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Spectroscopic Applications of Terahertz Quantum-Cascade Lasers

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Chapter 1

Introduction

Any matter in this universe is made up of particles called atoms or molecules. Research on the interaction between light and matter using spectroscopic techniques provides great flexibility to probe the matter in new ways. When electromagnetic radiation interacts with matter, it involves either absorption, emission, or dispersion of radiation by the system being studied. Absorption or emission of atomic and molecular spectra can provide detailed information about the structure and chemical properties of the system. Spectroscopy has thus produced a significant contribution to the current state of atomic and molecular physics, to chemistry, and molecular biology. It is used in material research to identify different materials due to their intrinsic spectral fingerprints. In principle, one can identify spectral fingerprints of media that rely on the transitions from the ground state to the excited energy states of the medium. The accurate knowledge of the laboratory transition frequencies of the molecules using diagnostic tools is the prerequisite for the analysis of astronomical observation. A detailed analysis of the data yields valuable information on molecular parameters and is essential for the prediction of previously unmeasured transitions, either in frequency regimes not yet explored experimentally or of intensities not in the range of even the most sensitive spectroscopic techniques.

1.1 Terahertz radiation

The terahertz (THz) range, from 1 to 10 THz (30–300 μm) of the electromagnetic spectrum lies between microwave and infrared frequency range just at the border between photonics and electronics [c.f. Fig. 1.1]. This frequency spectrum of electromagnetic waves has broad potential in a wide range of applications, from fundamental research, such as molecular spectroscopy and astronomy, to practical areas such as environmental science, biomedical and security applications. A significant reason for the increased interest in THz research comes from astronomy. Many important astronomical molecules and atoms have their fingerprint absorption and emission spectra in the THz frequency range. To date, around 200 molecular species have been detected in the interstellar medium (ISM) and circumstellar shells [1]. Nevertheless, the generation and detection of THz waves are not as mature as in other regions of the electromagnetic spectrum. This frequency range has proven to be one of the most challenging to operate because the frequency is typically too high for traditional electronics, and the photon energy is too low compared to visible and near-infrared (NIR) light. Therefore, it is difficult to control and manipulate THz radiation using existing electronics and optics. As a result, the large portion of the THz range was not mainly explored because there were no suitable emitters to send controlled THz signals or active sensors to collect and record information. Over the last two decades, intensive research and development activities in academia and industry have tried to bridge the gap between microwave and infrared spectra. New concepts, techniques, and methods have been applied for the generation and detection of THz radiation in recent years. As a consequence, compact THz sources and detectors are now being developed that are capable of generating, detecting, and manipulating THz signals. Recent innovations allowing both robust and reliable THz sources, combined with high-performance THz spectroscopy and imaging systems, have opened up remarkable new opportunities in science and technology.

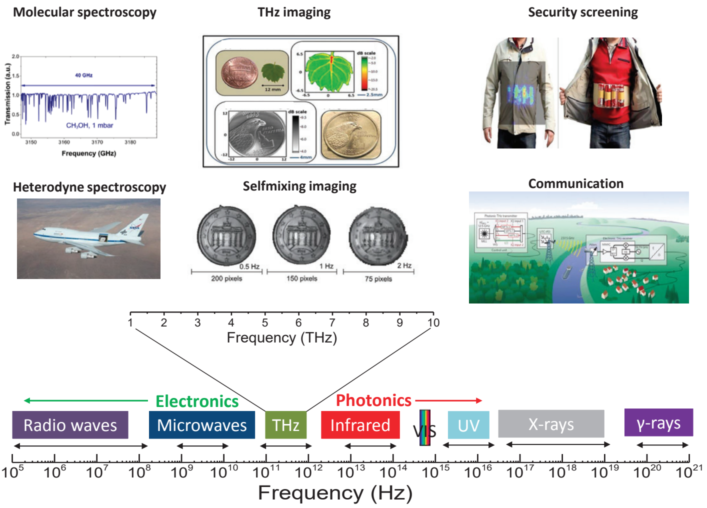

Figure 1.1: Electromagnetic spectrum and THz applications [2, 3, 4, 5, 6, 7].

1.2 Applications of THz radiation

The THz frequency spectrum is suitable for a variety of applications in different fields. Some of the very highlighted applications are discussed briefly in the following subsections.

