# 17 Nanostructures in the Terahertz Range

John W. Bowen

**Abstract** With advances in technology, terahertz imaging and spectroscopy are beginning to move out of the laboratory and find applications in areas as diverse as security screening, medicine, art conservation and field archaeology. Nevertheless, there is still a need to improve upon the performance of existing terahertz systems to achieve greater compactness and robustness, enhanced spatial resolution, more rapid data acquisition times and operation at greater standoff distances. This chapter will review recent technological developments in this direction that make use of nanostructures in the generation, detection and manipulation of terahertz radiation. The chapter will also explain how terahertz spectroscopy can be used as a tool to characterize the ultrafast carrier dynamics of nanomaterials.

**Keywords** Terahertz • Nanostructures • Nanoscale sensing • Nanomaterial characterisation

# 17.1 Introduction

Compared to other regions of the electromagnetic spectrum, the terahertz range has only relatively recently become accessible for imaging and spectroscopy. This is due to the considerable technological challenges that have had to be overcome in order to generate, manipulate and detect terahertz radiation. A few years ago, this state of affairs gave rise to the notion of the "terahertz gap" in the spectrum between the infrared and microwave regions, generally considered to cover frequencies from 100 GHz to 10 THz, corresponding to wavelengths from 3 mm down to 30  $\mu$ m. However, while the field of terahertz science and technology is still in its infancy, the

J.W. Bowen (🖂)

School of Systems Engineering, University of Reading, Whiteknights, Reading, RG6 6AY, UK e-mail: j.bowen@reading.ac.uk

technology has advanced to such an extent that terahertz imaging and spectroscopy are now no longer confined to the laboratory, and are beginning to find realworld applications in a diverse range of areas. Many of these applications are a consequence of terahertz radiation being able to penetrate through materials which are otherwise opaque in other regions of the electromagnetic spectrum, combined with the ability to achieve three-dimensional and depth-resolved imaging, ultra-fast time resolution, identification of materials via molecular resonances, and improved safety over X-ray imaging due to the non-ionising nature of the radiation. As an example, the development of relatively portable terahertz imaging systems has enabled the first use of terahertz imaging in situ at an archaeological site [1]. This work was carried out in the remote, hot and dusty environment of the Turkish desert, to image Neolithic wall paintings obscured by covering layers of plaster. Other examples of in situ terahertz imaging of historic wall paintings and of applications in cultural heritage in general are given in [2] and [3]. There is also much interest in the use of terahertz imaging and spectroscopy for the detection of weapons, explosives or contraband concealed about the body or hidden in luggage [4], the detection of trace elements of substances [5], in medical imaging and diagnosis [6], and in a variety of industrial non-destructive testing applications [7]. However, despite this pioneering work, the portable systems available to date are still rather large and cumbersome, are relatively fragile and easily upset by extremes of temperature and moisture, and, depending on the spatial and spectral resolution required, can have rather slow data acquisition rates. Therefore, with these applications in mind, there is a need for terahertz systems with greater compactness and robustness, enhanced spatial resolution, more rapid data acquisition times and the ability to operate at greater standoff distances. This in turn requires the development of higher power terahertz sources, more responsive detectors and imaging arrays, all of which can be operated without the need for cryogenic cooling. This chapter will explore recent developments in this direction made possible by the use of nanostructures in the generation, manipulation and detection of terahertz radiation. Topics covered include the nanostructure enhancement of sources and detectors, terahertz nearfield microscopy, terahertz sensing of molecules and nanoparticles, and terahertz molecular imaging. In addition, it will be explained how terahertz radiation can be used as an important tool to characterize the ultrafast carrier dynamics of nanomaterials. As a precursor to the discussion of these new developments, the next section of this chapter gives an overview of the principles of operation of some of the terahertz imaging and spectroscopy systems in current use.

# 17.2 Terahertz Systems

There are several techniques available to carry out terahertz imaging and spectroscopy. As this chapter will discuss the application of nanostructures to extend and improve the performance of some of these techniques, as well as discussing the use of terahertz spectroscopy to characterize nanomaterials, this section will give a brief overview of the principles behind their operation.

