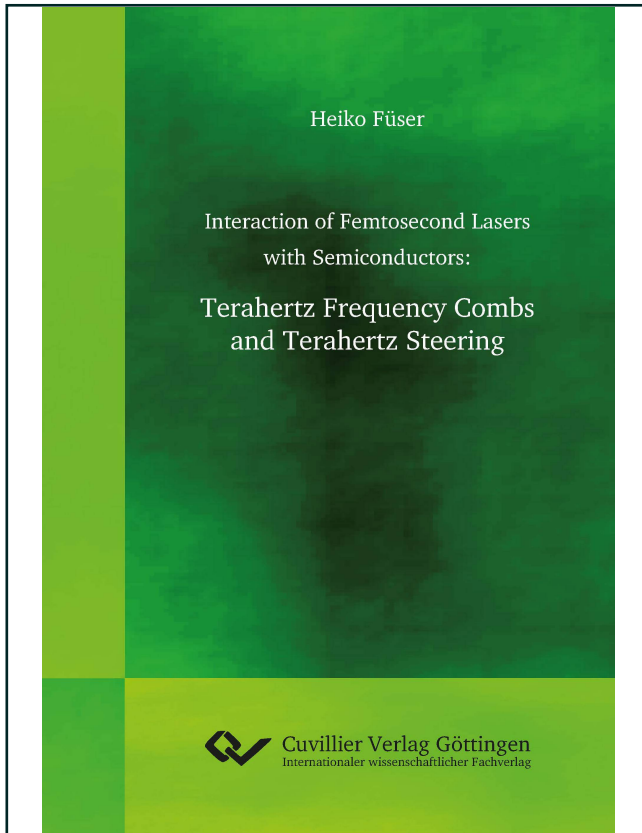




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**Interaction of Femtosecond Lasers with Semiconductors**  
*Terahertz Frequency Combs and Terahertz Steering*



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# 1 Introduction

## 1.1 Motivation

Research and development in the field of Terahertz (THz) science and technology have intensified significantly over the past 30 years [1–7]. The ongoing interest [1], motivated by both academic as well as economic means, reflects the huge technological progress in a specific part of the electromagnetic spectrum which has been difficult to access for many decades. Due to the lack of efficient generation and detection mechanisms of electromagnetic signals [2], the corresponding spectral region has long been referred to as THz gap. The exact definition of the boundary frequencies of this gap greatly varied, depending on the specific problem of interest. Typically frequencies from 100 GHz up to 30 THz have been considered (i.e. wavelengths between 3 mm and 10  $\mu\text{m}$ , respectively) [3]. Hereby the lower boundary was roughly marked by the frequency limit of typical all-electronic devices such as amplifiers and oscillators. The upper value of the THz frequency range has been determined from the optical side of the electromagnetic spectrum, given by the operational frequencies of typical laser sources such as the  $\text{CO}_2$  laser.

For a long time, the THz frequency region was only of interest for a scientific niche of mostly astronomy-related spectroscopy [4]. However, over time this part of the frequency spectrum became a matter of increasing interest for other fields, in particular for fundamental physics. Many interaction phenomena between radiation and matter are found in the far-infrared region, covering a wide range of elementary excitations such as phonons, plasmons, excitons and various transition effects [5–7]. With this development, the necessity for appropriate tools to be utilized in the corresponding investigations arouse. Consequently, the research on THz sources and detectors intensified. Various THz generation mechanisms have been established, ranging from electronic-based devices like solid-state oscillators [8] over free-electron oscillators like gyrotrons [9, 10] to THz lasers, i.e. gas lasers [11, 12], free electron lasers [13, 14] or quantum-cascade lasers (QCL) [15, 16], and other sources [17]. Moreover, broadband spectroscopic systems were realized, utilizing short opti-



cal laser pulses [18, 19]. In addition to the radiative approach driven by spectroscopic needs, also the progress in electronics has been steadily narrowing the THz gap. Here, especially the continually increasing demand of bandwidth of modern communication technology [20–23] acted as the main driver for the ongoing development of THz technologies. Wireless data transmission at high gigabit rates [24] has already successfully been employed, e.g. for the television broadcasting of events of major interest like the 2008 Summer Olympic Games in Beijing [25]. These technological breakthroughs are based on the properties of today's state-of-the-art integrated circuits, whose operational frequencies already reach far into the THz region. For instance, devices like voltage controlled oscillators operating at several 100 GHz [26] or integrated diode circuits and THz mixers for frequencies up to 2.5 THz [27] allow for the realization of photonic circuits. In combination with the novel THz sources available, e.g. QCLs, envisaged electro-photonic designs promise to bridge the THz gap [28].

