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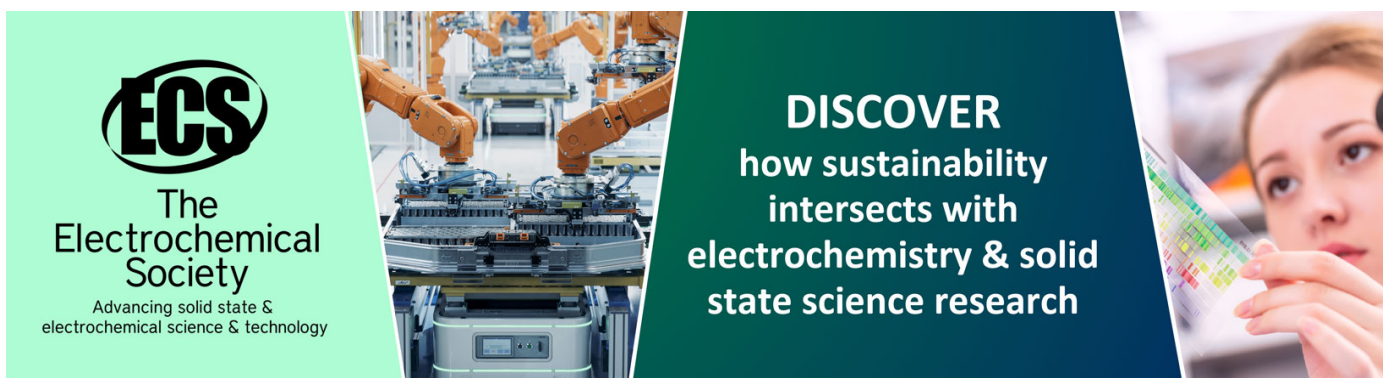
The 2023 terahertz science and technology roadmap

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Roadmap

The 2023 terahertz science and technology roadmap

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Abstract

Terahertz (THz) radiation encompasses a wide spectral range within the electromagnetic spectrum that extends from microwaves to the far infrared (100 GHz–~30 THz). Within its

frequency boundaries exist a broad variety of scientific disciplines that have presented, and continue to present, technical challenges to researchers. During the past 50 years, for instance, the demands of the scientific community have substantially evolved and with a need for advanced instrumentation to support radio astronomy, Earth observation, weather forecasting, security imaging, telecommunications, non-destructive device testing and much more. Furthermore, applications have required an emergence of technology from the laboratory environment to production-scale supply and in-the-field deployments ranging from harsh ground-based locations to deep space. In addressing these requirements, the research and development community has advanced related technology and bridged the transition between electronics and photonics that high frequency operation demands. The multidisciplinary nature of THz work was our stimulus for creating the 2017 THz Science and Technology Roadmap (Dhillon *et al* 2017 *J. Phys. D: Appl. Phys.* **50** 043001). As one might envisage, though, there remains much to explore both scientifically and technically and the field has continued to develop and expand rapidly. It is timely, therefore, to revise our previous roadmap and in this 2023 version we both provide an update on key developments in established technical areas that have important scientific and public benefit, and highlight new and emerging areas that show particular promise. The developments that we describe thus span from fundamental scientific research, such as THz astronomy and the emergent area of THz quantum optics, to highly applied and commercially and societally impactful subjects that include 6G THz communications, medical imaging, and climate monitoring and prediction. Our Roadmap vision draws upon the expertise and perspective of multiple international specialists that together provide an overview of past developments and the likely challenges facing the field of THz science and technology in future decades. The document is written in a form that is accessible to policy makers who wish to gain an overview of the current state of the THz art, and for the non-specialist and curious who wish to understand available technology and challenges. As such, our experts deliver a ‘snapshot’ introduction to the current status of the field and provide suggestions for exciting future technical development directions. Ultimately, we intend the Roadmap to portray the advantages and benefits of the THz domain and to stimulate further exploration of the field in support of scientific research and commercial realisation.

Keywords: terahertz, spectroscopy, photonics

(Some figures may appear in colour only in the online journal)

Contents

1. Introduction	
<i>John Cunningham and Michael B Johnston</i>	5
2. THz time-domain quantum optics	
<i>Alfred Leitenstorfer and Andrey S Moskalenko</i>	6
3. Terahertz spintronics and magnetism	
<i>Tobias Kampfrath and Junichiro Kono</i>	8
4. Terahertz time-domain polarimetry and ellipsometry	
<i>Kun Peng, Naser Qureshi and Enrique Castro-Camus</i>	11
5. Nonlinear terahertz spectroscopy	
<i>Dmitry Turchinovich and Koichiro Tanaka</i>	13
6. THz biological effects and sensing	
<i>A G Markelz, Martina Havenith and Cameron Hough</i>	15
7. Terahertz spectroscopy of emerging materials	
<i>Hannah J Joyce and Willie J Padilla</i>	18
8. THz air photonics	
<i>Binbin Zhou, Ki-Yong Kim, Xi-Cheng Zhang and Peter Uhd Jepsen</i>	21
9. Recent progress in terahertz intersubband devices	
<i>S S Dhillon, M S Vitiello, E H Linfield and A G Davies</i>	23
10. Laser-based terahertz sources	
<i>Matthias C Hoffmann and Roger Lewis</i>	26
11. Terahertz emission spectroscopy and imaging	
<i>Masayoshi Tonouchi, Tom S Seifert and Pernille Klarskov</i>	29
12. Terahertz scanning tunnelling microscopy	
<i>Yaroslav A Gerasimenko, Dragan Mihailovic and Rupert Huber</i>	32
13. Near field THz imaging	
<i>Jessica Boland, Oleg Mitrofanov, Pernille Klarskov and Paul Dean</i>	35
14. Terahertz instrumentation for satellites	
<i>Brian N Ellison, Peter G Huggard and Simon P Rea</i>	38
15. Status of THz astronomy	
<i>C K Walker, D Leisawitz and J R Gao</i>	41
16. Multipixel THz imaging	
<i>Chong Li, Gintaras Valušis and Qin Chen</i>	45
17. Medical applications	
<i>Vincent P Wallace and Emma Pickwell-MacPherson</i>	48
18. Waveguide-based THz standards and metrology	
<i>Xiaobang Shang, Jeffrey Hesler and Nick Ridler</i>	50
19. Terahertz communication	
<i>Cyril C Renaud, Ingmar Kallfass and Tadao Nagatsuma</i>	53
20. THz technology in industry	
<i>J Axel Zeitler and Don Arnone</i>	55
Acknowledgments	57
Data availability statement	58
References	58