### 17.2.1 Terahertz Time Domain Spectroscopy and Imaging

Terahertz time domain spectroscopy (TDS) is currently the most widely used laboratory based terahertz spectroscopy technique, and is also employed in the portable imaging system used for the work on wall paintings reported in [1] and mentioned above. TDS is based upon generating short, typically several hundred femtosecond long, pulses of broadband terahertz radiation and recording the resulting reflected or transmitted pulses from a sample or object of interest. The short pulse length makes it possible to resolve reflections due to interfaces between different layers within a stratified object, as these will appear as individual pulse reflection signatures occurring after different time delays within the recorded time domain signal, corresponding to the time of flight to each layer. This depth resolution can be further improved by using a deconvolution technique to achieve sub-pulse length and sub-wavelength depth resolution [8]. Moreover, the incident terahertz pulses are so short that they contain a wide range of frequencies, spanning at least 100 GHz to 3 THz (wavelengths of 3 mm down to  $100 \,\mu$ m) in typical systems. Therefore, broadband spectroscopy is possible by carrying out a fast Fourier transform of the time domain signals. As the technique is coherent, the resulting spectra contain both amplitude and phase information, enabling direct recovery of both the refractive index and absorption coefficient of a sample, without recourse to the Kramers-Kronig relations.

The TDS technique can be extended to enable imaging by focusing the terahertz beam to a small spot through which the sample can be raster scanned, or, if more convenient, in fibre coupled systems the beam can be raster scanned across the sample. The scanning is usually accomplished by mounting either the sample or the generator and detector heads on translation stages.

As there is a complete time domain signal behind each pixel in the resulting image, there are multiple parameters that can be used to construct an image, each of which can provide complementary information about the object being imaged. Typical imaging parameters include the peak amplitude in the time domain, the time delay, the integrated amplitude over a given range of frequencies, and the impulse response function [8].

The terahertz pulses are generated using a near-infrared femtosecond pulsed laser. Although the output pulses from the laser are short enough to have a bandwidth of several terahertz, their centre frequency is in the infrared range, and so a terahertz emitter has to be used to convert the output into the terahertz range. There are two main approaches to achieving this conversion. In photoconductive generation, the beam from the femtosecond laser generates photocarriers in a piece of semiconductor spanning the feed gap of a terahertz planar antenna. As a bias voltage is applied to the two halves of the antenna, a photocurrent is generated through the antenna for the duration of the incident infrared pulse and persists for the carrier lifetime, resulting in a short pulse of terahertz radiation being emitted from the antenna. In the second approach, known as optoelectronic generation, the terahertz pulses are generated by difference frequency mixing of the frequency components in the infrared pulse in a non-linear crystal with a non-zero second order susceptibility.

Detection of the terahertz pulses is achieved using a gated detection system. Again there are two main approaches. In photoconductive detection, the terahertz pulse is incident on an antenna structure similar to that used for photoconductive generation. This time, however, the antenna is not connected to a bias supply but is connected to equipment to record the current induced by the incident terahertz pulse. As current only flows when photocarriers are produced in the semiconductor due to an infrared pulse from the femtosecond laser being incident during the time of arrival of the terahertz pulse, the detector is gated by the femtosecond laser. In order to achieve this, a beam splitter is used to separate the output from the laser into pump and probe pulses to excite the generator and gate the detector respectively. Because the carrier lifetime results in the terahertz pulse being significantly longer than the infrared pulse, the time delay between the terahertz pulse and the infrared pulse can be scanned using an optical delay line so that the time domain waveform of the terahertz pulse can be mapped out over a series of successive output pulses from the femtosecond laser.

In the second approach, electro-optic detection, the terahertz pulse and the infrared gating probe pulse are incident on a crystal that displays the linear Pockel's effect. If no terahertz field were present, the incident linearly polarized infrared beam would have its polarization converted by the crystal, so that it would become circularly polarized. However, due to the Pockel's effect, the incident terahertz field results in an elliptically polarized output infrared beam. A Wollaston prism and pair of photodetectors are used to analyse the polarization of the infrared beam into linearly polarized components, the difference in intensity of which is directly proportional to the terahertz field amplitude. Again, both the terahertz pulse and infrared pulse need to be incident simultaneously for an output signal to be recorded, and so the terahertz signal can be recorded in the time domain by scanning the delay of the infrared probe pulse.

In portable systems, both the pump and probe pulses can be delivered to the emitter and detector using fibre optical umbilicals, allowing a high degree of flexibility for scanning different objects in situ.

#### 17.2.2 Photomixer Systems

An alternative technique that is beginning to be employed in commercially available terahertz spectroscopy systems is based around mixing together the output from two visible or infrared lasers to generate terahertz radiation at the difference frequency. The mixing is achieved within a semiconductor, usually GaAs, in which the intensity modulation of the laser beams due to their interference results in carrier generation with a terahertz periodicity. Often these laser beams are focused to a small spot in the semiconductor, and the resulting carrier currents feed a terahertz planar antenna

integrated on its surface. Typically, photomixers are pumped by distributed feedback diode lasers operating at a wavelength around 1,550 nm, which can be amplified by erbium doped fibre amplifiers. This approach has resulted in continuous wave terahertz outputs of between 20 mW at 100 GHz and 25  $\mu$ W at 0.914 THz. The output frequency can be swept over a wide range by sweeping the wavelength of the diode lasers, so that spectroscopy can be accomplished and depth information can be retrieved using frequency modulated continuous wave (FMCW) techniques. Again, imaging is possible by raster scanning. The resulting systems are very compact and portable. An in-depth review of terahertz photomixing is given in [9].

