Chapter 1
Introduction

1.1 The Terahertz Spectral Region

The terahertz (THz) spectral region, in this book considered as the range from 0.3 to 10 THz (1 mm–30 µm), is one of the most exciting and, at the same time, most frustrating sectors of the electromagnetic spectrum. Exciting because of the wealth of knowledge that can be gained from research at these frequencies, frustrating because, until recently, techniques have lagged behind those at lower and higher frequencies. But before introducing these real difficulties there is a more trivial dilemma – what is the best name for this spectral range?

For many years, the term “far-infrared” (FIR) was used to cover all wavelengths between ~20 µm up to the shortest millimeter region. Later suggestions were “sub-millimeter” but as this, used strictly, provides a cutoff at 1 mm this was not entirely satisfactory. In 1978, Blaney suggested an accurate but rather long title “short millimeter and sub-millimeter-wavelength range” which could be abbreviated to SMSMR [1], but this did not become widely accepted. “Terahertz” is a relatively recent introduction and was initially closely associated with THz time-domain spectroscopy (THz TDS) but is now generally accepted as the convenient description for this spectral range regardless of whether it is approached from the IR or microwave direction. There is some doubt as to who first suggested the term terahertz, but it seems to have become increasingly accepted following its use by J. W. Fleming in a 1974 paper [2].

A major problem for research in this spectral region is the very high absorption of the Earth’s atmosphere, over most of the frequency range, mainly due to the vibrational–rotational levels of water vapor. This limited THz astronomy, and research in the upper atmosphere, to high altitude observatories and balloon flights in the very few “windows” available. Figure 1.1 shows the atmospheric transmission, between 0.2 to 2.2 THz, from the site of the Receiver Lab Telescope (RLT) on Cerro Sairecabur in the Andes range of northern Chile. The site is at 5525 m and appears to have some of the best THz transmission available. The spectrum was obtained in January 2005 with a Fourier-transform spectrometer.
The intensity of atmospheric absorption is remarkable, reaching a maximum of over $2 \times 10^5$ dB/km at around 8 THz [6]. To put that number in perspective, less than 1% of light at these frequencies would travel from a source to a detector over a path length of 10 cm. As an aside, one of the authors of this book failed to locate a powerful line at 9.1 THz from the first high-power THz laser [7] because there was a short open-air section in the optical path. As a further aside, when the distinguished cosmologist Sir Fred Hoyle turned to science fiction he wrote a book entitled “October the First is Too Late.” In it a rocket is launched with a payload to measure the sun’s output at a “wavelength roughly a hundred times less than the shortest radio waves,” a wavelength at which no radiation reached the earth’s
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A powerful coherent signal is observed from a source with an apparent dimension of 10 times the radius of the sun. The scientists suggest that this would produce such a non-divergent beam that they must have located an inter-galactic relay station!

To achieve negligible absorption throughout the THz region, optical paths need to be evacuated to below 100 Pa, a pressure readily achievable with rotary vacuum pumps. However, if tunable sources are available, it is possible to find a frequency over most of the THz range, where short to medium path-length experiments can be performed at atmospheric pressure. Figure 1.2 shows the transmission under typical laboratory conditions.

Depending on the application, there are alternative units that may be more convenient for use in this spectral region. For example, the energy of the photon in electron-volts is very relevant for studying semiconductors, and the reciprocal centimeter (cm$^{-1}$), originally given the name “Kayser” after J. H. G. Kayser, who compiled a giant catalog of chemical spectra in the early 1900s, is convenient for describing spectra. The relationship between the various units is shown in Fig. 1.3. Before describing the history of the THz region, which began in the 1890s, it is relevant to consider the implications of the physical dimensions and electron energies shown in Fig. 1.3. In wavelength terms, 30–300 µm bridges the gap between the region where open path optical techniques are still convenient to use, up to the shortest wavelengths where waveguides can be employed. At 30 µm (10 THz), systems can be designed that are virtually identical to those used in the visible or near-IR regions with lenses, prisms, mirrors, etc., and spectroscopy can be performed with diffraction grating or Michelson interferometer instruments. However, as the wavelength is lengthened the size of optical components must be increased to avoid diffraction losses and interference effects. In the microwave region, these problems are avoided by restricting the radiation to a single mode in a waveguide. Although waveguides are manufactured for use up to ~2 THz, there

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**Fig. 1.2** Atmospheric transmission of a 1-m path at 1013 hPa and 40% relative humidity
are major difficulties when the dimensions are reduced to match short millimeter, or sub-millimeter wavelengths. A typical rectangular waveguide for single-mode transmission at a wavelength $\lambda$ has dimensions of approximately $\lambda/2$ by $\lambda/4$. Manufacture of very small cross-section guides of sufficient uniformity is difficult, but this is not the major problem. As a wave passes down a waveguide it sets up currents in the surface. These currents are required if the walls are to reflect the waves and, as the frequency of the electromagnetic wave is increased, only electrons near the surface of the waveguide have time to respond. In a resistive surface, this means that the effective resistance, through which the wall current flows, increases with frequency and the waveguide becomes increasingly lossy. Despite these difficulties, both optical and microwave-type arrangements are being employed over most of the THz region, and this has led to the development of what is called “Gaussian” optics. These are explained in more detail in Chap. 2.

While the physical dimensions of the wavelength range govern the types of spectroscopic systems used, it is the electron energy spread shown in Fig. 1.3 that makes this a fascinating region for research and application. The value of THz frequencies for a wide variety of studies can be readily understood by reference to Fig. 1.3. Room temperature corresponds to approximately 6 THz according to $h\nu = kT$. Hence, by varying the temperature from 14 K to 210°C, $h\nu$ can be made equal to $kT$ over the range from 0.3 to 10 THz with comparative ease. This is the unique feature of this part of the electromagnetic spectrum. In energy terms, the frequency range covers 1.2 to 37 meV, corresponding, for example, to the energy levels of many phonon bands, shallow impurities in semiconductors, and rotational interactions in gases. It should be stressed that the relationship between $h\nu$ and $kT$ is extremely important to a broad range of phenomena that can be studied in the THz region. For example, one noteworthy application is the investigation of the complex processes that lead to the formation and destruction of the ozone layer in the upper atmosphere. At THz frequencies, absorption and emission of radiation by gases are due to rotational transitions of molecules and the strength and the frequency of
maximum interaction is dependent on the molecular mass. The great advantage of
the THz region is that the strength of the interactions increases, as the square to cube
of the frequency, up to a mass-dependent maximum, but then falls exponentially at
higher frequencies. For the great majority of molecules of interest to those studying
the atmosphere, the peak absorption is in the THz range.