1.2.1 Security screening

The development of techniques for the inspection of plastic explosives, chemical, and biological weapons has become essential as public safety concerns have increased considerably in recent years. THz imaging takes advantage of relatively low levels of non-ionizing THz radiation to detect hidden objects in fabrics, ceramics, and paper. It can reasonably handle security screening tasks such as checking mail packages, envelopes, and small parcels for hidden objects and threats. Figures 1.2 (a)–(e) display samples of THz images of various objects hidden under clothes, paper, plywood, and wood. Being organic compounds, most explosives have their unique spectral features in the THz band due to their rotation and collective vibrational transitions. Using THz spectroscopy, it should be possible to detect and identify unknown substances through their transmission characteristics and reflective spectra.

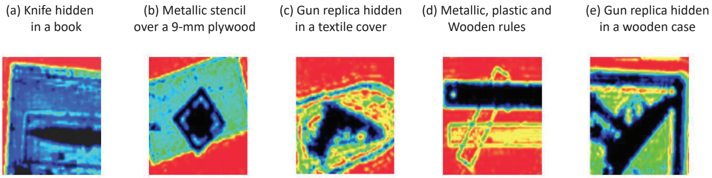

Figure 1.2: (a)–(e) Security screening applications: THz images of various hidden objects [6].

1.2.2 Biomedical research

THz waves have been used for the research of biological and medical applications. The investigation of biological features using THz radiation has both advantages and disadvantages. One of the advantages of THz radiation is the low photon energy, which is well below the ionization energies of atoms and molecules. This implies that one can study materials with THz radiation without any tissue damage unless too much power is applied. The other advantage is that biological molecules depend on hydrogen bonding, which is the most dominant bond in biological samples. For example, interactions between water and biomolecules cause hydrogen bonds to mediate and participate in a wide range of biomolecular interactions. THz radiation capable of detecting such spectral features of resonance and motion of molecules. The disadvantage of THz radiation is the high absorption in water in which biological molecules reside. Several biomedical studies have been performed in the THz frequency range. Among these studies are the determination of glucose concentration [8], monitoring DNA junction structures [9], breath gas analysis [10], water concentration determination [11], as well as many more.

1.2.3 Astrophysics and atmospheric sensing

Astronomy and astrophysics are the significant applications for continuous-wave (cw) THz spectroscopy. The THz spectral regime provides a wide range of spectral lines that are invaluable probes for star formation and planetary atmospheres [12]. It contains a plethora of spectroscopic features of atoms and molecules that are essential diagnoses of both the physical and chemical conditions of the gas and the energy sources in the astronomical environments. These spectral probes include rotational lines from simple molecules (e.g., HD, CH, OH, CO, H_2O, NH_3) and ground state fine structure lines from abundant atoms and ions (e.g. C, C^+, N^+, N^{++}, O, and O^{++}). As the wavelength of THz radiation is much larger than the typical size of the dust grains ($\sim$0.1 μm), the THz radiation is less scattered by interstellar dust as in the case for mid-infrared or optical radiation [12]. Observations in different ranges of the electromagnetic spectrum provide full information about the studied astronomical objects to the astronomers. One of the examples of astronomical observation is the Stratospheric Observatory for Infrared Astronomy (SOFIA), where the multi-frequency channels instrument GREAT (German Receiver of Astronomy at THz Frequencies) is used to acquire high-resolution spectra. The 4.7 THz heterodyne spectrometer system in the GREAT instrument has partly been developed in the German Aerospace Center, Berlin. This spectrometer system covers the frequency range around the fine structure line of the neutral atomic oxygen at 4.7448 THz [13].

1.2.4 Wireless communication

Over the past ten years, the rise of wireless devices and the rapid increase in data traffic have drastically increased the demand for spectral bandwidth, along with much faster data rates. Researchers, therefore, believe that switching to the carrier frequency above 0.1 THz could be a promising way to address such a huge demand. Numerous articles on various aspects of THz wireless communication measurements have been published in recent years. Carrier frequencies about 300 GHz are the most widely observed bands due to the availability of transmitter devices and components with sufficient output power.

To date, GaAs/InP electronics technologies are leading over 0.1 THz in all-electronic THz communication. They can cover frequency bands up to 300 GHz with a data rate of over 64 Gbit/s [14]. However, other semiconductor-based technologies have also been demonstrated for THz communication in the last few years. Si-electronics-based transceivers now easily enable 10 Gbit/s wireless links up to 260 GHz bands [15]. Communication links enabled by THz photonics technologies could play a significant role in the realization of an efficient THz wireless system. The highest data rate of about 100 Gbit/s for photonics-based transmitters has been demonstrated [16]. However, the transmission distance is limited by atmospheric attenuation as THz undergoes significant atmospheric absorption.