### 17.2.3 Quasi-optics

In order to keep terahertz systems as compact as possible, and bearing in mind that the wavelengths involved are a few millimeters to fractions of a millimetre, the terahertz beams produced by these systems are typically only a few wavelengths or tens of wavelengths across. As a consequence, just as with nano-optics at visible wavelengths, diffraction becomes a significant aspect of the propagation and similar techniques to those used to analyse nano-optics can be applied to terahertz optics, albeit scaled up in wavelength. Moreover, again for reasons of compactness, any lenses or mirrors used to control the spread of terahertz radiation tend to be used in the near to far-field transition region of the beam and have to be designed to take this into account. As a more general treatment than geometrical optics is needed to describe the behavior of such optical systems, they are generally referred to as being quasi-optical.

One convenient way to describe an arbitrary diffractively spreading paraxial beam is as a superposition of a set of beam-modes, each of which maintains a characteristic form as it propagates, albeit with a scaling width and changing phase-front curvature. These are usually Hermite-Gaussian or Laguerre-Gaussian beam-modes, each having a Gaussian transverse amplitude distribution modulated by either Hermite or Laguerre polynomials of different order. These beam-modes provide an analytical basis for modelling the propagation of a beam away from its narrowest point, termed the beam-waist, and enable the design of lens and mirror profiles to refocus the beam to a new beam-waist at a set distance. A full discussion of quasi-optics is given in [10].

The diffractive spreading imposes a number of size constraints on terahertz optical systems. Firstly there is a maximum distance from a lens at which a beamwaist may be formed, which depends purely on the width of the beam as it emerges from the lens and is independent of the lens focal length. Secondly, any apertures in a quasi-optical system have to have a diameter which is at least three times the 1/e amplitude half-width for a fundamental Gaussian beam-mode in order to avoid significant truncation. Truncation is undesirable, both because it would result in a loss of power and in the generation of higher order beam-modes, giving rise to dispersion. Finally, diffraction limits the smallest spot size to which a beam may be

focused to around one half of the wavelength, at which point the beam would be extremely non-paraxial. Although such extremely converging beams are not used in practice, the spot size in most imaging systems is typically a little larger than the wavelength and decreases in size with the wavelength across the system bandwidth. However, as will be discussed in Sect. 17.4, there are ways in which the radiation can be concentrated to much smaller dimensions, allowing nanoscale imaging and spectroscopy.

#### 17.3 Nanostructure Enhancement of Sources and Detectors

Recent work has shown that the incorporation of nanostructures in terahertz sources and detectors can offer considerable benefits in improving their efficiency. These advances will be discussed in this section.

## 17.3.1 Plasmonic Enhancement of Photoconductive Emitters and Detectors

One of the problems with both conventional photoconductive emitters and detectors and with photomixers is that it is difficult to obtain ultrafast and high quantum efficiency operation simultaneously. In a conventional emitter, the gap between the antenna electrodes cannot be smaller than the diffraction limited spot size of the pump laser, typically of the order of  $2 \mu m$ . Given the carrier drift velocity, only a small number of the photocarriers can reach the antenna electrodes on a subpicosecond timescale and so contribute to the terahertz radiation. The remaining carriers just result in a dc current, which lowers the bias field, in turn reducing the carrier acceleration between the electrodes. In order to suppress this unwanted dc current, most conventional emitters use short carrier lifetime semiconductors to ensure that carriers which are slow to reach the electrodes recombine before they get there. However, this means that only a small proportion of the photoexcited carriers contribute to the terahertz emission, limiting the efficiency.

Berry and Jarrahi [11] have shown how it is possible to overcome this problem by incorporating nanoscale plasmonic gratings into the emitter electrodes, eliminating the need for a short carrier lifetime semiconductor. Even though the gaps within the plasmonic gratings are very much smaller than the diffraction limited laser spot, the intensely localized surface plasmon fields generate carriers in the close vicinity of the gratings, enabling the carriers to quickly contribute to the current within the metal grating/antenna electrode structure. A very thin photoabsorbing semiconductor layer is used to ensure that carriers are not generated deeper within the semiconductor, as they would not reach the grating electrodes on a sufficiently short timescale. While electrons generated immediately under the positively biased grating will be drawn towards that grating and holes immediately under the negatively biased grating are drawn towards that grating to contribute to the terahertz emission, the carriers of opposite sign need to be collected by a grounded central contact between the two electrodes. Using this scheme, carriers that would take a relatively long time to reach the opposite electrode, and so contribute to an unwanted dc current, are removed, while around half of the electrons and holes that are generated contribute to the terahertz emission, which is a significant improvement on conventional emitters.

