



# Recent advances in terahertz imaging: 1999 to 2021

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## Abstract

We discuss the progress in the field of THz imaging based on time-domain spectroscopy during the last 20 years emphasizing several highlights. These include 3D mapping of the water distribution of plants, THz reflection imaging of samples with arbitrary shape, burn wound imaging and the early diagnosis of diabetic foot disease. These applications greatly benefit from the introduction of fibre-coupled THz time-domain system operated by rugged and portable femtosecond fibre-lasers. THz imaging is a versatile measurement method that has a plethora of practical applications and great promise for the future.

## 1 Introduction

The field of terahertz science and technology is gradually reaching a level of maturity that is reminiscent of, if not yet comparable to, that of its electromagnetic neighbors in the microwave and infrared regions. Much of this progress has been motivated by the idea of generating images with terahertz waves [1]. In this article, we will update and put in perspective the progress experienced by terahertz imaging over the last two decades. In a sense, it is a sequel of the article “Recent advances in terahertz imaging” [2] that shares two authors with this publication and that appeared in this same journal in 1999. We do not intend to provide an exhaustive literature review, but instead to give an overview of how terahertz imaging and its applications have evolved, particularly in the specific areas that were discussed in the original 1999 article. We aim to provide some examples to illustrate the perspectives of this fascinating and promising field.

Terahertz spectroscopy has seen enormous changes over the last two decades. After the appearance of terahertz time-domain spectroscopy (THz-TDS) in the 1980s [3, 4], which gave unprecedented access to the, at the time, elusive band of the electromagnetic spectrum that fell between the microwaves and the mid-infrared, it was more or less natural that people would start trying to produce images with this type of radiation [5]. Commercial terahertz systems did not exist, and the possibility of transporting these spectrometers or operating them outside a controlled laboratory environment was out of the question, mainly due to the dimensions and lack of stability of the solid-state lasers which were the heart of most THz-TDS systems.

Much has changed in the interim. A first step towards more flexible THz systems was made by Picometrix (now known as Teramatrix), who developed THz-TDS system with fiber-pigtailed antennas in 2001 [6]. This advance enabled many measurements that had previously been impossible such as those involving light scattering [7] or spatial characterization of wave fronts [8], due to the fact that the antennas could now be flexibly repositioned. At that time the system was still based on a titanium-doped sapphire femtosecond laser, and the fibre-coupled antennas were still made from GaAs. The advent of antennas based on low-temperature grown InGaAs/AlInAs multiple quantum wells [9] was a game changer, as these can be operated using rugged, compact and cost-effective rare-earth doped mode-locked fibre lasers. In subsequent years these antennas were significantly improved [10, 11] and now portable THz-TDS systems based on these antennas are available from several companies. In addition, numerous alternative techniques to access the THz band have been introduced [12–15] and

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many fixed wavelength CW sources and even planar detector arrays are offered commercially. In this article we will concentrate on results produced by the technique of THz-TDS, which were the main focus of the original article in 1999.

With the instrumental improvements, it is not surprising that the applications have also evolved. Terahertz radiation is now being used in fields as diverse as the study of fundamental properties of materials [16] to the inspection of the state of conservation of artwork [17, 18]. We aim to survey some of these exciting ideas, providing examples of how terahertz imaging is having impact in many areas, including both some that were anticipated in 1999 and some that were not.

## 2 T-ray imaging

Our original article focused on the, at that time, newly developed method for generating images using terahertz time-domain techniques. It included a discussion of the physics and engineering considerations required to construct and operate a time-domain spectrometer. These ideas have by now been reviewed many times [19, 20], and the systems have become commercially available from numerous vendors. So, it is probably not necessary to discuss those issues in great detail here, except perhaps to mention a few of the most exciting improvements that have been reported in the intervening years. The bandwidth of pulses, initially limited to only a few THz, can now be extended to over 100 THz using the shortest laser pulses and free-space electro-optic sampling [21]. The peak electric field strength generated in a TDS system, initially limited to only a few volts per cm, can now go into the tens of megavolts/cm range [22]. The average power, initially in the nanowatt range, can now reach tens of milliwatts [23]. These advances have enabled many scientific breakthroughs, although mostly not involving imaging.