As mentioned earlier, now that there are a number of ways of reaching heights
where atmospheric absorption is greatly reduced, or avoided altogether, the way has
been opened for astronomers and astrophysicists to make a wide range of studies
in the THz region. Because the vast majority of the Universe is either cold or very
cold, the peak of the blackbody curves for most emissions falls into the THz region.
For example, this frequency band contains information on the cosmic background
at 2.7 K, which has a peak emission at 270 GHz while the interstellar medium, of
great interest for star-formation studies, is as low as 10 K in the cooler regions to
above 100 K in the denser parts. Thus, the peak emission from these gases varies
from 1 THz to above 10 THz.

1.2 History of Terahertz Research

Research in the “gap” between the IR and microwave spectral regions took a
giant step forward with the advent of THz pulse emission from photoconductive
antennas (T-rays) but, reading some of the 1990s papers, one could be forgiven for
thinking that these were the first experiments in the THz spectral region. In fact,
the earliest studies were a hundred years earlier, almost entirely due to the work of
Heinrich Rubens (Fig. 1.4), who was professor at the Technische Hochschule Berlin-
Charlottenburg (now: Technische Universität Berlin) and at the Universität Berlin.
The large majority of papers published up to 1920 were by Rubens, or researchers
who had collaborated with him, but from then on there was a steady flow of papers,
eventually leading to a rapid expansion from the 1950s onward. Rubens’ research
concentrated on the extension of the IR spectral region to longer wavelengths into
what soon became called the far infrared (FIR). In the late nineteenth century,
there was also research on short “electric” waves, following the experiments of
Heinrich Hertz, and Rubens participated in this, studying the polarization and
reflection of electric waves, before beginning his IR work. There was a remarkable
amount of microwave research in the last decades of the nineteenth century,
some of it reaching to frequencies close to 0.1 THz. Distinguished scientists were
involved, including Lodge, Fleming, Righi, Popov, and Lebedev, with theoretical
contributions from Lord Rayleigh. The most remarkable experiments were those
of the Indian physicist, J. C. Bose. Bose studied at Cambridge University, where
he came under the influence of Rayleigh. On returning to India he took up a
post at the Presidency College in Calcutta, and there he carried out a variety of
microwave experiments, later returning to England to give demonstration lectures.
Working at frequencies up to 60 GHz, he constructed prisms, lenses, grid polarizers,
reflection gratings, a horn antenna and a double prism attenuator. For detection he
used primitive point contact diodes. Bose measured the current–voltage curve of his junctions, noting their nonlinear properties and plotting curves very similar to those of modern diodes [8]. Sir Neville Mott, Nobel Laureate for his own contributions to solid-state electronics, is quoted as saying that “Bose was at least 60 years ahead of his time” and that “he had anticipated the existence of p- and n-type semiconductors.”

By 1900, the experiments of Marconi and others had shown that lower frequencies were much more useful for communication and interest in very high frequencies died. It was to be nearly forty years before its revival and it was to take another decade before there was significant encroachment into the THz region from the microwave direction. Progress in reducing the “gap” in the electromagnetic spectrum was to come initially by extending the IR to longer wavelengths.

During his thirty years of research into long-wavelength IR, Rubens had many collaborators, including several young Americans, and it was with one of them, B.W. Snow, that he performed his first significant experiments using prisms of rock salt (NaCl), sylvine (KCl), and fluorite (CaF$_2$) [9], eventually extending the spectrum to nearly 20 $\mu$m. IR prism spectroscopy was to have a long life-span, as it was widely employed into the 1950s. But it was another of the young Americans, E. F. Nichols, who made the discovery that opened the way to the first significant experiments in the FIR region. Working with crystalline quartz, he discovered that its reflectivity over a narrow wavelength range near 9 $\mu$m rose from a few percent to nearly that of a silvered surface [10]. This phenomenon, which Rubens later called the “reststrahlen” (residual ray) effect, is due to lattice vibrations and, with other crystals, Nichols and Rubens were to find similar narrow bands at longer wavelengths. With multiple reststrahlen reflectors, they were able to produce nearly monochromatic light at specific wavelengths. The elegant spectroscopic system used
by Rubens and Nichols [11] is shown in Fig. 1.5. After Nichols left Berlin, Rubens and his colleagues extended the techniques to wavelengths beyond $50 \mu \text{m} (6 \text{THz})$, and this research produced a dramatic outcome.

Toward the end of the nineteenth century one of the major problems engaging the attention of physicists and mathematicians was that of radiation from hot-bodies and, specifically, its variation with wavelength. A brief roll call of some of those involved is more than impressive: Wien, Angstrom, Rayleigh, Lummer, Pringsheim, Paschen, Larmor, and Planck. Various formulae for the radiation from an ideal source, the blackbody, were derived but none fitted all of the experimentally obtained results. A particular problem was to obtain really accurate data at very long wavelengths. In 1900, Rubens and Kurlbaum [12], using the reststrahlen spectrometer, obtained the necessary data and Rubens immediately visited Max Planck to give him the results and he, that same day, wrote down the equation which is now called Planck’s Radiation Law.

After several weeks of what he later described as the most strenuous work of his life, Planck revolutionized physics by inventing quantum theory to explain the radiation formula, which he had derived empirically by combining the already well-known short wavelength radiation results with the long wavelength ones of Rubens. And after the death of Rubens in 1922 Planck was to write: “Ohne das Eingreifen von Rubens wäre die Formulierung des Strahlungsgesetzes und damit die Begründung der Quantentheorie vielleicht in ganz anderer Weise, vielleicht gar nicht einmal in Deutschland zustande gekommen.” (“Without the intervention of
Rubens the formulation of the radiation law, and for this reason the foundation of quantum theory, would have been accomplished in an entirely different way, and maybe not even in Germany.”) [13].