The advent of THz generation and detection techniques unlocked the door for a multitude of applications. In various fields such as medical diagnosis, quality control, chemical and biological sensing, imaging techniques for security applications and other purposes, THz techniques have proven to be a valuable tool [29–33]. The increasing economic interest in this particular frequency band indispensably calls for metrological developments to characterize devices, judge the reliability of measurement methods, precisely compare different technical approaches or define common standards. For about a decade, national metrology institutes (NMIs) all over the world intensified their work towards THz metrology [34] and, by this, accompanied the rise of this new technology. Large NMIs performing research in the field of THz metrology are, e.g., the National Institute of Standards and Technology (NIST) in the USA, the National Physical Laboratory (NPL) in the UK, the National Metrology Institute of Japan (NMIJ) and the National Institute of Information and Communications Technology (NICT) in Japan, the National Institute of Metrology (NIM) in China, the Korea Research Institute of Standards and Science (KRISS) in Korea and the Physikalisch-Technische Bundesanstalt (PTB) in Germany. To cover the wide range of THz applications, metrological research projects have been established. These activities comprise spectrometry applications [35–37], THz radiometry [38–43], characterization of high-frequency electronics [44–49] [C1], the calibration of THz sources [50–52], high-precision frequency measurement techniques [53–56] and more. Despite these projects, THz metrology constitutes a very recent branch in the portfolio of NMIs, thus requiring intensified ongoing research. A vital task for the future concerns an international coordination of these research projects and a thorough comparison of the various ap-



proaches realized by the different NMIs. Envisaged comparisons cover, e.g., the characterization of high-frequency electronic devices [C5] or the development of traceable THz detectors [57].

This description outlines the framework in which the work presented in the following chapters is placed. This thesis is divided into two main parts, discussing different aspects of the ongoing effort to close the THz gap. First, precision measurements of free-space, continuous-wave (cw) electromagnetic signals in the THz frequency domain are presented. An accurate measurement of different parameters of the radiation is realized by utilizing the concept of THz frequency combs [53]. Incorporating a data processing scheme to correct the measurement data for the noise influences of the laser system, high-precision frequency determination with an accuracy of the measurement system as high as  $(9 \pm 3) \cdot 10^{-14}$  is demonstrated using a 100 GHz source [A3]. This presents an improvement of two orders of magnitude as compared to previously reported results [54, 55, 58]. THz communication, imaging and sensing applications are not only based on frequency, but also on amplitude and phase information of electromagnetic signals. Thus, employing the same optoelectronic detection scheme as utilized for the frequency measurements, a spatially resolved analysis of these parameters is demonstrated. Here, standard deviations of the mean of 0.1 % and  $0.2^\circ$  are reached for the measurement of the relative amplitude and phase, respectively. With this, the full characterization of the emission properties of cw-THz sources is demonstrated [A1]. By utilizing the emission of a CO<sub>2</sub>-laser to access the far-end of the THz gap, the frequency limits of the detection process are analyzed. For a precise determination of the different parameters of electromagnetic radiation, a detailed understanding of the interaction processes of the electromagnetic signals with the detection system is necessary. To account for this, an evaluation and comparison of two techniques for the detection process is presented, namely photoconductive and electro-optic detection. By this, two entirely different light-matter interaction processes (with the polarization depending in first respectively second order on the optical field strength) are analyzed towards their usability for the aforementioned metrological tasks.

The second topic of this thesis addresses a THz generation mechanism. For many of the realized and envisaged applications operating in the former THz gap, specific properties of the THz emission are mandatory. Among these properties, the need for spatial control of the emitted fields has become more and more important. For instance, communications at high frequencies requires the realization of different propagation channels [48]. Furthermore, spatially resolved spectroscopy and imaging applications are based on a relative movement between the THz field and



the device under test [59]. One possible approach to accomplish such demands is given by a steerable THz emitter. In this thesis such an emitter is presented, based on an all-optical approach for the THz generation process as well as for the control of the emitted fields [A2]. Utilizing the optically induced emission of THz radiation from a gallium arsenide (GaAs) semiconductor material, adjustable THz steering angles reaching from below  $2^\circ$  to more than  $8^\circ$  are realized. The presented method allows for a fast, reliable and non-mechanical beam steering. A simple model is introduced, capable to relate the steering effect to different optically excited current contributions. With the help of the model an analysis of the underlying relations and dependencies between the THz field and the optical excitation parameters is presented. This work is the basis for future developments of next-generation THz emitters.