Next, we note that, as terahertz technologies have matured, the number of options available to researchers for creating images using terahertz radiation has grown significantly. Of course, the raster-scanning technique described in 1999 is still in widespread use, as it offers a number of important advantages over other methods. Because it involves the acquisition of a complete time-domain waveform at each pixel of an image, it is in essence a measurement of a hyperspectral data cube, which can be rich in information about the sample under study. From this one data set, images can be formed based on either the amplitude or the phase of any of the measured frequency components, which typically span a wide bandwidth covering more than one decade in frequency. This capability has been exploited in many interesting examples. For instance, Kawase and co-workers demonstrated the identification of “white powders”

in envelopes, distinguishing harmless substances from illicit drugs using spectroscopic signatures that are manifested in the amplitude of the measured spectrum [24]. Meanwhile, the ability to directly measure the phase (and/or time delay) is critical to the implementation of tomographic techniques using THz pulses [25], as well as THz holography [26].

Despite these advantages, the time-domain imaging technique also has some important disadvantages. One key concern is the time required to acquire images: since a complete waveform is measured at each pixel, and this is generally accomplished using a single-point detector in a serial process, the image acquisition time is generally on the scale of minutes, or longer. This issue has inspired many efforts to develop alternative techniques that are faster. One hot topic is to leverage recent work in computational imaging, which enables the formation of images using only a subset of the full pixel set. Early efforts explored the use of compressed sensing to perform single-pixel imaging [27], which can reduce the measurement time substantially at the cost of increased computational effort [28]. Subsequent work has built on this idea in a variety of ways, including Fourier single-pixel imaging [29, 30] and ghost imaging [31], as well as demonstrations of real-time video-rate image acquisition [32].

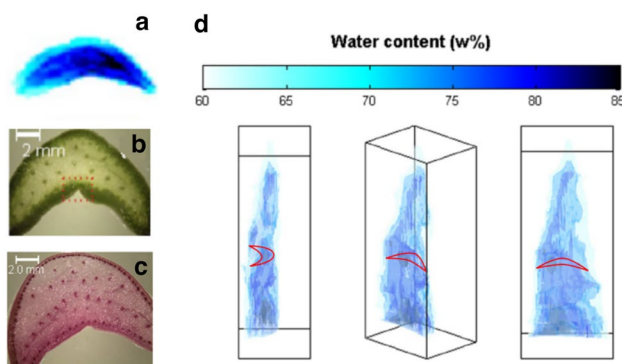
Another approach to faster image acquisition is the development of focal-plane arrays, which can be integrated into cameras for video-rate operation. The fabrication of a camera with sufficient sensitivity to be useful with a relatively low-power THz-TDS source is extremely challenging, so this approach most often leverages other types of THz source, typically with more power but much less spectral bandwidth. One early demonstration employed a micro-bolometer array camera together with a quantum cascade laser operating at 4.3 THz [33]. This report inspired many other efforts to design focal-plane arrays, using various material platforms and detection strategies [34–36]. Numerous THz cameras operating at video rate are now available on the commercial market [1].

These advances in imaging technologies have led to a commensurate growth in applications of terahertz imaging and sensing. As in 1999, it remains true that these ideas span a great variety of topics, from medical science to process monitoring to art conservation, ranging from very feasible [37, 38] to *highly* speculative. Some prominent recent successes in the realm of commercial applications include the monitoring of multi-layer paint coatings on automobiles [39, 40], the identification of defects in polymers and composites [41], and the characterization of chemical composition, porosity, and coating thickness in pharmaceutical tablets [42, 43].

One prominent possible application of THz imaging involves the contact-free mapping of the AC conductivity of materials or devices. After the early work of

Grischkowsky and co-workers demonstrated that THz-TDS can be used for electrical characterization of doped semiconductors [44], the first THz image mapping the conductivity of a doped wafer was reported in 1997 [45]. A more recent example described the mapping of the carrier mobility and chemical doping level in biased graphene layers [46, 47]. If the frequencies employed are well below the inverse carrier scattering time, as is often the case when broadband THz pulses are employed, the AC conductance obtained is directly comparable to the DC value. Hence, this application may be important for example, for the spatially resolved electrical characterization of solar cell or battery materials.

In addition to industrial applications, transmission imaging has provided a new tool in the area of plant science. The ability to image the water distribution within the plant has provided new and interesting information on the mechanisms to resist drought conditions of some desert plants such as Agaves. These plants produce a type of sugars called agave fructans, whose capacity to form large hydration shells [48] and therefore to retain water is remarkable. The 3-dimensional localization of these fructans obtained using terahertz imaging corresponds with the most hydrated parts of the plant, as seen in Fig. 1. Here, a 3-dimensional image and a 2-dimensional projection of the water content of the leaf are shown [49] together with visible images of cross sections of the leaf.