1. Introduction

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Terahertz (THz) radiation, which in the last century was also referred to as the sub-millimetre wavelength region of the electromagnetic spectrum, or as just as ‘far-infrared’ radiation, is typically defined as electromagnetic waves with frequencies between 100 GHz and 30 THz (wavelengths 3 mm–10 μm). While electronic methods of generation and detection are typically associated with microwaves at the lower-frequency end of the THz band, optical and photonic methods are used for infrared light at the upper boundary. The THz band is where the fields of electronics and optics meet up, and this convergence has required the expertise not just of electrical engineers, but also photonic engineers, physicists, as well as material scientists. Taking one example, the development of technologies to modulate THz light has involved a range of disciplines, requiring knowledge of free-space optics, light-matter interactions, and carrier transport from condensed matter physics. Often new, adapted or highly engineered materials are required, so an underpinning of materials science has also proved necessary. Furthermore, the technical developments arrived at have enabled new THz spectroscopy, imaging and microscopy methods that have drawn in an even wider range of scientists, including biologists and chemists. Beyond scientific discovery, THz technologies are now entering the realm of ‘real world’ applications ranging from weather monitoring and forecasting, communications, security scanning, and diagnostics for faults in semiconductor integrated circuits, to monitoring thickness of paint layers in both automotive and aerospace applications. A running theme is that THz science is not seeking to find one single ‘killer application’, rather as has been demonstrated in other spectral ranges, it has now shown utility in a host of worthwhile areas, each with its own drivers that will ultimately determine commercial success or failure. We may reasonably surmise the prospect of finding new areas with potential for commercial exploitation is strong, and set to drive further developments over the next decade and beyond; it is likely many such developments will occur within the thematic areas that we have identified here, but we must also acknowledge that inevitably some of the most exciting developments will come from entirely new directions that as yet are unforeseen.

In 2017 we thought that the field of THz science and technology had expanded in interdisciplinary breadth and publication output to an extent that it was difficult for any individual to have a satisfactory overview of the status of the field. Thus we compiled the 2017 THz Science and Technology Roadmap [1], which aimed to produce a snapshot of the field and assess the many future challenges and opportunities. The exploration and exploitation of the THz region of the electromagnetic spectrum has continued unabated since. In this second edition, we concentrate on the substantial scientific developments that have been made in the intervening six years, on new applications and technologies at the very forefront of scientific developments, and look to the future of the many technological areas that are already benefitting from the continued worldwide research investment and interest. Developments across 19 topical areas are featured, including in quantum optics and quantum device designs, in lasers and modulators, magnetic systems, and covering applications as diverse as automotive, biological and medical, along with the nascent underpinning metrology.

There is a strong focus on imaging running through our Roadmap, a result of the very rapid advances being made in the various THz modalities, which now offer the ability to observe THz properties down to single-molecular length-scales, a potential game-changer in terms of understanding fast charge-carrier dynamics and their spatial dependence across a wide range of micro- and nano-scale systems, and which may be reasonably expected to lead eventually to new devices in which the passage, emission, modulation and detection of THz radiation is tailored for particular applications.

Elsewhere, developments in astronomy and satellite systems are discussed, along with the aligned topic of THz communications. As hardware underpinning 6G and subsequent wireless technologies becomes ever more important over the next decade telecommunications is poised to transition from the microwave to THz domain; a prospect recognised by the international governments and regulatory organisations such as the International Telecommunications Union.

We again anticipate our Roadmap will be of interest both to new and established workers in the disciplines and topics covered, including those that may have heard of THz science but who would like a succinct summary of the state-of-the-art. It may also that it might act as a pointer both for funding agencies and for governmental organisations interested in assessing and understanding the range of fledgling to well-established technologies related to the THz region of the electromagnetic spectrum.

2. THz time-domain quantum optics

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Status

A detailed understanding of the physics of vacuum fluctuations is of paramount importance for answering fundamental questions related to a broad spectrum of advanced topics ranging from quantum technologies all the way to the properties of the universe. The emerging field of THz time-domain quantum optics brings the established technology for generation and characterization of classical THz electric fields to this quantum regime. In conventional quantum optics, the quantum state of light is accessed in a time-averaged way via homodyne detection or photon correlation measurements. Instead, electro-optic sampling based on ultrashort laser pulses provides sensitive access to the coherent THz field amplitude directly in the time domain. Consequently, generalizing this technique to quantum fields became attractive. The first experimental implementation led to free-space detection of electric-field vacuum fluctuations in the multi-THz or mid-infrared range, supported by a theory based on the paraxial approximation of nonlinear quantum optics [2]. Exploiting a quantum Green's tensor based [3, 4] or a direct polaritonic approach [5] allows considering also effects of absorption and dispersion as well as deviations from the paraxial limit. The results of a first attempt [6] to go beyond quantum vacuum by generation and sampling of few-cycle synchronal squeezed states, i.e. states with quantum noise patterns synchronized to the probe pulses, are clarified in the next sections. The concept of this experiment is sketched in figure 1.

An expanded scheme, implemented first for subcycle measurements of intensity correlations from a THz quantum cascade laser close to the lasing threshold [7], is capable of resolving the temporal and spatial correlations of quantum fields [8], see figure 2. Furthermore, the changes of the THz quantum vacuum induced by a perfectly reflecting plate or by a plate possessing a surface plasmon polariton were studied in this configuration [9]. Options to sample not only the electric component of the propagating quantum field but also its second generalized conjugated quadrature [10, 11] have been discovered recently [12, 13], paving the way towards a new type of quantum tomography operating completely in the time domain. In general, these subcycle studies of quantum fields are exploring deep connections between quantum and relativistic physics [11, 14], thus creating a new platform for future advances in this frontier region of contemporary science.

Current and future challenges

In THz quantum detection, a statistical readout with millions of repetitions for each relative temporal position t_D or τ is

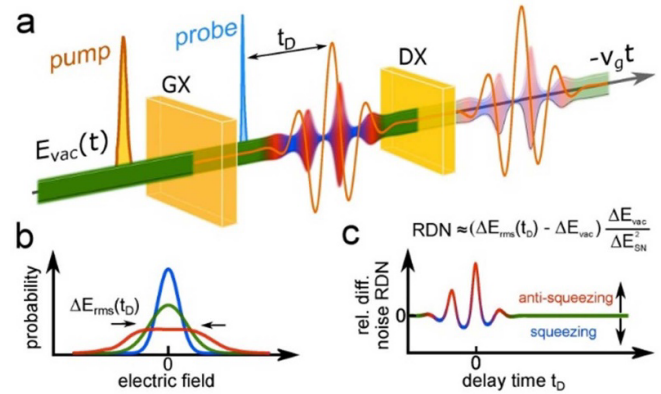


Figure 1. (a) Time-domain generation and sampling of THz quantum fields [2, 6]. A near-infrared (NIR) pump generates a THz coherent transient by optical rectification (orange) in a nonlinear crystal (GX). Cascaded action of the second-order nonlinearity modulates the refractive index seen by the co-propagating bare vacuum fluctuations $E_{vac}(t)$ (green band: rms amplitude), resulting in a synchronal squeezed field with increased (red) and decreased (blue) fluctuations with respect to $E_{vac}(t)$. This quantum waveform enters a detection crystal (DX) with a coherent NIR probe delayed in time by t_D . v_g : near-infrared group velocity. The polarization state of the probe encodes the amplitude of the THz field. It is analysed by an ellipsometer (see [2]). Reproduced from [6], with permission from Springer Nature. (b) Schematic probability distributions of the THz quantum field at three specific time delays t_D : bare vacuum (green line), squeezing (blue) and anti-squeezing (red). (c) Schematic of the relative differential noise (RDN) measured as a function of t_D . Detected variance also includes the shot noise of the NIR probe photons.