1.2.1 Early Twentieth Century Research

As mentioned earlier, Rubens continued to dominate the FIR region until the time of his death. Between 1892 and 1922 just over 150 papers were published which included FIR research. Only 18 of them do not have Rubens as an author or joint author. Much interesting work was done, and three discoveries made at that time are still relevant today.

In 1911, Rubens and Baeyer showed that the mercury arc lamp in a quartz envelope was an excellent long-wavelength IR source [14]. In this region, the arc plasma is opaque and has an equivalent blackbody temperature of over 4000 K and, in modern grating spectrometers and interferometers, it is still the preferred source. One year earlier R. W. Wood, of Johns Hopkins University (USA), had produced the first “blazed” diffraction grating, which allowed the concentration of the majority of light from a source into a single order [15]. Wood achieved efficiencies of over 75% for the first order and showed that gratings of this type could be used out to beyond 100\(\mu\)m (3 THz). Although Wood produced the first blazed gratings, the initial conception was by Rayleigh in 1874 [16]. Apart from their use in spectrometers, blazed gratings, with efficiencies close to 100% in one polarization, are employed in lasers as tuning elements. In carbon dioxide lasers, for example, replacing one of the mirrors with a blazed grating allows tuning over many wavelengths between 9 to 11 \(\mu\)m. These lasers are widely used as the “pump” for the optically excited THz lasers described in Sect. 4.4.2.

The third discovery, by Hagen and Rubens, concerned the reflectivity of metals [17]. They showed that the reflectivity depended on their electrical conductivity. The formula that they found empirically, which was later explained theoretically, shows that in the THz region any pure metal has a reflectivity of well over 99% so that even complex reflecting systems are virtually loss-free.

The “Rubens’ era” ended with an experiment where he used all his spectroscopic skills to measure the absorption of water vapor out to 400\(\mu\)m [18] (Fig. 1.6). This was a remarkable achievement, as it had to be done point-by-point without any electronic aids.

1.2.2 The Years 1920–1940

Between 1920 and 1930 some 40 FIR papers were published and from 1930 to 1940 another 80. In 1920, after a relatively short but glittering career after leaving Berlin, E. F. Nichols became Director of Pure Science at the National Electric Light Association in Cleveland, Ohio. Here, before his untimely death in 1923, he returned to long-wavelength research, working with J. D. Tear, and in 1923 they succeeded in
1.2 History of Terahertz Research

Fig. 1.6 Absorption of water vapor measured by Rubens in 1921 with a wire grid grating spectrometer. Traces A and B were measured with different illumination of the spectrometer slit by the mercury arc lamp. The dashed line around the minimum a’ was measured with higher resolution. Rubens attributed the two minima in trace B at the position a’ to water absorption. The origin of the minima b’ and c’ could not unambiguously be identified. Comparison with the spectrum in Fig. 1.2 shows that Rubens’ assignment was correct (from [18]).

joining the electric wave spectrum to the IR. Their experimental setup is shown in Fig. 1.7. Using a Hertzian oscillator, wavelengths down to 220μm were produced, which overlapped with the extension of wavelengths from a mercury arc source to 420μm [19]. In 1924, a Russian physicist, Glagolewa Arkediewa, showed that radiation of about 90μm could be generated by exciting small Hertzian oscillators in the form of brass filings immersed in oil [20].

Apart from Berlin, the main laboratories involved in long-wavelength IR studies were at the University of Michigan, where H. M. Randall was to be a dominant figure in IR research for many years, and the California Institute of Technology, where the usefulness of the FIR region for the study of the rotational levels of gases was realized by R. M. Badger [21], who was later joined by C. H. Cartwright and J. Strong. Both Cartwright and Strong made significant contributions to improving FIR techniques between 1930 and 1940, with Cartwright following in the footsteps of earlier American researchers by spending time in Berlin. He made detailed studies of the absorption of a variety of materials [22] and designed a liquid oxygen-cooled thermopile with more than ten times the responsivity of its room-temperature
Fig. 1.7 Nichols and Tear in a laboratory of the National Electric Light Association in Cleveland, Ohio, where Nichols was the Director. The Hertzian type oscillator is within the metal container on the left. The output is collimated by a paraffin wax lens onto a Boltzmann interferometer which Nichols is adjusting. The reflected beam is then focused by the second lens onto a radiometer detector (for details of the experimental setup see [19], photograph courtesy of J. D. Tear’s relatives)

counterpart [23]. Strong collaborated with Randall on a “Self-Recording Spectrometer” [24], went on to study the absorption of 14 gases between 20 and 200 μm [25] and later, at Johns Hopkins University, made outstanding contributions in the optical field. In his latter years, he was one of the first to pursue the transition from grating to interference spectroscopy at THz frequencies [26].

Although there were major improvements in spectroscopic systems throughout these decades, there were only marginal improvements in sources and detectors. Thermocouples, thermopiles, or bolometers, feeding into very sophisticated galvanometers, were the detectors, usually with a mercury arc or some heated material such as the Welsbach mantle as a source [27].

Probably, the most significant experiment of the 1930s was the observation of the molecular absorption of ammonia gas over the wavelength range 1–4 cm using a coherent source, a split-anode magnetron [28]. This was the prelude to the field of microwave spectroscopy, which expanded dramatically from the late 1940s onward, when new coherent sources, such as the klystron, became available for research applications.

1.2.3 The Years 1940–1950

Not surprisingly, 1940–1946 was a barren time for research publications but, of course, there were many developments during this period. Of particular note is the research in Germany to extend the range of prism spectroscopy. Up to 1940,
25 \mu m was the maximum wavelength using KBr prisms but, with the mixed crystal thallium–bromide–iodide, KRS-5 (KRS standing for “Kristalle aus dem Schmelzfluss”, crystals from the melt), coverage was extended to 37 \mu m [29]. The introduction of cesium bromide [30] and cesium iodide in the early 1950s allowed prism systems to operate out to nearly 60 \mu m [31].