In addition, as the pump laser radiation can be absorbed over the entire plasmonic grating area rather than just the gap between the electrodes, the active area can be increased without significantly increasing the capacitance of the antenna. This, combined with the fact that normal semiconductors have a higher thermal conductivity than those with a short carrier lifetime, means that higher pump power levels can be used.

As they have similar structures and are based on similar operating principles, the incorporation of plasmonic gratings within photoconductive detectors and antenna coupled photomixers can lead to similar improvements in quantum efficiency.

# 17.3.2 Terahertz Generation from Plasmonic Nanoparticle Arrays

An alternative to conventional terahertz generators may be provided by the illumination of nanostructured metal surfaces by femtosecond lasers. Different research teams have presented evidence for different mechanisms behind this phenomenon.

Kajikawa et al. [12] have demonstrated terahertz emission from surfaceimmobilized gold nanospheres. The emission was seen to have a quadratic amplitude dependence on the excitation field, suggesting it is a second-order nonlinear optical process giving rise to optical rectification.

On the other hand, Polyushkin et al. [13] have demonstrated a non-quadratic dependence in terahertz emission from metal nanostructures, including semicontinuous metal films and ordered arrays of nanoparticles. In this case, based on a model by Welsh et al. [14], they suggest that the mechanism is plasmon mediated multiphoton ionization and ponderomotive acceleration of electrons in the uneven surface plasmon evanescent field, giving rise to terahertz emission.

Greater understanding of these processes may eventually lead to the development of more efficient terahertz emitters and may even lead to the interesting possibility of using a single metal nanoparticle as a local terahertz emitter [13].

## 17.4 Terahertz Near-Field Microscopy

As discussed in Sect. 17.2.3, diffraction limits the smallest spot size to which a terahertz beam can be focused to around half a wavelength. However, a variety of techniques based around sub-wavelength apertures, probes, plasmonic parallel

plate waveguides, aperture-less near-field scattering microscopy and nanojunctions can dramatically increase the spatial resolution possible in terahertz imaging and spectroscopy systems, to the extent that nanometre-scale resolution is possible. This section gives an overview of these techniques.

#### 17.4.1 Imaging with Sub-wavelength Apertures and Probes

Imaging with sub-wavelength apertures and probes can be used to detect the evanescent terahertz fields in the vicinity of an object. The apertures or probes can be integrated with the terahertz detector to yield resolutions of a few micrometres [15, 16] and there is even a commercially available terahertz integrated microprobe range with a  $3 \,\mu m$  spatial resolution [17], which can easily be added to existing terahertz TDS systems. Images can be constructed by scanning the probe or aperture across the surface of the object.

Rather than using a physical aperture, it is also possible to tightly focus the probe pulse onto an electro-optic crystal to form a so-called dynamic aperture, as terahertz detection will only take place in the crystal within the confines of the probe pulse spot [18, 19].

#### 17.4.2 Plasmonic Focusing

An alternative terahertz microscopy approach is to concentrate terahertz radiation to sub-wavelength dimensions in tapered plasmonic parallel plate waveguides. Focusing down to  $10 \,\mu\text{m}$  by  $18 \,\mu\text{m}$  has been demonstrated at  $0.155 \,\text{THz}$  [20], although, theoretically, losses limit the smallest spot size to only  $100-250 \,\text{nm}$  [21].

#### 17.4.3 Aperture-Less Near-Field Scattering Techniques

Rather than integrating a detector with a scanning probe, it is possible to insert a sharply pointed probe tip into a terahertz beam and measure the scattered evanescent field as the tip is scanned across an object. With this technique, it is necessary to have some means to distinguish the scattered field from the incident field. In one approach, the tip is scanned near to the surface of an electro-optic detection crystal with the correct crystallographic orientation to detect just the scattered fields from the tip which are polarized normal to the crystal surface, while being insensitive to the incident terahertz radiation which is polarized parallel to the crystal surface [22,23]. Alternatively, a vibrating probe tip can be used with lock-in detection [24].

Huber et al. [25] have achieved very impressive results by using metalised atomic force microscopy tips as the scattering probe. Because of non-linearity in the field near the tip, they have been able to achieve background suppression of the incident radiation by detecting the higher harmonics of the scattered field using a Michelson interferometer. Using this technique with radiation from a 2.54 THz laser they have managed to produce terahertz images of single nano-transistors, differentiating regions of different carrier density, with a spatial resolution of 40 nm, which is 1/3,000 of the wavelength.