implemented to have access not only to the mean value of the field (coherent amplitude) which vanishes for the cases of bare and squeezed vacuum, but also to its full statistics (see figures 1(b) and 2). The key factors limiting the sensitivity are related to the shot noise at low fluxes of probe photons (weak measurement regime), followed by back-action and potentially also third-order nonlinear effects at higher probe intensities. Figuring out the optimum operational regimes and materials involved will be a challenge for the near future. Another challenge is an appropriate modification of the setup for simultaneous quadrature measurement, thus enabling subcycle-resolved quantum tomography. This step might require spectral filtering of the detected near-infrared (NIR) photons and variation of the static phase shift in the ellipsometer [12]. Generalization towards quantum imaging of THz fields with subcycle and subwavelength resolution [15] represents another milestone. Once such challenges have been met, the first quantum technology at far- and mid-infrared frequencies is available. While the photons in the coherent part of the THz fields which have been exploited so far are maximally uncorrelated with each other, equivalent to Poissonian statistics, quantum states of light with e.g. squeezed character comprise correlated photons which are e.g. coming in pairs. The ability to exploit this aspect of quantum light will be extremely attractive especially in the THz frequency range where the collective resonances of matter are located. For example, correlated two-photon absorption will provide access to different

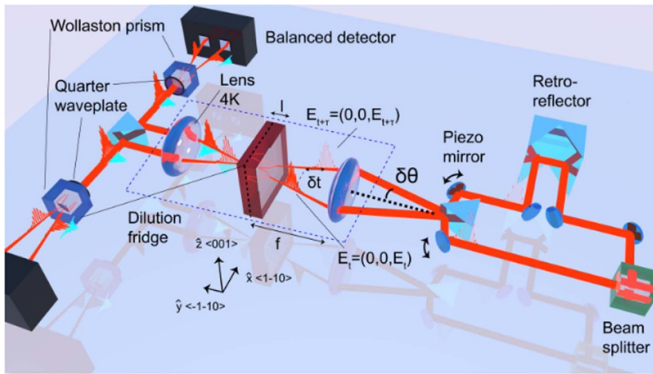


Figure 2. Two-probe setup for studying the correlations of the THz vacuum field [8]. At the entrance, a strong NIR probe is split into two beams (red bands) by a beam splitter. One of the beams acquires a time delay τ at a retroreflector. The beams are focused at an electro-optic crystal (EOX) of length l with a small difference $2\delta\theta$ in the angle of incidence. The beams may also have a spatial separation between their focus points at the EOX to access spatial correlations. After the interaction with the THz vacuum field in the EOX, the modified NIR beams are separated and directed towards two different ellipsometry setups, including quarter waveplates, Wollaston prisms and balanced detectors. The currents from the detectors are multiplied electronically and statistically analysed to obtain the correlation signal. To avoid thermal population of the photon states of the few-THz quantum fluctuations detected by this experiment, the interaction part of the setup is operated at cryogenic temperatures (Figure Courtesy of F F Settembrini and J Faist).

selection rules as compared to dipolar one-photon absorption while remaining in a linear regime. Potentially, vibrational or magnonic (i.e. spin-wave) transitions which are only Raman active for coherent light as well as elusive excitations of many-body states like superconducting condensates might become directly accessible by this quantum variant of infrared absorption without the severe restrictions in time resolution inherent to Raman spectroscopy.

Advances in science and technology to meet challenges

The most critical issues for advancing time-domain quantum optics are the stability of few-femtosecond laser sources [2, 6], operating sophisticated setups in cryogenic environments [8] and finding the right schemes of electronic readout for maximum efficiency of isolating the quantum aspects. For example, the changes of noise with t_D detected in [6] were heavily contaminated owing to an interplay of several spurious factors [12]: while the total photon statistics of the ultrabroadband probe pulses is almost perfectly Poissonian

in the radiofrequency range where lock-in quantum detection operates, anti-correlations in amplitude noise have been discovered between the high- and low-frequency spectral regions. Together with an immanent presence of the Hilbert transform of the THz field in the detected signals and minute deviations of the ellipsometer setup from optimum, positive and negative deviations from the noise level of bare vacuum input arise at the zero-crossings of the co-propagating coherent waveform. The route to avoid such artefacts is as follows: sources for more perfectly coherent pulses or even specially designed quantum states for probing [16] have to be deployed. This task might include in-depth studies of e.g. the properties of the femtosecond frequency comb structures of these sources, down to a resolution in the order of 100 Hz. At the same time, geometries for nonlinear generation of quantum noise patterns in time which are synchronal to those probes have to be worked out where the presence of a co-propagating coherent THz amplitude is maximally suppressed. This task may be accomplished e.g. by direct pumping of second-order orientation-patterned nonlinear crystals with intense and phaselocked THz transients from optical-parametric oscillators or by exploiting third-order nonlinear elements for direct conversion of NIR femtosecond pulses into THz quantum radiation. Finally, as in any quantum technology, strategies to avoid spurious losses e.g. due to reflections or absorption will minimize degradation of any quantum states due to the superposition of uncorrelated vacuum fluctuations. Potentially, the broad relative bandwidth and minimum dispersion required by the time-domain aspect will require advanced concepts such as e.g. moth's-eye nanostructures for antireflection coating.

Concluding remarks

THz time-domain quantum optics represents a new frontier, taking its place among other technologies contributing to the second quantum revolution. It might pave the way to time-domain tomography of quantum fields and further on to quantum spectroscopy [17] in this important spectral regime. It also provides a resource to address fundamental questions and to resolve puzzles in quantum physics, extending even to cosmological context. In particular, characterization of photon-antiphoton clouds at elementary timescales, observation of vacuum entanglement, direct detection of Unruh radiation and control of its properties may be envisioned. The technological challenges on this path are prominent but seem to be manageable. They include tasks like increasing detection sensitivity, eliminating spurious effects and finding advanced electro-optics materials. Finally, also new concepts might emerge with extended ranges of applications and efficiency.

3. Terahertz spintronics and magnetism

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Status

The central goal of terahertz (THz) spintronics is to interrogate and control spin dynamics in spintronic systems using THz electromagnetic pulses [18], as schematically summarized in figure 3. In particular, both THz electromagnetic transients and optical femtosecond laser pulses are used to push the rotation and transport of electron spins and the detection of spin dynamics from the sub-gigahertz to the THz range.

This approach provides new insights into the formation of spintronic effects and reveals new phenomena, which exist only in non-equilibrium states on ultrashort time scales. A major reason is that the THz range coincides with the natural frequencies and rates of electron transport, lattice vibrations (phonons) and spin waves (magnons) (figure 3). Finally, THz spintronics may not only enable faster spintronic operations, but also new THz-photonic functionalities such as the generation, modulation and detection of THz radiation.

Magnon spectroscopy. THz magnetic fields can be used to directly drive spin precession by the Zeeman torque and to excite magnons (figure 4(a)). This approach is useful to characterize antiferromagnets such as insulating ferrites (e.g. YFeO_3) [18, 19]. In $\text{Er}_x\text{Y}_{1-x}\text{FeO}_3$ in static magnetic fields above 3 T, one can additionally induce a THz precession of the Er^{3+} spins [19].