**Fig. 1** **a** Cross section water-content image of an agave leaf. The image shows a high hydration core surrounded by a lower hydration layer. **b** Microscope image of a cross-section of an agave leaf where the inner hydrenchyma (white) and the outer chlorenchyma (green) are visible and correlate with the image in **a**. **c** Periodic acid-Schiff (PAS) staining of the agave leaf revealing the presence of carbohydrates. **d** 3-dimensional water-content image of the entire agave leaf from 3 perspectives showing the outer low-hydration, and inner high hydration regions. Adapted from [49] licensed under CC-BY 4.0

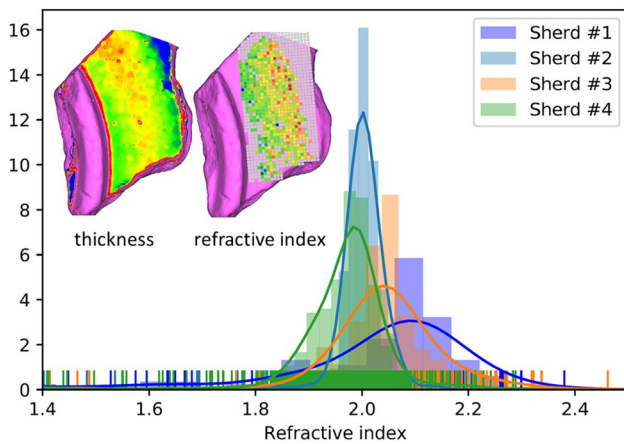
### 3 Reflection imaging with T-rays

Analogous to transmission imaging, a full time-dependent waveform is acquired for each pixel in reflection. So the data-set obtained in a reflection measurement is also a hyper-spectral cube, containing information about the composition of the sample under study. However, in many cases the useful information that can be extracted from these images is related not to spectroscopic composition but to the geometrical structure of heterogeneous samples, and particularly of layered media. When a sample has a complicated structure that incorporates interfaces within it, as long as all the materials involved are reasonably transparent, the contrast of their refractive indices will imply the existence of Fresnel reflections at each interface. Since the terahertz electromagnetic transients are rather short in time, owing to their small temporal coherence, their propagation through a sample with interfaces will lead to a series of reflected “copies” of the pulse separated in time. The time separation between them encodes the spatial separation between subsequent interfaces, therefore allowing the reconstruction of the internal structure of the sample in a similar fashion to B-scan ultrasonograms.

#### 3.1 Tomographic image reconstruction

The possibility of non-contact reconstruction of the internal geometry of complex layered material structures has attracted attention in many areas. In the earliest examples, the flatness of interfaces and prior knowledge about material composition led to relatively straightforward problem in image reconstruction [50]. Of course, when the sample under study has a complicated non-planar surface, a simple normal-incidence reflection configuration is no longer effective, and a more complicated approach is needed [8]. One interesting recent approach to overcome this challenge has been described by Stübbling et al. By placing a THz-TDS system at the end of a robotic arm, and extracting sample shape information from visible images or using structured light illumination, one can form THz reflection images of samples with arbitrary, even quite complex, shapes [51, 52]. This approach has been recently used in combination with photogrammetry to assess the age and composition of ancient pottery shreds, as shown in Fig. 2 [53]. Other groups have implemented more advanced signal processing approaches to tomographic image reconstruction, as discussed in Ref. [54].





**Fig. 2** A histogram of refractive index values at different locations on four pottery sherds from the Punic, Roman, and modern epochs. The ancient sherds were recovered from an archaeological site in Sardinia, Italy. The inset shows measured variations in thickness (left) and refractive index values (right) for one of the Roman samples. Adapted from [53]