## 1.2 Outline of the Thesis

This thesis is organized as follows. In chapter 2, the theoretical and experimental fundamentals are given, common for both topics of THz generation and THz detection by optoelectronic means. Indifferent whether the generation of pulsed THz signals or the detection of cw-THz radiation is regarded, the corresponding processes described here are based on the interaction of light with a semiconductor material. Thus, the concept of semiconductor susceptibility is introduced (section 2.1), with a focus on the aspects being relevant for this thesis. Subsequently the basic properties of the light source used throughout this work are presented (Section 2.2), a femtosecond solid-state laser utilizing a titanium-doped sapphire crystal. Finally the findings of both sections are combined (section 2.3), introducing the resulting effects and dependencies of the light-matter interaction relevant for the remainder of the thesis.

Chapter 3 represents the treatment of THz detection by use of THz frequency combs. After a brief motivation and review of the related literature (section 3.1) the high-precision frequency measurement scheme (section 3.2), the spatially resolved amplitude and phase measurements (section 3.3) and the frequency range of the detection methods (section 3.4) are discussed. In all sections, a comparison of both electro-optic and photoconductive detection is performed. Hence, the chapter closes with a conclusion and outlook regarding the advantages and disadvantages of both methods (section 3.5).

Chapter 4 introduces the concept of coherently controlled THz emitters. Again a brief motivation and overview of the corresponding literature is given (section 4.1). A schematic description outlines the concept of



the THz steering mechanism (section 4.2), which is then experimentally demonstrated and analyzed (section 4.3). A more detailed understanding of the steering effect is obtained by simulating the radiation pattern using a dipole model (section 4.4). Finally, the obtained insights and conclusions allowing for an improvement of the technique are summarized (section 4.5).

An outlook for future work is given in the last part of this thesis, chapter 5.



## 2 Theoretical and Experimental Fundamentals

In this chapter, a description of the fundamental relations necessary in the remainder of the thesis is given. First, the concept of susceptibility is introduced in section 2.1. Going from a classical description to a quantum mechanical approach, the basic expressions for the light-matter interaction utilized in the following chapters are derived. Second, the properties of the laser system used throughout the presented work are summarized in section 2.2. Here, especially the concept of optical frequency combs is introduced, as well as the noise contributions inherent to the laser process. Section 2.3 combines the hitherto outlined insights and describes the fundamental equations for the generation and detection of THz signals based on light-matter interaction.

### 2.1 Susceptibility of Semiconductors

The interaction of light with matter reveals a rich and complex field in modern physics. Atomic systems offer specific responses if excited by electromagnetic waves under appropriate conditions. Especially if the atoms are arranged in a regularly ordered scheme like crystal lattices, light may be used as a tool to induce well-defined effects. To introduce the fundamental relations of light-matter interactions necessary in the scope of this work, the following section briefly reviews the corresponding literature, e.g. [60–62], as well as recent publications for a more specific examination of the topic. Depending on the level of detail of the investigation, more or less accurate models can be used to describe the appearing interaction phenomena. Hence, first a macroscopic picture is presented to establish a general overview and introduce basic phenomena. In a second step, a more elaborated approach based on a quantum mechanical description is given, which is necessary to explain the detailed material response after excitation with an electromagnetic field of high power.





### 2.1.1 Lorentz Oscillator Model

The basic concept of susceptibility is best introduced using a highly restricted model. Regarding a material system like an atom or a solid composed of a large group of non-interacting atoms, the system consists of positively charged ions and negatively charged electrons bound to the ions. If such an electron is exposed to a time-varying, monochromatic electrical field  $\mathbf{E}'(t) = \mathbf{E}'(\omega) \exp(-i\omega t) + \text{c.c.}$  with a frequency  $\omega$  and an amplitude  $\mathbf{E}'(\omega)$  and if this field is not strong enough to break the bond between ion and electron, a relative displacement  $\mathbf{x}(t)$  of the electron from its equilibrium position is caused. Thus, a dipole moment  $\mathbf{p}(t) = e\mathbf{x}(t)$  results, with  $e$  expressing the elementary charge. Under the influence of the temporal variation of  $\mathbf{E}'(t)$  and the restraining field of the positive charge, the electron movement may be described as a damped driven oscillation [61]

$$m_0 \frac{d^2 \mathbf{x}(t)}{dt^2} = -2m_0 \gamma \frac{d\mathbf{x}(t)}{dt} - m_0 \omega_0'^2 \mathbf{x}(t) + e\mathbf{E}'(t), \quad (2.1)$$

where  $m_0$  is the electron mass,  $\omega_0'$  and  $\gamma$  are the resonance frequency and the damping constant of the oscillator, respectively. The latter corresponds to the radiative losses caused by an accelerated charge particle. This expression is solved by the ansatz  $\mathbf{x}(t) = \mathbf{x}(\omega) \exp(-i\omega t)$ , which inserted into (2.1) results in  $\mathbf{x}(t) = -e\mathbf{E}'(t) / [m_0 (\omega^2 + 2i\omega\gamma - \omega_0'^2)]$ . Hence a proportionality between the dipole moment and the electrical field is given,

$$\begin{aligned} \mathbf{p}(t) &= \frac{e^2}{m_0} \frac{1}{\omega_0'^2 - \omega^2 - i2\gamma\omega} \mathbf{E}'(t) \\ &= a_p(\omega) \mathbf{E}'(t), \end{aligned} \quad (2.2)$$

where the complex proportional constant defines the optical polarizability  $a_p(\omega)$  of the Lorentz oscillator model.