Remarkably, linear THz spectroscopy revealed that the coupling of the Er^{3+} spins with a Fe^{3+} magnon mode can become ultrastrong if their precession is tuned in resonance with the magnon frequency (figure 4(a)). As a result, the magnon acts akin to a cavity photon mode in cavity electrodynamics. Such condensed-matter-based THz quantum simulators provide new opportunities to reveal novel phenomena that are predicted for light-matter coupling Hamiltonians but have remained experimentally elusive so far [21] (see section 2).

Magnetic-order switching. For spin switching, indirect pathways through electron or phonon excitation are often more promising than direct magnetic-field driving through Zeeman torque (figure 3).

For instance, resonant phonon excitation by intense THz electric fields was used to switch ferrimagnetic [22] order or induce ferromagnetism in an antiferromagnet [23]. Such THz-nonlinear studies (see section 5) are not only relevant for

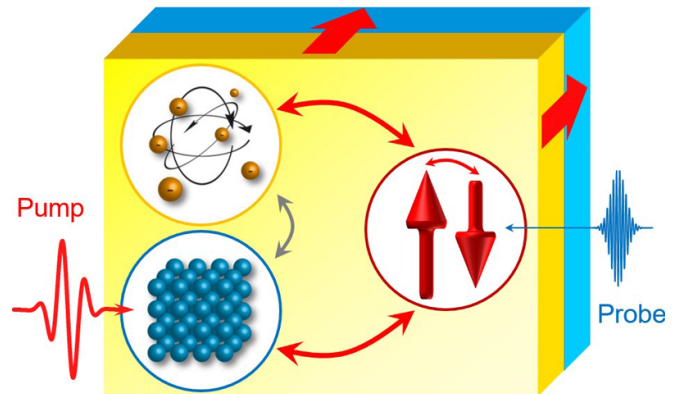


Figure 3. Basic concept of THz spintronics. An ultrashort electromagnetic pump pulse (THz or optical) induces changes in the magnetic order of a solid by excitation of electron spins, crystal lattice (as shown here) or electron orbital degrees of freedom. The resulting torque on spins or spin transport into an adjacent layer are monitored by a suitable probe pulse. At least one of the two pulses is a THz pulse.

THz spintronics, but also provide fundamentally new insights into spin-phonon and spin-electron interactions.

Laser-driven THz spin transport. Femtosecond optical laser pulses were shown to drive THz spin transport in stacks made of magnetic layers and layers without magnetic order (figure 4(b)). Macroscopically, the ultrafast spin currents arise from optically induced gradients of (generalized) electron temperature and spin voltage [24], both of which are established spintronic concepts. Interestingly, the transient spin voltage is an ultrafast route and sizeable only before spins and hot electrons have equilibrated. To detect the THz spin current, one uses spin-to-charge-current conversion, which transforms the spin flow into a perpendicular charge current and, thus, a measurable THz pulse [24] (figure 4(b)).

This scheme is highly useful to rapidly characterize conversion in the bulk and at interfaces [24, 25] or the spin-current generation by various magnets [24, 26] of as-grown thin films without microstructuring. One can even extract the temporal evolution of the spin current and obtain a better understanding of electron- and magnon-driven spin currents [20] (figure 4(b)). Finally, this concept allows one to efficiently generate THz pulses in optimized spintronic structures, covering the full 1–30 THz range and offering functionalities like rapid THz polarization modulation [24].

Current and future challenges

With regard to photonic applications, the efficient optical generation and detection of THz electromagnetic pulses remains a central goal of THz spintronics. With regard to spintronics, it is important to note that spintronic devices are typically driven and probed by low-frequency voltages.

Therefore, to explore spintronic functionalities at THz frequencies, one should directly use THz electric fields rather than rectified optical laser pulses. In this way, all flavors of electrically driven spin transport (conduction-electron- and

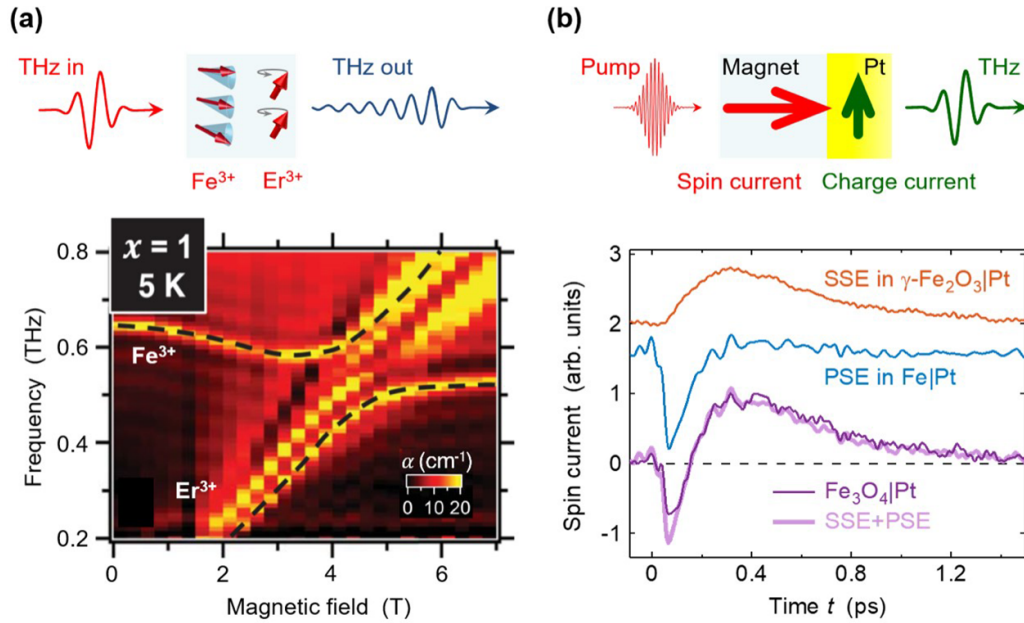


Figure 4. THz spin torque and THz spin transport. (a) Zeeman torque of an incident THz pulse induces spin precession and THz emission. Measuring the THz in and THz out signals yields the absorption coefficient of ErFeO_3 vs frequency and external magnetic field at 5 K. The anticrossing of the Fe^{3+} magnon mode (field-independent feature) and the Er^{3+} spin precession (diagonal feature) indicates ultrastrong coupling [19]. From [19]. Reprinted with permission from AAAS. (b) Optical excitation launches spin transport from a magnetic layer into a Pt layer, where the spin flow is converted into a charge current. Measurement of the emitted THz pulse allows one to retrieve the spin-current evolution. For the insulating ferrimagnet $\gamma\text{-Fe}_2\text{O}_3$ and the metallic ferromagnet Fe, respectively, the spin transport to Pt is mediated by magnons and conduction electrons through the pyrospintronic effect (PSE) and spin Seebeck effect (SSE). For the ferrimagnetic half-metal Fe_2O_3 , a superposition of both spin-current flavors with their distinctively different dynamics is found [20]. Reproduced from [20]. CC BY 4.0.

magnon-carried), spin torque (e.g. field- and damping-like spin-orbit torque) and magnetoresistive spin read-out can be studied over the full THz range of 1–30 THz. An ultimate goal is to implement these operations in on-chip circuitry [27].