A very significant post-war event was the introduction of the pneumatic detector, now almost always referred to as the “Golay” [32]. This elegant device came within a factor of three of the theoretical limit for a room temperature detector. Although the detector is named after Golay, it is interesting to note that the first published paper was by H. A. Zahl and M. J. E. Golay [33] and Zahl filed a patent for a “pneumatic cell detector” in 1938. In 1939, Zahl also patented a “System for detecting Sources of Radiant Energy” with co-inventor M. J. E. Golay. At that time, there was considerable effort toward methods of detecting hot objects by their radiant emission. The first cooled bolometer was also a 1940s invention. This used the superconducting transition point of tantalum at 4.4 K, thus requiring the use of liquid helium [34]. A later version was made with niobium nitride (NbN), which has a transition at 14.3 K, a temperature which could be obtained by pumping on the then more readily available liquid hydrogen [35].

There were huge advances in electronics during the Second World War and this produced a dramatic change in spectroscopic systems. Virtually, all experiments up to 1940 used DC recording systems but from 1945 onward detectors were designed with shorter time constants, the incoming radiation was “chopped” and AC amplification employed. One earlier exception was a recording spectrograph for the FIR [36] covering the range 18–200 \mu m which used a very low frequency amplifier that had been designed as early as 1932 [37].

The development of high-frequency radar systems had led to new magnetron and klystron sources, and these were employed for a number of microwave spectroscopy experiments following on from Cleeton’s earlier measurement of the ammonia inversion spectrum. With a klystron, frequency doubled in a crystal harmonic generator, the absorption of oxygen between 50 and 60 GHz was measured in 1944 at the Massachusetts Institute of Technology (MIT) with the results published in 1946 [38]. Among those involved were W. Gordy, who later wrote a review covering this early research [39], and C. H. Townes and his colleagues [40] who, by 1952, had reached frequencies of 270 GHz [41]. In 1954, microwave spectroscopy was extended into the submm region by Gordy’s research group [42], thus providing a clear overlap with the longest wavelength IR spectroscopic systems. Gordy had a remarkable career, beginning his research with studies of hydrogen bonding. During World War II, he was at the MIT Radiation Laboratory, where he participated in the development of microwave radar, immediately realizing the potential of microwave technology for molecular spectroscopy. Joining the Physics Department at Duke University in North Carolina in 1946, he was instrumental in setting up a microwave laboratory to pursue research in what he described as the “gap in the electromagnetic spectrum.” This laboratory, which moved to Ohio State University in 1990, remains prominent in submm research after six decades. Notable among Gordy’s 75 PhD students is Frank de Lucia, who has been at the forefront of THz research since 1970.
1.2.4 The Years 1950–1960

By the end of the 1940s, the stage was set for a major expansion of IR spectroscopy and for the extension of microwave techniques into the THz region. The distinguished IR and FIR researcher, Armand Hadni, has described the period as the start of the “explosive” years [43], but the number of scientists involved in what was still considered to be a very difficult spectral region was relatively small, with less than 200 papers published in this decade. The majority of these came from the relatively few laboratories which had significant FIR groups. The important centers could almost be counted on the fingers of one hand: J. D. Strong at Johns Hopkins University, USA, later to be joined by G. A. Vanasse; E. E. Bell, and R. A. Oetgen at Ohio State University, USA; Earl Plyler at the National Bureau of Standards, USA; A. Hadni at the University of Nancy, France; L. Genzel at the University of Frankfurt, Germany, who was later to move to Freiburg; H. Yoshinaga, at Osaka University, Japan, and H. A. Gebbie at the National Physical Laboratory, England. There were also other more isolated researchers who were to have great significance in the opening up on the FIR region, including P. B. Fellgett at Cambridge in England and P. Jacquinet at Orsay, France. Toward the end of the 1950s new names appeared and these, with others, were to launch the FIR into the mainstream of optics research. Notable figures were P. L. Richards (USA), N. G. Yaroslavski (USSR), E. H. Putley (UK), and D. H. Martin (UK).

Grating spectroscopy still dominated, with the mercury arc as source and the Golay as detector, but improved electronics and recording systems greatly speeded up research. However, it was in this decade that the foundations were laid for the transition to FTS, which was to revolutionize FIR spectroscopy. Although the transition from gratings to FTS progressed comparatively slowly, mainly due to the limitations of early computers, the outcome marked a development that was certainly as important as the more dramatic arrival of THz TDS some three decades later. It was well known that interferometers such as the Michelson were excellent for the accurate study of monochromatic light but when there were more than three or four wavelengths from a source the interferogram produced was very difficult to analyze. Even so Michelson, with infinite patience and his own remarkable invention of a “harmonic analyzer” [44] (which was in essence a mechanical analogue computer) was able to derive spectra by observing fringe intensity. Both Michelson and Rayleigh were aware that the full spectrum could be obtained by Fourier transformation but had no way of performing this. The first person to derive an actual spectrum was Peter Fellgett, while studying stars in the IR. In the visible and very near-IR, a spread of wavelengths could be observed by placing a photographic plate at the exit of a diffraction grating monochromator but, at longer wavelengths, until the comparatively recent availability of detector arrays, only one wavelength could be studied at a time. After various efforts at coding different wavelengths with an ingenious chopping system – a method also employed by Golay [45] Fellgett realized that if a two-beam interferometer were to be placed in the path of undispersed radiation and its path difference scanned at a uniform rate, the
resultant signal from a single detector would contain information at all wavelengths simultaneously. Fellgett had to transform his interferogram by hand, using devices called Lipson–Beaver’s strips, a very tedious task [46].

The ability to study a wide range of wavelengths simultaneously became known as the multiplex or Fellgett advantage. What he had not immediately realized was that, while for high-resolution grating spectroscopy very narrow slits were required, thus reducing the amount of light passing through the instrument, interferometers had the further advantage of having large apertures. This was first pointed out by Pierre Jacquinot and came to be known as the Jacquinot (or throughput) advantage. In the 1950s, a number of experimenters, including Jacquinot, Connes, Strong, Vanasse, and Gebbie, began research with interferometers. The throughput, multiplex and resolving power advantages of interferometers were soon recognized but the limitations of early computers meant that it might take days to analyze the interferograms. When this limitation was overcome in the following two decades, FTS became the dominant force in spectroscopy in the FIR.