For homogeneous materials composed of many of such oscillators, it is convenient to express the proportionality between the overall dipole moment  $\mathbf{P}(t)$  and an electrical field  $\mathbf{E}(t)$  by the electric susceptibility  $\chi$ ,

$$\mathbf{P}(t) = \epsilon_0 \chi \mathbf{E}(t). \quad (2.3)$$

Here and in the following, the International System of Units (SI) is used, thus the constant  $\epsilon_0$  denoting the electric permittivity of free space is introduced in (2.3). Following the description of the oscillator model, the susceptibility is connected to  $a_p(\omega)$  via the Clausius-Mossotti Relation. The latter takes into account that the local field  $\mathbf{E}'(t)$  at the position of an atom is influenced by the surrounding polarization and, thus, differs from the

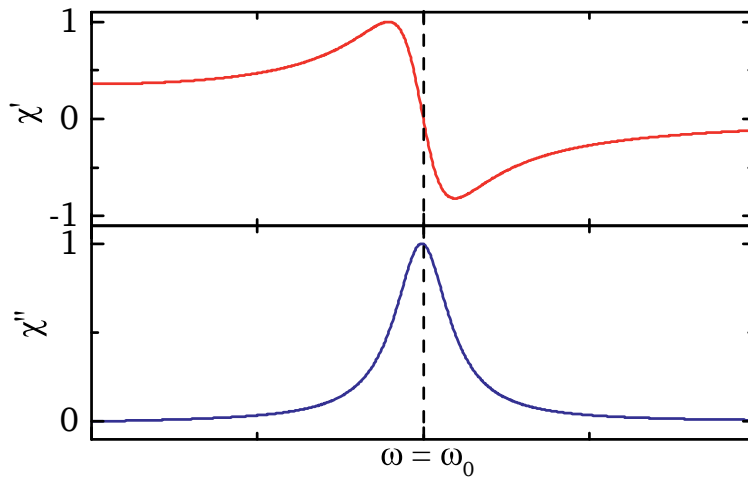


Figure 2.1 Real and imaginary part of the normalized susceptibility around the resonance frequency  $\omega_0$ , denoted as dashed line.

externally applied field  $\mathbf{E}(t)$  [61]. Together with the number of oscillators per unit volume  $N$ , it follows for the susceptibility

$$\chi(\omega) = \frac{Ne^2}{m_0} \frac{1}{\omega_0^2 - \omega^2 - i2\gamma\omega}, \quad (2.4)$$

having a slightly detuned resonance frequency  $\omega_0$  with respect to  $\omega'_0$  due to the local-field correction. Separating real and imaginary part of  $\chi = \chi'(\omega) + i\chi''(\omega)$ , it follows

$$\chi'(\omega) = \frac{Ne^2}{m_0} \frac{\omega_0^2 - \omega^2}{(\omega^2 - \omega_0^2)^2 + 4\omega^2\gamma^2} \quad (2.5a)$$

$$\chi''(\omega) = \frac{Ne^2}{m_0} \frac{2\gamma\omega}{(\omega^2 - \omega_0^2)^2 + 4\omega^2\gamma^2}. \quad (2.5b)$$

The behavior of  $\chi'$  and  $\chi''$  around the resonance frequency  $\omega_0$  is shown in figure 2.1. The course of the real part describes dispersion, whereas the imaginary part is related to absorption effects as will be discussed in the following. Using the definition of the dielectric permittivity  $\epsilon(\omega) = \epsilon_0(1 + \chi(\omega))$ , the ability of a material to transmit an electrical field is described. Due to the complex quantity  $\chi$ , the material's refractive index  $n \approx \sqrt{\epsilon(\omega)/\epsilon_0}$  is complex as well, having a real and an imaginary part  $n = n'(\omega) + in''(\omega)$  expressed by

$$n'(\omega) = \sqrt{\frac{1}{2\epsilon_0} \left( \epsilon'(\omega) + \sqrt{\epsilon'(\omega)^2 + \epsilon''(\omega)^2} \right)} \quad (2.6a)$$

$$n''(\omega) = \sqrt{\frac{1}{2\epsilon_0} \left( -\epsilon'(\omega) + \sqrt{\epsilon'(\omega)^2 + \epsilon''(\omega)^2} \right)}. \quad (2.6b)$$