Coherent spin pumping is a ubiquitous spintronic technique where processing spins transfer spin angular momentum into an adjacent metal layer. It was recently demonstrated at sub-THz rates [28, 29]. Importantly, the spin-pumping amplitude scales with the driver frequency, suggesting even better performance at THz rates.

Spin-orbit torque is a powerful mechanism that allows one to switch magnetic order by applying charge currents. It recently approached ultrafast time scales by using 6 ps wide electric-field pulses [27], which reversed the magnetization of ColPt stacks. Pushing this approach to THz frequencies is particularly interesting for antiferromagnets, where resonant coupling to THz long-wavelength magnon modes can be exploited.

Magnetoresistive effects are important spintronic tools to characterize magnetic order of ferro-, ferri- and antiferromagnets. Anomalous Hall effect and giant, tunnel and anisotropic magnetoresistance were recently transferred to the range below 5 THz. Broadband anisotropic-magnetoresistance measurements at 1–30 THz allowed one to straightforwardly separate extrinsic (scattering-related) vs intrinsic (scattering-unrelated) contributions [30]. However, a direct measurement of the THz spin Hall effect, a central spintronic phenomenon,

and related effects such as spin Hall magnetoresistance has remained challenging and is an important future goal.

Advances in science and technology to meet challenges

THz technology. To fully cover the 1–30 THz window, broadband THz emitters are required, for instance, based on gas plasmas or spintronic materials [24] (see sections 7 and 9). To interrogate the THz-field-driven dynamics with a high signal-to-noise ratio, ultrashort intense optical pulses (duration <30 fs) at a high repetition rate are desirable, for instance, through fiber-laser technology.

Suitable probes. Reliable ultrafast detection of antiferromagnetic order will be a necessary breakthrough for advancing the emerging technology of antiferromagnetic spintronics. Coming from the optical side, ultimate options are ultrafast x-ray magneto-optic effects or x-ray-induced photoelectrons, which may simultaneously provide high spatial resolution. Coming from the electrical side, THz magnetoresistive effects and emission spectroscopy (see above and sections 3, 10 and 12) are a promising ultrafast monitor of magnetic order.

Likewise, in technologically relevant spintronic thin-film stacks, interfaces critically impact the spin dynamics [24, 25]. To separate the interface from bulk contributions, THz emis-

sion spectroscopy (see figure 4(b) and section 11) offers a large potential that needs to be explored further.

Theory development. The description of spintronic and magnonic effects at THz frequencies is generally non-trivial because complex processes such as electron-momentum relaxation [30], electron thermalization [24], electron–spin [24], electron–phonon [24] and spin–phonon [22, 23] equilibration become relevant on sub-picosecond scales in highly correlated solids [31]. A novel approach in this direction is

time-dependent density functional theory, which will soon also capture phonon dynamics [32].

Concluding remarks

Applying ultrabroadband THz radiation and femtosecond laser pulses to spintronic and magnetic structures provides new insights into fundamental spintronic processes. On the other hand, exciting applications for spintronic information processing and THz photonics emerge.

4. Terahertz time-domain polarimetry and ellipsometry

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Status

Since the late 1980s the number of applications of terahertz (THz) radiation has experienced an extraordinary growth. Most of the studies that have since emerged present either transmission or reflection spectroscopy, or imaging, where the main variables used for the data analysis is either the amplitude or phase of the THz wave. The polarization of the radiation [33] has also provided rather interesting insights on the anisotropy of many samples, leading to new techniques known as THz polarimetry and ellipsometry. For instance, characterization of birefringent materials where the birefringence originates from the mesoscopic structure provides exciting possibilities for industrial inspection [34]. Furthermore, THz polarimetry opens possibilities in the characterization of externally induced effects on materials: for example external magnetic fields allow the determination of Hall conductivity in a contactless fashion or the cyclotron effective mass of charge carriers on a picosecond timescale [35]. In addition, measurement of the full polarization state allows the determination of the complex optical properties of non-transmissive materials by ellipsometry [36].

The accurate measurement of the polarization of THz beams, particularly using pulsed time-domain spectroscopy has been challenging. However, the introduction of polarization sensitive devices [37–39] as well as the possibility of finding appropriate polarization sensitive geometries for electro optic sampling (utilizing beamsplitters [39], polarization modulation [40] and spinning electro optic sensor [41]), has opened the possibility to measure the polarization of THz beams more accurately and rapidly. Some examples of polarization sensitive devices that have been introduced are shown in figure 5 as well as a schematic of the polarization sensitive measurement based on electro optic sampling.

Current and future challenges

One of the main challenges in THz polarimetry lies in improving the accuracy of currently available devices and techniques for the measurement of the polarization state. The precision of the measurements of dielectric properties of materials by ellipsometry strongly depends on the capacity to resolve changes in polarization. Likewise, the determination of subtle effects,

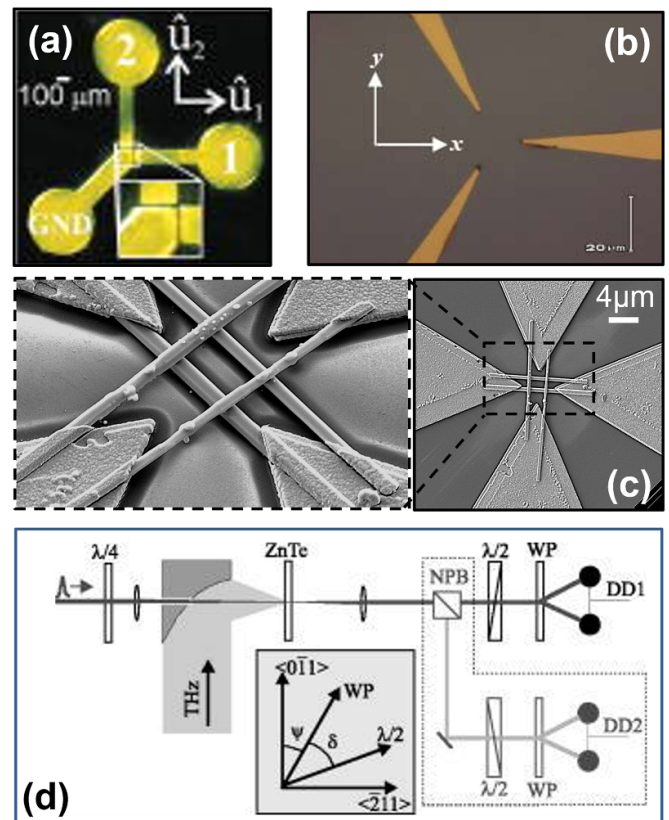


Figure 5. THz polarization measurements based on three different polarization sensitive photoconductive detector designs where contacts allow simultaneous measurement of more than one component of a terahertz transient (a)–(c) and based on electro-optic sampling where external polarizers are used (d). Reprinted from [37], with the permission of AIP Publishing. Reprinted with permission from [38] © The optical Society. From [39]. Reprinted with permission from AAAS. Reprinted with permission from [38] © The optical Society.

such as THz circular dichroism (TCD) in biomolecules [43], which could be a game changer for structural biology, require the possibility to sense very small changes in polarization.