### 17.4.4 Oxide Nanojunctions

Ma et al. [26] have demonstrated terahertz generation and detection at 10 nm oxide nanojunctions, opening up the possibility of terahertz spectroscopy and control of individual nanoparticles and molecules. The nanojunctions are formed by writing nanowires at the interface between layers of LaAlO<sub>3</sub> and SrTiO<sub>3</sub> using an atomic force microscope tip. This writing can be accomplished reversibly, so the junctions can be formed or reformed at will, potentially enabling the object to be firstly located with the atomic force microscope and then a nanojunction formed around it for terahertz nano-spectroscopy. The terahertz fields are generated and detected by  $\chi^{(3)}$  processes under illumination from a femtosecond laser, and the same junction can potentially be used for both generation and detection at the nanoscale, four orders of magnitude smaller than the diffraction limit.

# 17.5 Terahertz Sensing of Molecules and Nanoparticles

Because molecules and nanoparticles are considerably smaller than the wavelength, their absorption cross-sections are quite small at terahertz frequencies, making it difficult to detect them unless they are present in large quantities. However, it has recently been demonstrated that their absorption cross sections can be enhanced by over three orders of magnitude by placing them across or inside nanoslot resonators. This yields a dramatically increased absorption coefficient, allowing much smaller quantities to be detected.

Using a slot with a length of 90  $\mu$ m and width of 50 nm, formed in a 50 nm gold layer on a quartz substrate, Park et al. [27] have used TDS to demonstrate detection of as little as 40 ng of the explosive RDX drop-cast over an area of 10 mm<sup>2</sup>. This corresponds to only 22 fg of RDX within the slot. Without the nanoslot resonator, it is necessary to use of the order of 1 mg to see a noticeable difference in transmission through the RDX on the bare quartz substrate.

As the radiation is funneled through the nanoslot, the electric field becomes enhanced within the slot by orders of magnitude compared to the magnetic field. Because the absorption cross-section of the molecules is directly proportional to the wave impedance, which is the ratio of the electric to the magnetic field, the absorption cross-section increases accordingly. In the case of the RDX experiment mentioned above, the molecular absorption cross-section was enhanced by a factor of 2,800 times over that with the bare substrate.

The absorption enhancement can be tuned by changing the length of the slot so that it resonates at different frequencies, which can correspond to specified absorption bands in the target molecules. Park et al. [27] have demonstrated this by carrying out a second experiment with a 150  $\mu$ m long slot, resonant at 0.5 THz, which shows a similar absorption cross-section enhancement, but for lactose molecules rather than RDX.

The same group have also shown that similar nanoslot resonators are sensitive to even single nanorods placed across them, the nanorod changing the resonant conditions of the slot, depending its size and placement [28, 29].

### 17.6 Terahertz Molecular Imaging

A number of studies have indicated that conventional terahertz imaging may be a useful medical diagnostic tool, particularly for skin cancer [30] or tumours near the surface of organs [31]. However, terahertz molecular imaging (TMI) [32], which uses nanoparticle probes as contrast agents, offers enhanced performance by increasing contrast and enabling micrometre spatial resolution. Additionally, it offers the scope to track nanoparticle drug delivery.

The mechanism by which TMI operates is as follows. After injection into the biological tissue to be imaged, the nanoparticle probes target cancer cells by antibody phase conjugation, whereupon they become absorbed inside the cell by a process known as endocytosis. The tissue is illuminated with a chopped near-infrared laser beam which gives rise to surface plasmon polaritons on the nanoparticles, inducing a water temperature increase in the cells. As the refractive index of water in the terahertz range is temperature dependent, this results in a terahertz reflection change which can be detected using terahertz time domain imaging. Modulating the near-infrared laser enables differential detection, which is necessary to achieve the required sensitivity.

The TMI technique has been compared with magnetic resonance imaging (MRI) by transfecting superparamegnetic iron oxide (SPIO) nanoparticles into ovarian cancer cells in a mouse. Good correlation was seen between the TMI and MRI images, implying that TMI may be a useful technique in the operating theatre to complement pre- and post-operative MRI [32].

#### 17.7 Terahertz Characterisation of Nanomaterials

Terahertz spectroscopy is an important probe of the ultrafast carrier dynamics of nanomaterials because it provides information about charge transport on subpicosecond to nanosecond timescales and nanometre length scales which cannot be provided by other techniques. Terahertz frequencies closely match typical carrier scattering rates of  $10^{12}$ – $10^{14}$  s, making terahertz spectroscopy ideal for studying conductivity.

