\epsilon_0 \chi^{(2)} E_0^2}{2} + \epsilon_0 \chi^{(2)} E_0^2 \cos(2\omega t) \quad (2.18)$$

It is obvious from expression (2.18) that a field with twice the frequency as well as a constant DC field is emitted. The latter one is called optical rectification (OR).

Assume that the incident electric field $E(t)$ consists of two superimposed contributions $E(t) = E_1(t) + E_2(t)$ at different frequencies. Then these two waves also interact according to eq. 2.17 in a nonlinear process. The output is a mixture of the sum $(\omega_1 + \omega_2)$ and the difference $(\omega_1 - \omega_2)$ of the participating frequencies (and of course both transmitted fundamental frequencies). Which particular contribution is stronger depends on the phase matching condition because the process is described by a set of coupled differential equations.

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

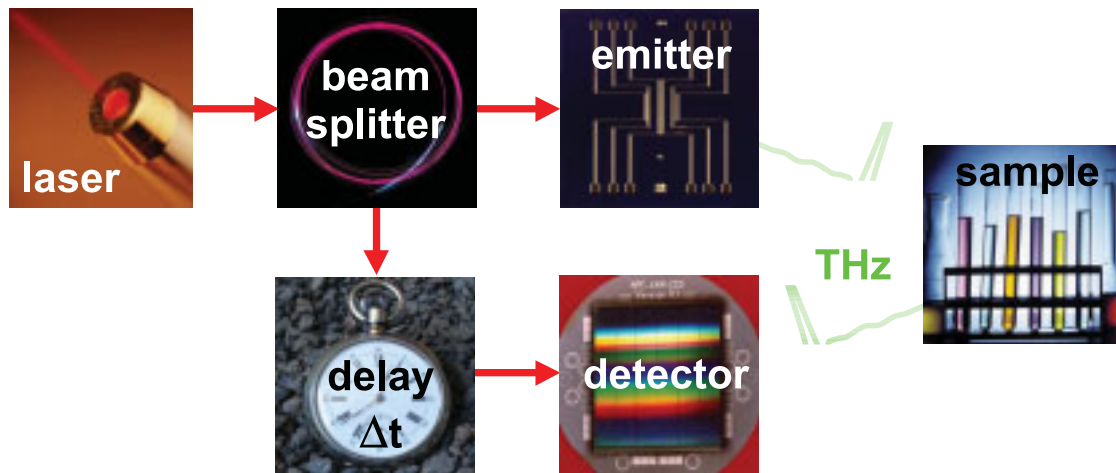


Figure 3.1: Principle of the THz time-domain spectroscopy system (THz-TDS). Major components: laser, emitter, detector and a delay producing device

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