Jacquinot [47], Strong [48], Fellgett [49], and Gebbie [50] have written short review papers concerning the development of FTS in this period. Connes, in a detailed paper [51], has described the contribution of Michelson and even earlier researchers in the field. He points out that the modern form of FTS was almost discovered by Rubens and his collaborators in 1910 [52]. On the detector side, the Golay was virtually the only FIR detector of the 1950s but in 1959 the first convenient-to-use cooled device, the carbon bolometer, was invented [53] and the same year saw the first photoconductive detector to reach wavelengths longer than 100 μm [54].

Although no fundamental electronic source was produced for frequencies above 300 GHz in the 1950s, the foundations were laid with the first backward wave oscillator (BWO) or carcinotron [55, 56]. Extension to higher frequencies followed in the 1960s and BWOS are now available to above 1 THz, with a tuning range of ±10% of the center frequency.

Of the papers published in this decade, 80% were on applications, which included the first studies on semiconductors, including cyclotron resonance measurements [57], many papers on the absorption spectra of gases and, at 0.2 THz, observation of solar and lunar radiation [58]. Early work on the transmission of superconductors began, which was to lead to the direct measurement of the energy gap predicted by the BCS theory [59]. This decade also saw the first FIR diagnostics on the high-temperature plasmas produced in the attempts to achieve controlled thermonuclear fusion. These early experiments succeeded in measuring both the average electron density and the electron temperature in several different plasmas [60].

The late 1950s also saw the first steps on the long journey that would lead to the development of high-power gyrotrons. The earliest suggestions appeared in theoretical papers published in Australia [61] and the USSR [62], with the first experimental studies being made at microwave frequencies in the USA [63].
1.2.5 The Years 1960–1970

This was a flourishing decade for the THz region, with well over 1000 published papers and several new groups appearing. One of these, that of Fritz Kneubühl in Switzerland, was to have far-reaching consequences because, apart from his group doing outstanding FIR research, he, from the mid-1970s, organized a series of conferences in Zürich devoted to the IR and FIR spectral regions. Another series of conferences was to have longer-term importance. These began with a meeting in the USA at Atlanta in 1974, entitled “The First International Conference on Submillimeter Waves and their Applications.” The guiding hand was that of K. J. Button, then at MIT, who predicted then that it “would be the first of a long series because the submillimeter wave specialty has become well-defined and moderately well-populated.” Rome, in 2010, saw the 35th Conference in the series. But returning to the 1960s, the significant feature of this period is that it produced much of the instrumentation that is widely used today. If one looks back to the time before 1960 the only inventions which have not been superseded, apart from the spectrometers themselves, are the mercury arc source and the Golay detector. And for many applications the Golay detector is now replaced by the cheaper, and more convenient pyroelectric detector. Pyroelectric detectors date from this era although the use of the pyroelectric effect for detection had been suggested as early as 1938 [64]. The 1960s onward saw great progress in detector development with the n-InSb electron bolometer [65], the Ge bolometer [66], and a tunable FIR detector [67]. One of the most widely used detectors, the extrinsic photoconductor Ge:Ga, also dates from this decade [68]. When stressed this responds out to 200 μm [69]. While the “Putley” detector is still very widely used the Ge bolometer has been largely superseded by the more versatile Si composite bolometer [70]. A further helium-cooled detector based on the Josephson effect was also introduced [71]. One of the authors of this paper was Paul Richards who, in 1966, set up his research group at the Berkeley campus of the University of California and has been an outstanding figure in FIR/THz research over more than fifty years. Another important step was the invention of the so-called “honeycomb” diode chip design for Schottky diode mixers by D. T. Young and J. C. Irvin [72]. Originally applied to mmW frequencies this design became widely used in the 1970s and 1980s.

The major discovery at the start of this decade was the laser. The first THz laser, the water vapor laser, was invented in 1964 [7]. This was followed by several other long-wavelength gas lasers, including HCN, which provides several continuous wave (cw) lines close to 1 THz [73]. Lasers were to become a major tool for THz research in the proceeding years.

What was perhaps the most far-reaching paper published in this decade was a two-page Letter in the journal “Nature,” which began “A Michelson two-beam interferometer can be used for both refractive index determination and absorption by measuring the shift of the achromatic fringe when a specimen of known thickness is placed in one arm of the interferometer.” This was the birth of “dispersive Fourier-transform spectroscopy” (DFTS), which has proved to be a powerful
technique for studying the optical constants of solids, liquids, and gases in the IR and THz regions [74]. Even today, when much spectroscopic work is performed with TDS systems, considerable routine and innovative research is still performed with DFTS instruments. Credit must also be given to E. E. Bell, who reported independently on the advantages of using both arms of a Michelson interferometer to provide absorption and refractive index information [75]. In 1969, D. H. Martin and E. Puplett introduced a polarizing version of the Michelson interferometer that has become widely used in the THz region [76].

Grating spectroscopy was still widely used in this decade because obtaining spectra from interferograms was so time-consuming, although the use of a fast Fourier-transform algorithm introduced by J. W. Cooley and J. Tukey had reduced the time required by some two orders of magnitude [77]. A specific problem of grating systems is the need to suppress higher order radiation which, with thermal sources, is at a much stronger level than the first order. This difficulty was to a large extent overcome by the use of capacitative grid filters by R. Ulrich in 1967 [78]. These are described in Sect. 3.5. Another very helpful discovery was that the polymer TPX is transparent over much of the FIR [79]. This is a hard plastic, particularly useful for windows and lenses, with the great advantage of having virtually the same refractive index in both the visible and THz spectral regions.

The year 1968 saw the birth of time-domain spectroscopy (TDS), although in the microwave region, with the publication of a paper entitled “BroadBand Microwave Transmission Characteristics from a Single Measurement Transient Response” [80].

### 1.2.6 The Years 1970–1980

The year 1970 saw the arrival of one of the most useful laser sources, the optically excited THz gas laser [81]. These now produce hundreds of cw and many thousands of pulsed wavelengths throughout the THz region. On the strongest cw lines output powers of above 100 mW are available from commercial instruments with pulsed powers of several kW.