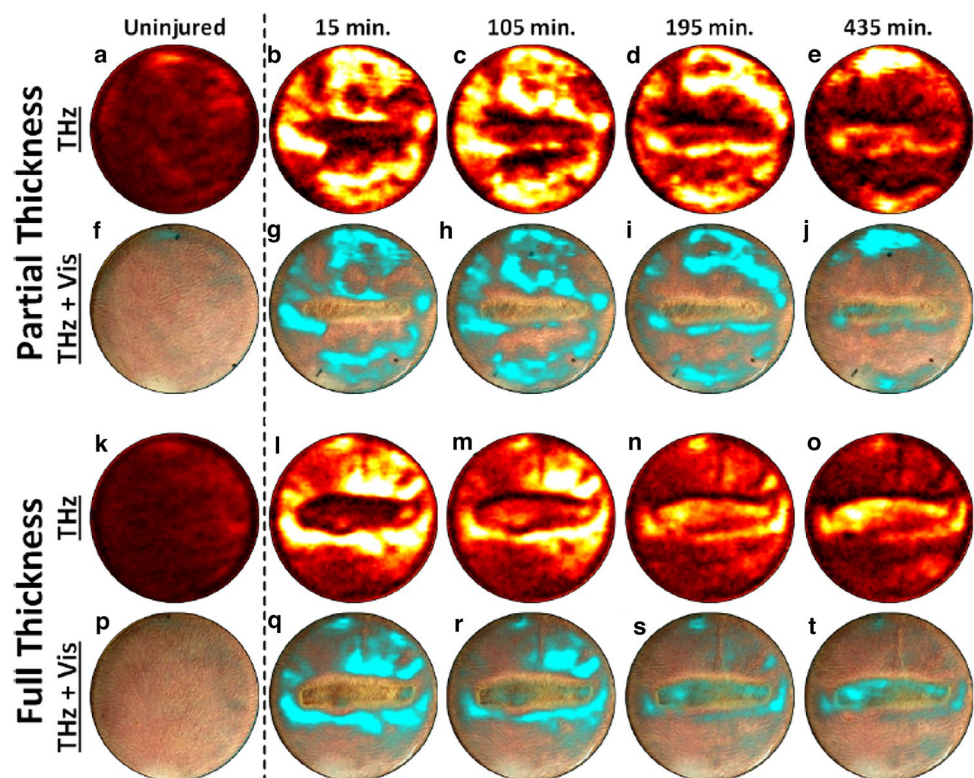
### 3.2 Medical imaging

Using terahertz as a probe in the medical area is an attractive option. For instance, terahertz radiation is non-ionizing, and therefore very safe to use as long as the average power is low enough to avoid producing significant heating of the tissues. Furthermore, it allows the water content

of tissues to be determined in a contactless fashion. A topic that was highlighted in the original article was the assessment of tissue burns by reflection geometry. A few preliminary measurements consisting of one-dimensional scans across a burnt section of chicken breast tissue were presented at that time. Since then, a wealth of progress has been made on that front, particularly over the last decade. Terahertz imaging has proven its potential for the discrimination of the depth of the burns, which is hard to assess clinically. Such studies have been done both *ex vivo* [55] and *in vivo* [56] on various animal models such as Sprague–Dawley rats [57] and Landrace Yorkshire cross pigs [59] whose skin have similarities with humans. Furthermore, differences in the post-burn water accumulation dynamics caused by edema have been well studied with terahertz methods [59, 60]. In such studies, an initial depletion of the hydration at the burn site, in the minutes following the injury, produces a decrease of the terahertz reflection at the cutaneous surface. However, the subsequent inflammatory response generates an accumulation of water over the following few hours, which increases the reflection. This effect is more pronounced in third degree “full thickness” burns than in first- or second-degree “partial thickness” burns as shown in Fig. 3.

A wealth of other applications of terahertz imaging in the medical field have been proposed over the last two decades. Of course skin is a type of tissue that is a natural candidate for terahertz inspection, since it is easily accessible. The

**Fig. 3** Burn wound time series imaging results. **a–e** THz images of a partial thickness burn. **f–j** Partial thickness THz images superimposed on the registered visible frame. **k–o** THz images of a full thickness burn. **p–t** Full thickness THz images superimposed on the registered visible frame. THz contrast is distinct for each burn wound severity. In time series THz images of a partial thickness burn, the contact area shows a drop in TWC and an edematous front superior to the burn over time. In contrast to the partial thickness wound the contact area of a full thickness burn does not display a significant drop in TWC. Additionally, the contact area is surrounded by a ring of TWC which runs concentric and coincident with the burn contact zone. [Reprinted] with permission from [60] ©The Optical Society