Another challenge to be addressed is the cost, portability and flexibility of THz time-domain systems with polarization measurement capability. One of the most significant advances towards real-world applications that THz time-domain spectrometers have seen in the last years is the shift from using free-space optics and Ti:sapphire lasers to using rare-earth-doped fiber lasers, and fiber-coupled optics. With this change, TDS systems have become cheaper, more robust, portable, less sensitive to environmental conditions, and do not require special laser safety training and equipment for the operators. Furthermore, fiber coupling of the THz emitters and detectors makes realignment of the setup quick and easy, and allows imaging of immobile objects. We can expect these advances to influence the application of THz polarimetry.

The emergence of practical applications of THz polarization imaging [44], especially in biomedical sciences, has highlighted the need for more accessible and stable continuous wave sources that can replace and extend the frequency

range of traditional sources based on electron tubes and Gunn diodes. Continuous wave imaging continues to attract attention, particularly because its monochromatic nature allows for simpler and more sensitive polarization components. On the other hand, high-speed polarization imaging will become a research hotspot to meet the industrial application criterion.

Advances in science and technology to meet challenges

The introduction of fiber coupled polarization sensitive photoconductive detectors seems to be the most viable solution to achieve economical, robust and flexible THz time-domain spectrometer systems that could, in principle, be available commercially in the near future. This of course requires the fabrication of such detectors on narrow bandgap materials such as InGaAs, and finding the appropriate way to fiber couple the detector while maintaining appropriate illumination of the gaps. The introduction of such devices would increase the number of laboratories with access to such measurements, and therefore the investigation of anisotropic properties of materials, as well as the availability of ellipsometry THz systems, which is at the moment rather limited.

The accuracy of the polarization measurements will require the introduction of new geometries and the development of improved polarizers and other optical components (with higher signal to noise ratio and/or polarization extinction ratio) in order to characterize and demonstrate the improvement in performance. The establishment of calibration and measurement protocols will be needed.

THz polarimetric imaging has regained public attention in recent years thanks to technological progress in THz polarization sensing, and the spatial resolution provided by imaging technologies offers a significant advantage in polarimetric studies. For example, personnel security screening has been demonstrated with THz polarimetric imaging [45], offering enhanced object contrast while providing additional polarization state distribution characteristics for information extraction of human bodies and concealed objects. Considering the ease of implementation, single-pixel THz imaging [46] seems to be the most feasible technique for realization of spatial, spectral, and polarization-resolved THz imaging systems with reasonable speed and accuracy, but related data

interpretation and image reconstruction algorithms need to be carefully developed. Concurrently, incremental progress in more conventional scanned polarimetric imaging continues. Hybrid detection methodologies [44, 47], which include interferometry and confocal techniques, that significantly improve the sensitivity and contrast of polarimetric imaging are emerging. Benefiting from the advances in polarization-resolved THz system development, imaging via TCD spectroscopy has become increasingly practical. In one aspect, the ability to resolve the full THz polarization state is key to the characterization and design of novel materials (such as plasmonic modulators [43]) that can produce circularly-polarized THz radiation for use in TCD. In another aspect, the TCD system itself is a polarization-resolved system. The practical realization of TCD will lead to a revolutionary change in biomedical science. Because many biological systems are composed of chiral molecules and their functions depend strongly on their chirality that can change during biochemical reactions. TCD has the potential to accurately probe such changes in real-time while allowing one to precisely distinguish various biomolecule groups.

Emerging techniques are rapidly increasing the range of applicability of THz polarimetry. For example, electro-optic sampling can increase the typical working bandwidth [48] or precision [49]; advances in photoconductive systems can increase efficiency [50] and complementing polarimetry with other techniques adds rigor to measurements [51].

Concluding remarks

The study of anisotropic properties of materials with the implementation of high accuracy ellipsometry systems at THz frequencies opens new and exciting possibilities in material science, biochemistry and many other areas. The development of methods and devices that allow for THz time-domain based polarimetry and ellipsometry has been an important step towards a broader use of such techniques. However, the number of laboratories in the world with such capabilities is limited, and the number of publications that benefit from probing the properties of materials with THz polarimetry is small. We foresee that when commercial THz time-domain systems offer the possibility of determining the full polarization state of the THz radiation, the use of this technique will expand and may also contribute significantly to imaging.

5. Nonlinear terahertz spectroscopy

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Status

The progress in nonlinear THz spectroscopy is largely driven by the advances in the development of efficient THz sources, delivering signals with peak electric field strength in the range of 100s of kV cm^{-1} to MV cm^{-1} [52]. Demonstration of strong-field THz generation on the table-top in 2000s, based on the tilted pulse front (TPF) excitation of lithium niobate, enabled nonlinear THz spectroscopy as a popular and widespread experimental technique, mostly used in fundamental research in materials science. Besides TPF, other popular strong-field THz generation techniques on the table-top are collinear THz generation in organic nonlinear crystals, air-plasma and, since recently, use of spintronic THz emitters in conjunction with mJ-pulse laser pumping. Apart from the table-top strong-field THz sources, large-scale accelerator-based THz sources become more and more accessible to the user community. Such sources demonstrate unique characteristics typically not accessible with table-top approaches, such as ability to generate strong-field quasi-monochromatic, multi-cycle THz and sub-THz waves, and thus enabling e.g. experiments on high-order THz harmonics generation [53, 54]. Further, strong-field phase-stable multi-THz quasi-monochromatic multi-cycle pulses generated via difference-frequency generation in laser-pumped nonlinear crystals are used in table-top nonlinear spectroscopy experiments extending into the IR and even into visible spectral range [55].

Since the free carrier response typically represents the most prominent, and also most broadband, contribution to the optical conductivity of materials in the THz range, the strongest THz nonlinearities usually also originate from the free carrier effects. Here, the driving THz electric field couples to the electronic system of the material via optical conductivity mechanism, thus facilitating the efficient transfer of energy from the driving THz field, and typically leading to carrier heating and modification of particle distribution within the bandstructure of the material. This in turn leads to the significant modification of the dielectric function of the materials, typically semiconductors or semimetals, and hence of their THz optical properties, manifesting in the pronounced THz nonlinearity. Examples of such effects are carrier heating in doped semiconductors [56], graphene [53, 57], topological insulators [54] etc. Graphene in the THz frequency range was recently shown to have by far the largest nonlinear coefficients of all known functional materials [53] (see figure 6). THz-driven impact ionization and Zener tunneling, resulting in the increase of free carrier density in semiconductors, also lead to pronounced THz nonlinearities [58, 59]. Infrared-active THz-frequency optical phonons can also contribute to

the THz nonlinearity in polar crystals [60]. Natural THz nonlinearity of materials can be further enhanced using local field-concentration methods, such as application of plasmonic metallic gratings [61].