Also in 1970 carbon monoxide (CO) was discovered for the first time in the interstellar medium with a Schottky-diode heterodyne receiver operating at 115 GHz [82]. Seven years later, T.G. Phillips et al. made the first real THz detection of an interstellar molecule [83]. They detected CO at 0.345 THz with a heterodyne receiver based on an InSb hot electron bolometric mixer. The 1970s were the pioneering years for astronomy at THz frequencies, mainly because of significant progress in receivers and because high altitude observatories became available. Most notably, the Kuiper Airborne Observatory (KAO) started its operation in 1974.

The first THz imaging experiments for other than astronomical applications were reported by D. H. Barker et al. in 1975. Already at that time the authors pointed out that “an FIR imaging system could be developed for industrial, military, law enforcement, and medical applications in the next few years” [84].
Early in this decade the first THz TDS results were obtained by the Paul Richards group, using a mode-locked Nd:glass laser and lithium niobate as an electro-optic (EO) crystal [85]. Due to the comparatively long pulse length of about 2 ps the shortest wavelengths produced were about 700 μm. Similar results were obtained independently by a Japanese group [86].

There was further significant progress toward improved detectors in this decade. Detection using metal–semiconductor contacts, so-called cat’s whiskers, dated back to Bose’s research in the 1890s. These are also the devices that Gordy used to provide harmonics of the fundamental frequency. Now described as Schottky diodes, these were already familiar devices for both direct detection and as mixers in microwave and mmW heterodyne systems. It was realized that if these diodes could be designed to operate at higher frequencies, they would have the great advantage, compared with other fast THz detectors, of operating at room temperature. Because various relatively high-power sources were also becoming available, there was the opportunity to extend microwave-type heterodyne systems to higher frequencies. A very useful review of the remarkable progress in Schottky diode optimization was written in 1980 [87]. As Gordy had shown, Schottky-type diodes are not only useful for heterodyne detection systems. Due to their nonlinear characteristic they are valuable as harmonic generators. Gunn and IMPATT sources were producing significant power levels in the mmW region and the use of these with Schottky structures began to provide significant harmonic power deep into the THz range.

A main reason for developing room-temperature systems for the THz region was that various platforms were becoming available for both astronomy and upper atmospheric research. However, video detection remained important and it was well known that the background noise that limited the ultimate detectivity of extrinsic photoconductive detectors, such as Ge:Ga, when used in a 300 K environment, would be greatly reduced in space applications. Intensive research on improving Ge:Ga detectors led to an improvement of four orders of magnitude in their performance [88]. Another He cooled detector that dates from this era, and was to become very important, particularly for THz heterodyne systems, is the superconductor–insulator–superconductor mixer (SIS) [89, 90].

### 1.2.7 The Years 1980–1990

The improvements in THz technology over the previous thirty years had led to a widespread recognition of the usefulness of this spectral region in many disciplines. Developments in computer technology had made FTS systems the choice for spectroscopy throughout the IR and THz regions. But, with an increasing interest in astronomical and upper atmosphere research, it was realized that heterodyne systems were required for these and other applications to achieve the necessary resolving power. This can be understood by looking at a paper published in 1984, which reported a balloon-borne FT interferometer study of stratospheric emission at ~40 km altitude [91]. At the shortest wavelength observed of 110 μm the resolving
power was \( \sim 27,000 \), falling to just over 2,000 at the longest wavelength of 1.4 mm. This instrument had a path length difference of up to 1.5 m between the fixed and moving mirror and was approaching a realistic limit in the resolving power of an FTS instrument. Despite this impressive achievement, the resolving power is not sufficient to determine the true lineshape of a molecular line.

The need for higher resolving powers was pointed out in a review paper outlining the state-of-the-art of diagnostics on interstellar plasmas [92]. What was required was the extension of the high resolution of heterodyne systems familiar in the microwave region up to THz frequencies. Until that time the only heterodyne studies had been with the “Putley” InSb electron bolometer, which has a very limited bandwidth. In the short-mm region rectifying diodes, usually Schottky devices were normally used as the mixer with electronic sources providing the LO power. The main problem in reaching higher frequencies is that losses increase, resulting in a requirement for more local oscillator (LO) power, typically in the mW range. At that time, the only practical cw sources available for frequencies above \( \sim 0.5 \) THz were optically excited gas lasers. Although these only produce specific wavelengths, a successful heterodyne detection of an interstellar CO line at 434 µm was achieved in 1981 [93] by mixing the CO line with a laser line of similar wavelength. Although optically excited gas lasers are normally bulky, a sufficiently compact version for use in the Kuiper Airborne Observatory (KAO) was designed and used for astronomical studies [94, 95]. The KAO was one of a number of platforms that were becoming available for both astronomy and upper atmospheric research, and for these and other applications the search for better detectors and mixing devices, as well as for improved sources to act as LOs, was intensified. The 1980s also saw the first IR/THz space-borne observatory. The Infrared Astronomical Satellite (IRAS), launched in 1983, was the first observatory to perform an all-sky survey at IR wavelengths, including two THz bands around 60 and 100 µm. This was the first of a very successful series of space-borne IR/THz observatories.

As already mentioned, conventional extrinsic Ge detectors had been hugely improved and were becoming widely used in astronomy and atmospheric studies. However, there was a particular problem with these detectors when used in space applications. This was because excess noise was produced when high-energy photons struck the detectors. This disadvantage was to a large extent overcome in the 7–10 THz region by the invention of blocked-impurity band detectors [96].

A fundamental source covering the frequency range from 1 THz to above 4 THz, invented in Russia and Japan in this decade, was the p-type Ge laser [97, 98]. Operating at or near liquid helium temperature, this is a useful tunable pulsed source that can also be operated in near cw mode [99]. Ge lasers are electrically excited and, as the power requirement, especially for small versions, is quite low, these can be conveniently packaged with a mechanical cooling device [100].

The first free electron laser (FEL) operated in the early 1970s and by 1984 Elias and his colleagues [101] had constructed a laser operating between 0.3 and 0.77 THz, with a peak power of 10 kW, as a user facility at the University of California at Santa Barbara. One of its main application areas was in biology, including photobiology. Since then a number of THz laser user facilities have been
installed in France, Germany, Italy, Russia, The Netherlands, and the USA, which are providing very valuable results in a wide range of disciplines. The tunable high-power and short-pulse output of FELs means that they are particularly useful for pump-probe experiments and time-resolved spectroscopy.