first *in vivo* measurements of skin [61] were reported in the early 2000s; however, the degree of sophistication required for actual medical applications has maintained interest in the topic up to the present day [62]. As one important example, diabetic foot syndrome, a consequence of diabetes, is a condition for which there is currently no objective diagnostic test, particularly in early stages. Many diabetic patients experience a combination of symptoms, that include the loss of sensation (neuropathy) related to a neurological deterioration in their lower limbs, poor irrigation (vasculopathy) and dehydration of the skin of their feet. In many cases this condition evolves into ulcers in the feet, and can result in partial or total amputations of the limbs. Indeed, this is the most common cause of amputations after traumatic accidents. Since skin dehydration accompanies this condition, terahertz imaging has been proposed as a potential tool for its objective and early diagnosis [63, 64], showing potential to become a valuable tool on this front. The most recent imaging study of diabetic foot [65] included a cohort of 178 human subjects (see Fig. 4), being to the best of our knowledge, the largest human population to ever be imaged with terahertz radiation for medical purposes. This work provides evidence that supports neurological deterioration as the underlying cause for dehydration, which is an open debate among clinicians.

Another tissue that is easily accessible without the need of endoscopes or surgery is the cornea. The possibility of non-contact monitoring of the corneal hydration has also attracted attention recently [66, 67]. The water content of the cornea is an important indicator of the ocular health [68],

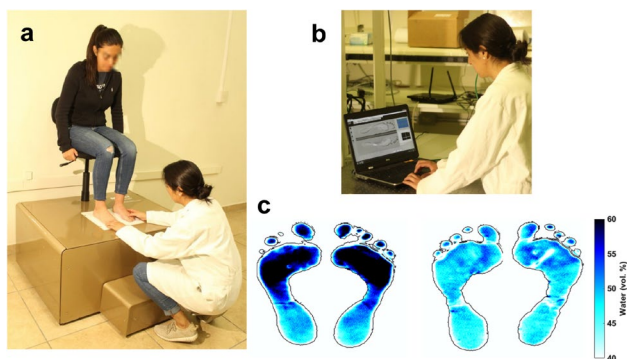
and could be used for the diagnosis and progression assessment of various ophthalmological conditions such as post-transplant corneal rejection, Fuchs dystrophy, keratoconus, trauma, infection, and other vision-impairing diseases [69].

Finally, we would be remiss if we did not mention that many efforts have been made to use terahertz imaging in oncology [70, 71]. Studies have focused on detection of breast [72], skin [73, 74], and colon [75–77] cancer, among other types [78–80] of cancer. In many of these studies, it has been demonstrated that terahertz imaging can indeed distinguish cancerous tissue from the surrounding healthy tissue. However, the exact mechanisms that lead to contrast in terahertz images is still a topic of debate [81], and this uncertainty has hindered widespread adoption of the technique in clinical settings.

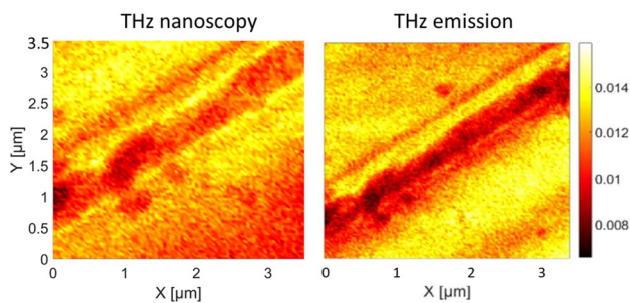
#### 4 THz near-field imaging

Another topic that was a focus of discussion in the original article related to the application of near-field techniques in the terahertz range. The 1999 paper displayed the very first near-field terahertz image, obtained using a sub-wavelength aperture, consisting of a conical tapered horn with an output opening of about 100  $\mu\text{m}$  diameter [82]. With a focus on the use of apertures for sub-wavelength confinement, the exciting developments in apertureless scattering-type scanning near-field microscopy (s-SNOM) taking place at almost the same time [83–85] were not mentioned at all. Despite some very impressive subsequent progress in aperture-based methods [86, 87], the s-SNOM approach, in which radiation is scattered from a sharply tapered metal tip held in close proximity to the sample under study, has become the dominant tool for nanoscale near-field spectroscopy and imaging in the terahertz range.