THz nonlinear spectroscopy has also been applied to solids where many-body interactions are important, such as superconductors and multiferroics. In particular, in superconductors, where the energy of the quasiparticles belongs to the THz range, Higgs mode excitation and related harmonic generation have been reported [62]. Future developments in these studies are expected. One needs to further mention remarkable developments in the nonlinear THz spectroscopy on ferroelectric materials with structural phase transitions. Since the soft mode involved in the phase transition also belongs to the THz range, large-amplitude excitation is possible with intense THz light [63]. It has been reported that the spatial inversion symmetry in a ferroelectric disappears due to strong THz light irradiation, in the experiment involving the optical second harmonic generation as a probe [64].

The methods of nonlinear THz spectroscopy currently in use are nonlinear THz time-domain spectroscopy (NL THz-TDS), where the transmitted THz field is measured and analyzed in a fashion similar to standard THz-TDS, but now dependent on the field strength in the incident THz wave [53]; THz pump—THz probe spectroscopy, where the transmission of a weak THz probe pulse following the strong THz field excitation is analyzed [57]; and THz pump—optical probe, where the transmission or reflection of a weak optical-range probe signal following the THz pump is analyzed [65]. A separate class of nonlinear THz spectroscopy experiments is multi-dimensional THz spectroscopy, often involving complex modulation schemes [66]. Finally, a very new class of experiments involving strong-field THz excitation of materials is THz-assisted surface science experiments combining the THz excitation of materials with UV/EUV-driven photoemission spectroscopy, such as the pioneering angular resolved photoelectron spectroscopy experiment under THz excitation [67].

Current and future challenges

In the recent 10 years the nonlinear THz spectroscopy became a well-established method of fundamental research used by many groups around the world. Almost every group involved in THz time-domain spectroscopy using mJ-level amplified laser is currently capable of performing nonlinear THz spectroscopy experiments of a certain kind. This is the result of wider availability of amplified laser systems, commissioning of new strong-field THz beamlines at large-scale facilities open to a broader user base, and, above all, constructive sharing of expertise within the ever-growing THz spectroscopy community. Nevertheless, the challenges in further development of nonlinear THz spectroscopy remain.

One of the key outstanding challenges still remains to be the availability of strong-field quasi-monochromatic table-top THz sources in the frequency range of ca 5–15 THz, corresponding to the reststrahlenband of most THz-emitting materials [1]. Such sources are needed for highly selective

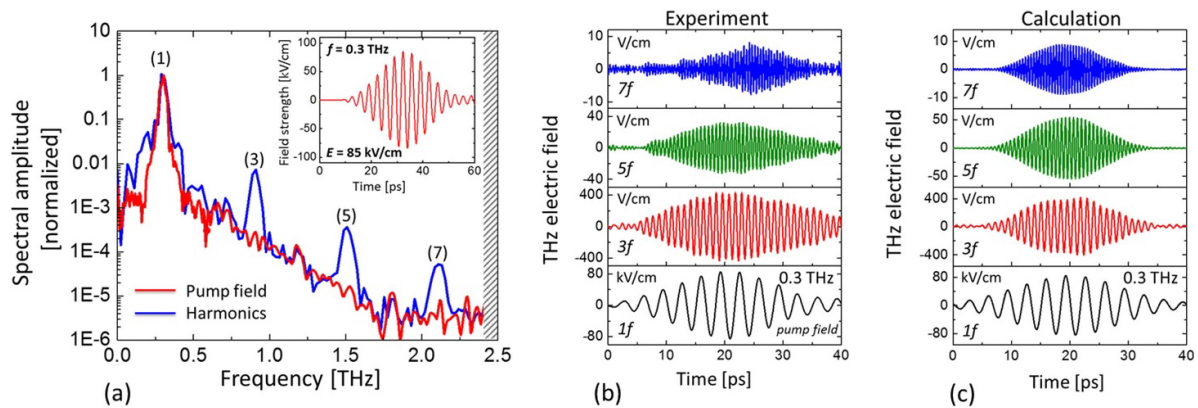


Figure 6. THz high-harmonics generation in graphene, enabled by the thermodynamic response of electron population in graphene to intense THz excitation, which results in strongly nonlinear THz conductivity of graphene [53, 57]. (a) Amplitude spectra of incident and transmitted THz wave. Higher odd-order harmonics up to the seventh order are clearly visible in the spectra of the transmitted THz wave. Inset—temporal profile of the pump wave at $f = 0.3$ THz. Measured (b) and calculated (c) THz harmonics in the time domain. Reproduced from [53], with permission from Springer Nature.

resonant excitation of e.g. polar lattice modes, or for resonant many-photon THz excitation of materials. Even though the air plasma-based THz sources and THz spintronic emitters are able to provide single-cycle strong-field THz pulses covering the range 5–15 THz, the spectral density within a narrow bandwidth at these frequencies remains low, thus precluding efficient nonlinear THz spectroscopy with quasi-monochromatic fields.

Another challenge is the improvement of the signal-to-noise ratio and the dynamic range in nonlinear THz spectroscopy. This will enable the detection of very weak generated nonlinear signals, such as e.g. very high THz harmonics with correspondingly very low electric field amplitudes [53].

The future development of a recently demonstrated THz-assisted photoemission spectroscopy [67] will similarly pose certain challenges in the mitigation of THz streaking effects on the detected photoemission spectra, and general increase of signal-to-noise ratio in this novel and highly promising type of nonlinear THz spectroscopy.

Advances in science and technology to meet challenges

In the coming years a major effort will be in overcoming the above challenges in nonlinear THz spectroscopy. Possible solution to generation of multi-cycle, quasi-monochromatic strong-field THz waves in the range 5–15 THz could be the difference frequency generation—type excitation of gas plasmas or metallic spintronic emitters, which do not have inherent spectral gaps in their response related to the restrahlenband. Further, the emerging availability of mJ-class femtosecond lasers operating at very high repetition rates of 50–100 kHz

based on various technologies such as rod-fiber or optical parametric chirp pulse amplification will enable the generation of strong-field THz pulses at very high repetition rates—see the pioneering demonstration [68]. Such an approach will naturally lead to a corresponding order of magnitude increase of signal-to-noise ratio in ultrafast THz spectroscopy due to ca 100-fold increase of the repetition rate as compared to present generation of mJ-class pump lasers typically operating at 1 kHz repetition rate. However, the new challenge will arise, namely the thermal management of the experiment, since the repetition rate of 100 kHz combined with the pulse energy of several mJ range entails average laser power in hundreds of Watts. On the side of large-scale THz facilities, the increase of both repetition rate to above 100 kHz, and the peak THz fields in generated multi-cycle THz waves to MV cm⁻¹ range will enable unprecedented opportunities of nonlinear THz spectroscopy with resonant excitation, and field-resolved detection of THz fields using electro optic sampling.

Concluding remarks

We envisage a stable increase of high-quality research output in nonlinear THz spectroscopy in the coming years, driven by wider availability of strong-field THz sources and related laboratory expertise, as well as by the emergence of novel materials potentially demonstrating very high THz nonlinearity, such as e.g. Dirac materials. Further, novel modalities in nonlinear THz spectroscopy will be demonstrated and further developed, for example based on the combination of strong-field THz excitation with other analytical methods such as scanning tunneling microscopy and photoemission spectroscopy.