Alternative facilities for THz research are the IR ports of certain synchrotrons [102]. These provide a significantly higher average source brightness than an arc lamp in the 0.3–30 THz region and the ps pulse structure is useful for a number of applications. Stable coherent synchrotron radiation (CSR), first observed in 2002, has produced a power increase of $10^5$ over the 0.3–1 THz range and is an exciting development for spectroscopy [103].

The dramatic event of this decade can almost be described as the “rediscovery” of the FIR region of the electromagnetic spectrum. With the advent of high-power, very short-pulse length near-IR lasers, the pioneering research of Nicholson in the microwave region, and Richards and Yajima, with their colleagues, in the FIR, came to fruition. The essential features of this system of THz research are fairly simple. When a very short pulse, typically 50 fs or less, of near-IR radiation falls on a photoconductor, THz pulses are generated within the photoconductor, which then radiates. The highest frequency produced depends on both the length of the laser pulse and the mobility of the electrons in the photoconductor. An alternative arrangement is to use an EO crystal where the ultra-short laser pulse changes the electrical polarization and causes THz radiation to be emitted. In this case, the upper frequency produced only depends on the length of the laser pulse, as no charge carriers are involved. In practice, the essential difference between the two systems is that photoconductive devices produce higher power but are less efficient above 3 THz. EO crystals provide less power, in comparison with photoconductive devices with the same excitation power, but can reach to above 100 THz. In both cases, the spectrum is obtained from the Fourier-transform of the emitted pulse.

In early experiments, the power levels produced were quite low. However, in 1984 Auston and his colleagues [104] introduced what became known as the “Auston switch,” to produce higher powers, and used these to study the transmission and absorption of various materials. This paper created widespread interest and, as mentioned previously, some of the early accounts seemed to suggest that this was an entirely new field of research.

Just before the close of this decade the first results of a new detector that was to become widely used as a THz mixer were published by G. N. Goltsman and coworkers. This was the hot electron bolometer (HEB), remarkable for the fact that it is a thermal detector [105]. Essentially, this relies on the same mechanism as the first helium-cooled detectors, reported in 1942 [34], as it operates in the narrow temperature range that marks the transition from the normal to the superconducting state of a superconductor. The particular advantage of this bolometer made of a thin NbN film is its combination of a small heat capacity with high heat conductivity, giving a time constant of approximately 40 ps for small devices. This is equivalent to a bandwidth of several GHz, thus making it very useful as a mixer. Furthermore, as it is a fast thermal detector it is useful throughout the IR and THz regions.
1.3 Reasons for Increased Interest

From being a rather specialist sector of the electromagnetic spectrum for research, the THz region has become of great importance in a wide range of disciplines. These include many aspects of space research, astronomy, atmospheric studies, biology, medicine, plasma physics, chemistry, nondestructive testing, and process control. There are also a number of important defense and security applications. For all of these applications, imaging as well as spectroscopic systems have been developed, and there are continuous, intense efforts around the world to improve their performance and capabilities.

A major reason for the increased interest in THz research comes from astronomy. The THz part of the electromagnetic spectrum possesses an amazing scientific potential for spectroscopy and imaging. Many absorption and emission lines of the important astrophysical and astrochemical molecules and atoms occur in the THz frequency region. By 2011 approximately 150 molecules and atoms have been detected in space. There are fairly simple molecules, such as CO, but molecules consisting of ten or more atoms have also been detected. Besides their existence and abundance, valuable information about the physical conditions such as temperature, density, and dynamics of the observed astronomical object can be obtained from spectroscopy. Many galaxies or interstellar clouds emit most of their energy at THz frequencies, either as broadband blackbody-like emission or as line emission from molecules and atoms. Imaging and photometry allow the study of the structure and morphology of these objects. Ground-based telescopes at many locations around the world, as well as the space-borne THz observatories IRAS, ISO, SWAS, Odin, Akari, and Spitzer, have contributed significantly to the exploration of the THz universe. At the time of writing this book ESA’s Herschel Space Observatory is in operation and will revolutionize our view of the THz universe.

It is the demand from these observatories that has driven forward the development of detectors, radiation sources, and spectrometers. The requirement for very sensitive detectors and large format detector arrays has led, for example, to the development of Ge:Ga photoconductive detector arrays with hundreds of elements, and superconducting composite bolometer arrays with thousands of elements. Amazingly, the sensitivity of these detectors is background noise limited. For heterodyne spectrometers SIS and HEB mixers approach the quantum noise limit, and small heterodyne arrays are now operational at several telescopes. The development of local oscillator (LO) sources has made all-solid-state multiplier-based THz sources with μW to mW output power available up to 2 THz. New observatories such as the Stratospheric Observatory for Infrared Astronomy (SOFIA), or the Atacama Large Millimeter Array (ALMA), which comprises an array of 66 giant 12-meter and 7-meter diameter telescopes for mmW and submm/THz observations, will continue to provide a strong incentive for the development of THz receivers and spectrometers.

Imaging has become an important application in the THz region. Some of the initial excitement of the early TDS research was the “discovery” that a variety
of materials, including clothing and packaging materials, which are opaque in the visible and near-IR regions, are reasonably transparent over much of the THz range. Imaging had already been performed using conventional THz techniques [106] but TDS imaging has found wide application over diverse disciplines, since the first paper was published in 1995 [107]. This also has the advantage of providing three-dimensional imaging. Because THz TDS does not require cooling, while providing large spectral coverage and high sensitivity at the same time, this technique rapidly spread and found new applications in research as well as paving the way for commercial uses. Along with THz TDS, breakthroughs in component development have contributed to the rapid development of the field since 1990. Most notable has been the appearance of THz quantum-cascade lasers (QCLs) in 2002 [108], which now cover a large part of the THz region with cw and frequency-tunable radiation. In addition, these only require modest cooling and provide mWs of output power. Such systems have been employed for a large variety of proof-of-principle experiments [109] for applications in biomedicine, nondestructive testing, security, and other fields such as art conservation [110].