1.3 Organization of thesis

The main objective of this thesis is to explore spectroscopic applications of THz QCLs. The first introductory chapter provides an overview of the THz spectrum and its applications. In the second chapter, the fundamental overview of the sources available for THz radiation will be addressed. Besides, the detectors used in this dissertation and the components, including mirrors, windows, polarizers, and many others, will be discussed. The third chapter provides a basic overview of the working principle, the waveguide technique, and the temperature performance of the THz QCLs. The fundamentals of laser absorption spectroscopy, as well as its principle and nomenclature, will also be addressed here. In addition, various techniques for high-resolution spectroscopy will be briefly presented. The fourth chapter will then discuss the technical and experimental methods, including the cooling systems used for THz QCLs and the fundamental characterization techniques. Finally, the high-resolution molecular spectroscopy with THz QCLs will be discussed. Chapter 5 will present the technique and results for Doppler-free spectroscopy. The main objective of this type of spectroscopy is to study spectral features below the temperature-imposed Doppler linewidth limit. In Chapter 6, the wide-band frequency tuning of THz QCLs with a near-infrared (NIR) optical excitation will be addressed. The feasibility of this approach for molecular laser absorption spectroscopy is demonstrated in this work. Finally, Chapter 7 will demonstrate the technique for stabilizing the frequency and output power of a THz QCL. The technique exploits frequency and power variations upon near-infrared illumination of the QCL with a diode laser.

Chapter 2

THz sources, detectors, and components

This chapter provides a brief introduction to THz sources, detectors, and passive components. In section 2.1, the sources others than THz quantum cascade laser (QCL) are briefly presented. The detectors used for this thesis will be discussed in section 2.2. The materials used for passive THz components (lenses, mirrors, waveplates) will be addressed shortly in the last section.

2.1 THz sources

The THz generation process can be divided into two categories: the direct and the indirect generation process. Sources that directly generate THz waves through oscillation in electronic (electron beam or solid-state sources) or optical devices (QCLs) are known as direct processes. On the other hand, sources that generate THz radiation by photo-mixing in a non-linear medium or a medium with accelerated electrons are referred to as indirect processes. Sources that are relevant competitors of the THz QCL (frequency range, output power, and operating principle) will be briefly discussed in the following paragraphs.

2.1.1 Solid-state sources

Electronic solid-state sources such as oscillators and amplifiers are generally limited in frequency due to the transit time of carriers through semiconductor junctions, which cause the high-frequency roll-off. The output power drops off as $1/\nu^{\alpha}$ as the frequency ν increases. The values of α are between 1 and 3 [17]. Solid-state sources include resonant tunneling diodes (RTD), Gunn or transferred electron devices (TED), and transit time devices such as impact avalanche transit time (IMPATT) diodes and tunnel injection transit-time (TUNNETT) diodes. They are compact devices and can operate at room temperature. Gunn devices that are generating 0.2–50 μW power at 400–560 GHz frequency range are now feasible [18]. TUNNETT diodes with operational frequencies as high as 355 GHz with 140 μW output power have been reported [19]. In a THz frequency multiplier, the frequency of a driver source is multiplied in a nonlinear device to generate higher-order harmonic frequencies up to 2 THz. Planar Schottky diodes are commonly used in frequency multipliers, taking advantage of GaAs substrate-less technology to reduce the loss in the substrate.

2.1.2 Electron beam sources

Gyrotrons, free-electron lasers (FELs) and backward wave oscillators (BWOs) are electron beam sources capable of generating comparatively high output power, high-frequency THz radiation. All of these instruments are based on the interaction of a high-energy electron beam with a powerful magnetic field inside a resonant cavity or a waveguide, which results in an energy transfer between the electron beam and an electromagnetic wave. For gyrotrons, a power of 5 kW at 1 THz was demonstrated in pulsed mode [20] and 100 W were obtained for cw gyrotrons at frequencies ranging from 0.2 to 0.5 THz [21]. The FEL can provide broad frequency tuning, ultra-short pulses, and very high intensity in the THz region. BWOs can be operated in the THz region with moderate output power levels. They can be electrically tuned over a bandwidth of more than 50% of their operational frequencies and can generate a few mW at 1 THz [22]. The primary drawbacks of such a system are a large size, high cost, and system complexity. Complete systems require high bias voltages and usually water cooling.