To give ultrafast time resolution, a modification of the TDS technique, known as time-resolved terahertz spectroscopy (TRTS) or optical pump/terahertz probe (OPTP) spectroscopy is used [33]. In this implementation, as well as providing the pulses to drive the terahertz generator and detector, the femtosecond laser provides another near-infrared pump beam to induce transient photoconductivity in the sample. A further optical delay line is included so that the terahertz behavior of the sample can be monitored at different times after photoexcitation of the sample.

The relaxation dynamics of the sample's transient photoconductivity can be probed by carrying out a "pump scan", wherein the delay in the pump beam is scanned while holding the terahertz detector gating delay at the peak of the terahertz time-domain waveform. The photoconductive lifetime can be determined from this type of measurement. Example measurements of the photoconductivity decay of GaAs, InAs and InP nanowires are given in Joyce et al. [34].

Conductivity can be determined by carrying out a "pump scan", wherein the pump beam delay is held constant while the terahertz gating delay is scanned to record the full terahertz waveform at a fixed time after photoexcitation. After fast Fourier transformation to the frequency domain and normalizing to a reference spectrum without the sample, the frequency dependent absorption coefficient and refractive index n of the sample can be found from the transmission amplitude and phase. In order to determine these quantities for nanostructures rather than bulk media, it may be necessary to make use of an effective medium approximation [35]. The frequency dependent complex conductivity can then be found from  $\alpha$ and n, although in doing so, it is usually necessary to include a component of the permittivity representing the contribution from bound modes, i.e. lattice vibrations and core electrons [36]. Once the conductivity has been determined experimentally, it can be fitted with an appropriate conductivity model (of which there are a number) to determine parameters such as the mobility, carrier scattering time and plasma frequency. A detailed overview of the whole procedure is given in [36]. Examples of terahertz characterization of a variety of nanostructures can be found in [35, 37-39].

#### 17.8 Conclusions

This chapter has shown how, despite the relatively large size of the wavelengths involved, nanostructures are beginning to make a significant impact in improving the performance of terahertz systems and devices applicable to a wide range of situations. In the case of terahertz sources and detectors excited by femtosecond lasers operating in the near-infrared, the terahertz performance improvements are the result of nanostructure enhancements of the interaction of the near-infrared laser radiation with the device. Recent developments in terahertz near-field microscopy mean that it is now possible to achieve nanoscale spatial resolution at terahertz frequencies. The use of nanoslot resonators can dramatically increase the terahertz absorption cross-section of molecules to allow the detection of small quantities of target substances, as well as providing a means to sense individual nanoparticles. Terahertz molecular imaging makes use of nanoparticle contrast agents to enhance the contrast and improve the spatial resolution of terahertz medical imaging for cancer diagnosis and nanoparticle drug delivery. Furthermore, terahertz spectroscopy provides unique information to characterize the ultrafast behavior of nanomaterials.

In addition to the new developments that have been covered in this chapter, work is underway on terahertz detection and generation via plasma waves in nanometre field effect transistors [40], on nano-hot electron bolometers [41], plasmonic and graphene based terahertz modulators [42, 43], terahertz polarisers based on carbon nanotudes and nanowire arrays [44, 45], terahertz vacuum nanoelectronics [46], graphene lasers [47], terahertz-driven molecular surface rotors for molecular machines [48], and wireless nanosensor networks for nanorobots, nanodiagnostic techniques and co-operative drug delivery systems [49]. It is clear that nanostructures are likely to play an increasing role in the future development of terahertz systems for many real-world applications.

### References

- Walker GC, Bowen JW, Matthews W, Roychowdhury S, Labaune J, Mourou G, Menu M, Hodder I, Jackson JB (2013) Sub-surface terahertz imaging through uneven surfaces: visualizing Neolithic wall paintings in Çatalhöyük. Opt Express 21:8126–8134
- Jackson JB, Bowen J, Walker G, Labaune J, Mourou G, Menu M, Fukunaga K (2011) A Survey of terahertz applications in cultural heritage conservation science. IEEE Trans Terahertz Sci Technol 1:220–231
- 3. Walker GC, Jackson JB, Giovannacci D, Bowen JW, Delandes B, Labaune J, Mourou G, Menu M, Detalle V (2013) Terahertz analysis of stratified wall plaster at buildings of cultural importance across Europe. In: Pezzati L, Targowski P (eds) *Optics for Arts, Architecture and Archaeology IV*, Proc. SPIE vol. 8790. Munich, Germany, p 87900H
- Kemp MC (2011) Explosives detection by terahertz spectroscopy-a bridge too far? IEEE Trans Terahertz Sci Technol 1:282–292
- Brown E, Khromova T, Globus T, Woolard D, Jensen J, Majewski A (2006) Terahertz-regime attenuation signatures in bacillus subtilis and a model based on surface polariton effects. IEEE Sens J 6:1076–1083
- 6. Pickwell E, Wallace VP (2006) Biomedical applications of terahertz technology. J Phys D: Appl Phys 39:R301–R310
- 7. Morita Y (2005) Terahertz technique for detection of microleaks in the seal of flexible plastic packages. Opt Eng 44:019001
- Walker GC, Bowen JW, Labaune J, Jackson J-B, Hadjiloucas S, Roberts J, Mourou G, Menu M (2012) Terahertz deconvolution. Opt Express 20:27230–27241