Fourier-transform spectroscopy is commercially available and is the workhorse in chemistry for spectroscopy of gases, liquids, and solids. THz spectroscopy with these spectrometers is widely used in chemistry not only with pyroelectric detectors, but also with liquid helium-cooled bolometers. Early research on astrochemistry activated advances in gas spectroscopy of simple molecules in the laboratory, but current increased interest in THz spectroscopy is related to the extensive computer power now available. This allows calculation and simulation of an expanding number of molecules with many atoms, weak interaction forces related to the structure of biomolecules like DNA and proteins, and interaction of molecules with a large number of solvent molecules, for example, for the investigation of chemical reactions in solvents such as water.

Both large mass and weak interaction forces lead, independently, to low vibration frequencies located in the THz frequency range. Also, collective modes in crystal lattices consisting of many, and even assorted, molecules, as well as liquids, show THz spectra and complex THz responses, respectively. Polymorphs are very difficult to separate by IR spectroscopy, but show significantly different spectral signatures in the THz frequency range. High signal-to-noise ratios, now available with room temperature THz spectrometers, in contrast to THz Fourier-transform spectrometers, may lead to applications in the pharmaceutical industry to distinguish generic drugs, and to support patent applications on newly developed pharmaceuticals. Molecular systems, and reactions with surfaces and interfaces, are of major interest for the development of new catalytic and energy efficient processes, as well as in energy storage, such as batteries, which are relevant for future electromobility. THz spectroscopy can investigate the interaction of molecules with surfaces because the heavy mass of the surface lowers vibrational frequencies into the THz range.

While common chemical reactions are performed in solvents, the reaction formula is typically written down neglecting solvent molecules. It has recently been recognized that the solvent can play a more active role in many chemical reactions, beyond just controlling the diffusion of molecules for the reaction. Especially for
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biomolecules, water, the solvent of life, actively participates in interactions, and these can be probed on ps and sub-ps timescales corresponding to THz frequencies. A current large new class of solvents, ionic liquids, is also being considered for industrial and research applications. Since many macroscopic properties, for example viscosity, may be related to THz responses, it is conceivable that THz spectroscopy may also help to develop designer fluids.

THz TDS will, in general, have a strong impact in chemistry, medicine, and biology. With the development of sub-ps mode-locked dye lasers in 1972 by Ippen and Shank [111] and subsequently, in 1986, the development of titanium-sapphire (Ti:Sa) lasers by Moulton [112], it did not take long for chemists to use such lasers to investigate chemical reactions. Short sub-ps pulses and pump-probe techniques allow the study of kinetics and dynamics of chemical reactions, which range from femtoseconds via milliseconds up to seconds. Since misfolding of proteins triggers a variety of illnesses, unraveling the process of protein folding, which takes place in many subsequent stages from femtoseconds to seconds, will be of prime medical interest.

THz pulses also permit the study of processes which are far from equilibrium, because the time scales involved are much shorter than thermal time scales. Plasma reactions for research and industrial applications can be studied in detail, with the added advantage that the thermal background of the typically hot plasma is negligible when coherent detection systems are used. Many processes, for example near equidistant rotational lines of molecules, are coherently locked for short time scales after excitation, and THz short pulses can provide coherence and dephasing times relevant to the understanding of molecular processes. Short pulses are also very relevant to condensed-matter research.

Although optical and electronic components to manipulate amplitude and phase of short pulses are readily available in the optical range, such components are in their infancy in the THz range. This, in part, has fuelled interest in systems based on plasmons, metamaterials, photonic band gap materials, and fast semiconductor switches, which may contribute to new components for THz pulse manipulation and shaping. Nuclear magnetic resonance is a standard technique for identifying chemicals, whilst in medicine the technique can be used to generate images in magnetic resonance imaging. Special pulses, pulse trains, and sequences of pulses allow a variety of applications and methods related to multi-dimensional spectroscopy. Similar techniques permeate into the THz field and may provide new insights into materials. Medical imaging is still in its infancy but, for example, the high absorption of water and low absorption of fatty tissue contribute to THz contrasts, and may lead to medical THz imaging augmenting the presently available infrared and ultrasound techniques.

The increasing need for extra bandwidth for communication may also impact heavily on THz technology and component prices, especially in the low THz frequency range where research applications are approaching 0.3 THz. However, the strong absorption of THz waves in air will restrict their use to local networks. This is beneficial because it allows an increased occupancy of frequency bands for spatially separated networks. Therefore, in-house and, in general, short-range (and
therefore locally secure) broadband communication is possible, without interference between neighboring networks in the same frequency band. Proximity initiated communication links are in service in some countries in the 0.1 THz range.

Many research centers, such as laser facilities, synchrotrons, and free electron lasers are increasing the number of THz sources and beam lines for external users, but much of the increased interest stems from the close connection between electrons and THz incoherent and coherent radiation. THz spectroscopy is an excellent tool for the diagnostics of charged particle beams [113,114] and may even revive interest in electron systems and tubes, although, most probably in micro- and nanoscale devices [115], currently being investigated for new types of plasma displays.

This close relationship of electrons and THz radiation is also reflected in the use of THz spectroscopy to investigate charges in semiconductors, and to measure conduction without the need to apply physical contacts and electrodes. The latter is especially relevant, because an increasing number of nanostructured and nanoscale materials are being applied to devices, ranging from fast electronics to systems converting, for example, solar energy into electrons, which are key fields of interest.

The increased interest in THz techniques is also related to their broader availability and, in many situations room temperature operation, which allows users from different disciplines to incorporate THz technology into their research or applications. This is in part a consequence of enabling technologies such as ultra-fast optics, nanopositioning, fast electronics for regulation and stabilization, advanced material processing, increased speed in simulations, and computer power. The proliferation of THz techniques from research into everyday life is benefiting from declining prices of commercially available components such as femtosecond fiber lasers, liquid-cryogen free cooling systems requiring little maintenance, sub-nanometer precise piezo-positioning systems, and off-the-shelf GHz electronics.

To summarize, there has been a push resulting from major THz technological breakthroughs, leading to significantly improved performance, greater ease of use and, last but not least, more affordable systems. This is being accompanied by a pull from both research and “real-world” applications. The combination of both push and pull has stimulated the dramatic increase in THz research and development that has occurred since about 1990, and will continue in the foreseeable future.
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