2.1.3 THz gas laser

THz gas lasers exploit rotational transitions in the excited vibrational state of a low-pressure polar gas molecule. The optically-pumped THz gas lasers rely on the selective absorption of tunable infrared radiation to create a population inversion between rotational states. Fig. 2.1 displays the lasing process: first mid-IR radiation passes through an absorption cell. If the energy of the mid-IR photon matches the transition between the ground and the excited vibrational state, it becomes absorbed by the gas molecules. This process results in a population inversion between rotational states, and an inverted rotational transition causes stimulated emission at the THz frequency. The molecule remains in the excited vibrational state, which must return to the ground state before the next pump cycle.

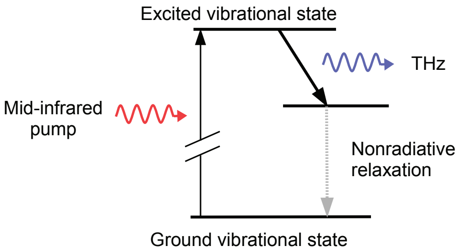

Figure 2.1: Lasing process of a optically excited THz gas laser. Adapted from [23].

For many years, CO_2 pumped gas lasers have been the major source of cw THz radiation above 0.3 THz. Typical CO_2 pumped THz gas lasers cover a frequency range of 0.15–8 THz with output power varying from several μW to several 100 mW [24]. Several groups have recently demonstrated THz radiation in an optically pumped ammonia pumping provided by a mid-IR QCL [25, 26]. Such lasers can provide a THz power level of 20–34 μW for 40–60 mW of pump power [26].

2.1.4 Difference frequency generation

Dual-wavelength mid-IR QCLs are currently the only electrically pumped semiconductor sources operating at room temperature that allows for difference-frequency generation (DFG) within 1–6 THz frequency ranges. These sources rely on the giant susceptibility ($\chi^{(2)}$) in the active region of mid-IR QCLs and a Cherenkov emission scheme [27]. The performance of THz DFG-QCLs has rapidly improved, but still, it can generate an only cw output power of 14 μW [28]. Another device for THz generation is photoconductive antennas (THz-PCAs), which have been extensively used for the generation of THz broadband pulsed and single-frequency cw signals. In cw mode, two laser beams, with their frequency difference in the THz range, combined either inside an optical fiber or overlapped adequately in space, are mixed in a photo-absorbing medium (photomixer) and generate a beat frequency signal. The output of two cw lasers is converted into cw THz radiation exactly at the difference frequency of the lasers. THz signals with the frequency linewidth as low as a few kHz can be generated by photo mixers. The frequency of the THz signal can be tuned by tuning the wavelengths of the lasers. On the other hand, the output power in conventional photomixers falls from 2 μW at 1 THz to below 0.1 μW at 3 THz [22].

2.2 THz detectors

THz detectors can be divided into two broad categories: direct photon detectors and thermal detectors. The operation of photon detectors is based on a photo effect where the radiation is absorbed within the material by interacting directly with bound or free-electrons. The direct photon detectors show a selective wavelength dependence of response per unit incident radiation power. To achieve excellent signal-to-noise performance and very fast response, a photoconductive detector requires cryogenic cooling. This is necessary to prevent the thermal generation of charge carriers. In this thesis work, Ge:Ga photoconductive detectors are commonly used for most of the measurements. On the other hand, in the case of a thermal detector, the incident radiation is absorbed to change the temperature of the corresponding material. The resultant temperature change alters the physical property to generate an electrical signal and does not rely on the photonic nature of the incident radiation. Thermal detectors are generally wavelength-independent, and the signal depends upon the radiant power. The thermal detector can be mainly divided into two categories: room-temperature and helium-cooled detectors. Golay cells, pyroelectric detectors, and microbolometers are room temperature thermal detectors. Highly sensitive detectors such as bolometers in the THz frequency range are mostly helium-cooled.

2.2.1 Ge:Ga photoconductive detectors

In general, THz photon energies are not sufficient to overcome the bandgap energy of an intrinsic semiconductor. In order to detect THz radiation via the photoconductive process, it is necessary to add impurities to the semiconductor. In such an extrinsic semiconductor, a donor or acceptor state exists close to the conduction or valence band so that a low energy photon can excite an electron out of a donor state or a defect electron (hole) out of an acceptor state. An extrinsic photoconductor detector based on Gallium doped Germanium (Ge:Ga) is used to detect THz radiation in this thesis. The detection mechanism is based on a transition from the Ga ground state to the valence band (Fig. 2.2 (a)). Such sensitive photoconductive detectors have been used for many years for low-noise photon detection for wavelengths from 60 to 120 μm [29]. These kinds of devices have high input impedance at low temperatures. The detector mostly used in this thesis has an input impedance of 150 kΩ at 4.2 K.