- 9. Preu S, Döhler GH, Malzer S, Wang LJ, Gossard AC (2011) Tunable, continuous-wave Terahertz photomixer sources and applications. J Appl Phys 109:061301
- 10. Martin D, Bowen J (1993) Long-wave optics. IEEE Trans Microw Theory Tech 41(10):1676–1690
- Berry CW, Jarrahi M (2012) Terahertz generation using plasmonic photoconductive gratings. New J Phys 14:105029
- Kajikawa K, Nagai Y, Uchiho Y, Ramakrishnan G, Kumar N, Ramanandan GKP, Planken PCM (2012) Terahertz emission from surface-immobilized gold nanospheres. Optics Lett 37:4053–4055
- Polyushkin DK, Hendry E, Stone EK, Barnes WL (2011) THz generation from plasmonic nanoparticle arrays. Nano Lett 11:4718–4724
- Welsh G, Hunt N, Wynne K (2007) Terahertz-pulse emission through laser excitation of surface plasmons in a metal grating. Phys Rev Lett 98:026803
- Mitrofanov O, Lee M, Hsu J, Brener I, Harel R, Federici J, Wynn J, Pfeiffer L, West K (2001) Collection-mode near-field imaging with 0.5-THz pulses. IEEE J Sel Top Quantum Electron 7(4):600–607
- Kawano Y (2011) Highly sensitive detector for on-chip near-field thz imaging. IEEE J Sel Top Quantum Electron 17:67–78
- 17. TeraSpike, AMO GmbH. Available at http://www.amo.de/thz\_tip.0.html
- Seo MA, Adam AJL, Kang JH, Lee JW, Ahn KJ, Park QH, Planken PCM, Kim DS (2008) Near field imaging of terahertz focusing onto rectangular apertures. Opt Express 16:20484
- Adam AJL, Brok JM, Seo MA, Ahn KJ, Kim DS, Kang JH, Park QH, Nagel M, Planken PC (2008) Advanced terahertz electric near-field measurements at sub-wavelength diameter metallic apertures. Opt Express 16:7407
- Zhan H, Mendis R (2010) Mittleman DM, Superfocusing terahertz waves below lambda/250 using plasmonic parallel-plate waveguides. Opt Express 18:9643–9650
- Rusina A, Durach M, Nelson KA, Stockman MI (2008) Nanoconcentration of terahertz radiation in plasmonic waveguides. Opt Express 16:18576
- van der Valk NCJ, Planken PCM (2004) Towards terahertz near-field microscopy. Philos Trans Ser A Math Phys Eng Sci 362:315–319; discussion 319–321
- Planken PCM, van Rijmenam CEWM, Schouten RN (2005) Opto-electronic pulsed THz systems. Semicond Sci Technol 20:S121–S127
- 24. Astley V, Zhan H, Mittleman D, Hao F, Nordlander P (2007) Plasmon-enhanced Terahertz near-field microscopy – OSA technical digest series (CD). In: Conference on lasers and electrooptics/quantum electronics and laser science conference and photonic applications systems technologies, Baltimore. Optical Society of America, p CTuJJ5
- Huber AJ, Keilmann F, Wittborn J, Aizpurua J, Hillenbrand R (2008) Terahertz near-field nanoscopy of mobile carriers in single semiconductor nanodevices. Nano Lett 8:3766–3770
- 26. Ma Y, Huang M, Ryu S, Bark CW, Eom C-B, Irvin P, Levy J (2013) Broadband terahertz generation and detection at 10 nm scale. Nano Lett 13:2884–2888
- Park H-R, Ahn KJ, Han S, Bahk Y-M, Park N, Kim D-S (2013) Colossal absorption of molecules inside single terahertz nanoantennas. Nano Lett 13:1782–1786
- 28. Park H-R, Bahk Y-M, Choe JH, Han S, Choi SS, Ahn KJ, Park N, Park Q-H, Kim D-S (2011) Terahertz pinch harmonics enabled by single nano rods. Opt Express 19:24775–24781
- Park H-R, Bahk Y-M, Ahn KJ, Park Q-H, Kim D-S, Martín-Moreno L, García-Vidal FJ, Bravo-Abad J (2011) Controlling terahertz radiation with nanoscale metal barriers embedded in nano slot antennas. ACS Nano 5:8340–8345
- 30. Woodward RM, Wallace VP, Pye RJ, Cole BE, Arnone DD, Linfield EH, Pepper M (2003) Terahertz pulse imaging of ex vivo basal cell carcinoma. J Investig Dermatol 120:72–78
- 31. Ashworth P, O'Kelly P, Purushotham A, Pinder S, Kontos M, Pepper M, Wallace V (2008) An intra-operative THz probe for use during the surgical removal of breast tumors. In: 2008 33rd international conference on infrared, millimeter and terahertz waves, Pasadena
- 32. Son J-H (2013) Principle and applications of terahertz molecular imaging. Nanotechnology 24:214001