Early work in translating scattering-based near-field imaging to the terahertz range involved the use of home-built microscopy systems, with limited control and resolution [88, 89]. Nevertheless, these early efforts established some of the key distinctions between the initial studies at either optical, infrared, or microwave frequencies, and those involving terahertz time-domain spectrometers. These included effects related to the broad spectral bandwidth of the source such as antenna effects associated with the tapered metal tip [90], as well as the recognition that the tip itself can act as an effective and low-loss waveguide in this spectral range [91]. Unlike in other regimes of the spectrum, free-space electro-optic sampling of the broadband subpicosecond terahertz pulse enables direct measurement of the electric field in the near field of the tip apex [92], which can be used to develop a better understanding of the scattered far-field signals [93]. This work led rapidly to the demonstration of images with nanoscale resolution using terahertz waves [94, 95].



**Fig. 4** The use of terahertz imaging in medicine has implied adapting the conventional terahertz spectrometers to perform images of the human body. **a** A prototype of a foot scanner where the patient can sit while the terahertz spectral image is acquired by a time-domain spectroscopy setup mounted on an X–Y raster scanning stage underneath. **b** The raw image acquisition procedure. **c** The processed images of two patients, which have been converted into a water-content map. On the left are the feet of a diabetic with no complications, and on the right a diabetic showing nephropathy. The hydration levels observed in these two patients are remarkably different. The images are part of the data-set reported in [65]



**Fig. 5** Two images of a scratch on the surface of an InAs wafer, acquired using THz s-SNOM methods. The left image is formed by scattering an incident THz pulse from the AFM tip. The right image is formed by illuminating the tip with a near-infrared femtosecond pulse, and collecting the tip-modulated THz emission from the surface. The emission mechanism is the photo-Dember effect. Both images are collected by referencing the lock-in amplifier to the 2nd harmonic of the tip tapping frequency. Adapted from Ref. [105]

More recently, research in terahertz near-field science has surged, as a number of groups have reported exciting advances in the study of nanoscale materials and their fundamental excitations. Measurements of the tip-induced propagation of plasmons in single-layer graphene [96, 97] launched a tremendous wave of interest in the study of 2D materials using terahertz nanoscopy. Researchers have branched out to explore the possibilities offered by other terahertz sources, both pulsed and continuous-wave [98–100]. Meanwhile, the first report of time-resolved pump-probe experiments coupled to an apertureless s-SNOM [101] has inspired interest in studying terahertz photophysics with both femtosecond temporal resolution and nanometer-scale spatial resolution [102, 103]. Several other variations of these experiments have been reported. One notable example is nanoscale terahertz emission microscopy [104], in which an incident visible or near-infrared pulse induces THz emission in the sample, and this emission is coupled out to the far field by the AFM tip. This approach can be used to perform emission spectroscopy on single nanostructures. Because of the nonlinearity of the THz generation mechanism, this emission technique can offer improved resolution compared to the more common THz s-SNOM approach [105, 106]. Figure 5 shows a side-by-side comparison of a THz scattering image (linear optics) and a THz emission image (non-linear optics) on the same region of a sample.

Finally, this discussion would not be complete without also mentioning the use of terahertz pulses in conjunction with scanning tunneling microscopy (STM) [107]. An incident THz pulse can act as a transient bias across a tunneling junction [108], enabling STM measurements with sub-picosecond temporal resolution in addition to their intrinsic atomic-scale spatial resolution [109, 110]. A recent review article discusses these and other terahertz scanning probe techniques in great detail [111].

## 5 Conclusions

One can state that the field of terahertz science and technology has evolved considerably during the last 20 years. New THz sources such as QCLs or the TECSEL [112] and new detector arrays have been demonstrated. This can enable very fast and inexpensive THz imaging systems. Yet, also the specific field of THz imaging based on time-domain spectroscopy made considerable progress. On one hand many practical applications have been reported, some of which have been reviewed in this article. On the other hand the THz-TDS system itself has evolved. Fibre-coupled systems are widely available which introduces a great deal of flexibility. The fibre-coupled antennas can be easily translated enabling fast and flexible imaging configurations, as illustrated in Sect. 3.2. The antennas can also be attached to a robotic arm allowing for THz tomography of samples with arbitrary shape. Although the use of femtosecond fibre-lasers reduced the systems cost significantly, the price level of THz systems should further drop in the future to allow a wider market access. One way to achieve this could be the use of mode-locked semiconductor lasers as demonstrated recently [113]. Although 20 years ago one would have expected THz imaging systems to be more widely used in industry today, overall expectations have been met. Terahertz imaging is a valuable measurement tool that has opened a variety of applications in many fields.

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