- Beard MC, Turner GM, Schmuttenmaer CA (2002) Terahertz spectroscopy. J Phys Chem B 106:7146–7159
- 34. Joyce HJ, Docherty CJ, Gao Q, Tan HH, Jagadish C, Lloyd-Hughes J, Herz LM, Johnston MB (2013) Electronic properties of GaAs, InAs and InP nanowires studied by terahertz spectroscopy. Nanotechnology 24:214006
- 35. Baxter JB, Schmuttenmaer CA (2006) Conductivity of ZnO nanowires, nanoparticles, and thin films using time-resolved terahertz spectroscopy. J Phys Chem B 110:25229–25239
- Lloyd-Hughes J, Jeon T-I (2012) A review of the terahertz conductivity of bulk and nanomaterials. J Infrared Millim Terahertz Waves 33:871–925
- 37. Jung GB, Myung Y, Cho YJ, Sohn YJ, Jang DM, Kim HS, Lee C-W, Park J, Maeng I, Son J-H, Kang C (2010) Terahertz spectroscopy of nanocrystal-carbon nanotube and -graphene oxide hybrid nanostructures. J Phys Chem C 114:11258–11265
- Parkinson P, Joyce HJ, Gao Q, Tan HH, Zhang X, Zou J, Jagadish C, Herz LM, Johnston MB (2009) Carrier lifetime and mobility enhancement in nearly defect-free core-shell nanowires measured using time-resolved terahertz spectroscopy. Nano Lett 9:3349–3353
- 39. Strait JH, George PA, Levendorf M, Blood-Forsythe M, Rana F, Park J (2009) Measurements of the carrier dynamics and terahertz response of oriented germanium nanowires using opticalpump terahertz-probe spectroscopy. Nano Lett 9:2967–2972
- 40. Shur M (2010) Plasma wave terahertz electronics. Electron Lett 46:s18-s21
- Karasik BS, Sergeev AV, Prober DE (2011) Nanobolometers for THz photon detection. IEEE Trans Terahertz Sci Technol 1:97–111
- 42. Vasić B, Gajić R (2012) Broadband and subwavelength terahertz modulators using tunable plasmonic crystals with semiconductor rods. J Phys D Appl Phys 45:095101
- 43. Weis P, Garcia-Pomar JL, Höh M, Reinhard B, Brodyanski A, Rahm M (2012) Spectrally wideband terahertz wave modulator based on optically tuned graphene. ACS Nano 6:9118–9124
- 44. Ren L, Pint CL, Arikawa T, Takeya K, Kawayama I, Tonouchi M, Hauge RH, Kono J (2012) Broadband terahertz polarizers with ideal performance based on aligned carbon nanotube stacks. Nano Lett 12:787–790
- Cartwright AN (2011) Wavelength-independent optical polarizer based on metallic nanowire arrays. IEEE Photon J 3:1083–1092
- 46. Jang K-H, Park SH, Lee K, Park G-S, Jeong YU (2012) High-order photonic bandgap reflex klystron using carbon nanotube multi-beam cathode. Electron Lett 48:707
- 47. Otsuji T, Boubanga Tombet SA, Satou A, Fukidome H, Suemitsu M, Sano E, Popov V, Ryzhii M, Ryzhii V (2012) Graphene-based devices in terahertz science and technology. J Phys D Appl Phys 45:303001
- Neumann J, Gottschalk KE, Astumian RD (2012) Driving and controlling molecular surface rotors with a terahertz electric field. ACS Nano 6:5242–8
- 49. Jornet JM, Akyildiz IF (2011) Channel modeling and capacity analysis for electromagnetic wireless nanonetworks in the terahertz band. IEEE Trans Wirel Commun 10:3211–3221