6. THz biological effects and sensing

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Status

THz light exquisitely overlaps the energy range for water relaxational motions and intermolecular vibrations of water and biomacromolecules. In addition to the spectroscopic insight that low power high dynamic range terahertz systems can provide for molecular studies, the development of high field pulsed THz sources are emerging as unique tools for unprecedented contactless membrane potential manipulation within cellular systems. Over the last 25 years, THz optical studies have provided insight into how picosecond biomolecular dynamics may influence biology and biomolecular function. Investigations have moved from initial cataloging of biomolecular linear THz transmission to THz optical measurements combined with multiple probes to answer specific biological questions, such as how picosecond dynamics effect catalytic cycles, and allosteric regulation. As instrumentation has continued to improve, systematic measurements have further shown that THz response can be used as an alternative measure for establishing chemical kinetic properties. THz optical studies are now poised to address questions such as the role of water in biology, the purpose of protein structure, the mechanisms of allosteric control, and the balance of membrane permeability and transcription regulation.

THz optical measurements of biomolecules was motivated by early modeling of the intramolecular vibrations, suggesting that only a few vibrations would dominate, and these vibrations would profoundly affect biological function. Many of the early, and sadly even recent studies used lyophilized powders of proteins and polynucleotides. To date, it is well established that a minimal hydration is necessary for biologically relevant dynamics, and a majority of measurements are now performed on solutions or hydrated films. Some early measurements also suggested narrow band absorption resonances, however these were the result of interference effects. Experienced investigators now routinely use multiple concentrations and/or sample cell thicknesses to avoid these well-known artifacts. With such controls in place, it is established that biomacromolecules have broadband THz absorbance, peaked in the 3–6 THz range. The broad response observed for unaligned samples is consistent with the large vibrational density of states for these complex macromolecules, as well as the configurational switching of the macromolecular structure on nanosecond time scales [69]. At 1.0 THz protein and polynucleotide molar absorptivity is

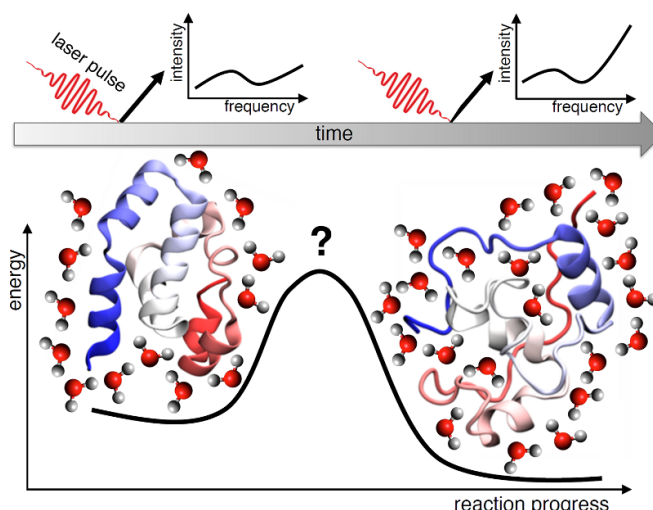


Figure 7. Plot of the free energy G during a reaction (here: protein unfolding). During the reaction, not only the protein structure but also protein hydration is changed, which is probed by THz spectroscopy as a function of time during protein folding.

found to be $\sim 900 \text{ M}^{-1} \text{ cm}^{-1}$ [70]. While considerably more than that of 25 °C water ($\sim 1.8 \text{ M}^{-1} \text{ cm}^{-1}$) [71], the typical saturation concentration for biomacromolecules is $< 20 \text{ mM}$, thus water absorption dominates. In spite of the very large water background, even the broad isotropic THz response of hydrated biomacromolecular samples is sensitive to temperature, ligand binding and photo excitation [72]. Indeed a variety of THz techniques [73–75] established underdamped protein intramolecular vibrations exist, supporting the notion that the vibrational ensemble for an intermediate conformation may provide access to conformational change.

This sensitivity to the intermolecular interaction between water molecules provides an opportunity to determine the contribution of the solvent to the total free energy of a reaction (see figure 7). Systematic spectroscopic studies revealed that the THz spectrum fingerprints any changes with respect to hydrogen bond strength, tetrahedrality, dynamics, which are all of major importance to rationalize and predict the outcome of a reaction. Even small changes in ion and protein hydration causes significant changes in the spectrum and could be investigated in detail. While the individual changes might be small, the large amount of solvent involved makes this contribution a major driving force for fundamental reactions. THz spectroscopy quantifies these changes during protein folding and upon introducing local mutations. Recent studies revealed that these spectroscopic signatures of hydration can be directly correlated to enthalpic and entropic changes of the solvent, H and S , which determine the reaction path and reaction rates [76]. Typically, for biological reactions, there is a subtle balance between a favourable enthalpy and an unfavourable entropic term, which almost cancel, thus allowing the fine-tuning of reactions by temperature. THz spectroscopy maps local water changes revealing whether these changes are more a cause or a consequence of fundamental biological reactions, such as enzymatic catalysis or protein aggregation.

While THz technologies are a valuable tool for sensing solvent and biomolecular dynamics, sufficient intensities may also induce structural and functional change in cells and tissue that must be characterized to establish safe exposure limits or develop novel therapeutic technologies [77–80]. Narrow-band CW sources are interesting for studying biological effects as they are capable of approximately single-mode excitation. However, these sources induce significant heating in bio-systems at moderate power levels, and the biological heat-shock response often obscures non-thermal biological effects.

Exposure systems that utilize pulsed, often single-cycle, THz sources are implemented to induce biologically negligible heating ('biologically negligible' is dependent on the specific system under study and must be verified), and effects observed may be attributed to non-thermal interaction mechanisms. The temporal localization of pulsed sources correspond to broad power spectra, and so a wider range of oscillatory dynamics may be excited. Several studies have reported significant biological effects that are not explained by the estimated heating, such as severe genotoxic stress, membrane permeabilization, and polymer disassembly.

Current and future challenges

The demonstration of underdamped intramolecular vibrations is an important first step towards understanding what role these dynamics may play in biology, such as whether a strongly excited vibrational band alters the catalytic rate. Such control of function by terahertz has not yet been demonstrated. The ideally the frequency bands relevant to specific motions would be known. Calculations have begun to indicate that some generalized displacements (such as hinging motions) may be concentrated in narrow energy bands even when configurational heterogeneity is accounted for [69], however experimental determination of these bands is a struggle given the broadband absorbance. Current focus is on selective detection of vibrations. One successful method is selection by transition dipole direction using anisotropic terahertz microspectroscopy [73]. Other methods under consideration include isolating vibrations strongly coupled to a chromophore excitation [81] or determining the displacements induced by a specific THz frequency by hydrogen deuterium exchange mapping after high power THz excitation [82].

Key concepts of characterizing solvent thermodynamics are firmly established in the sense of global properties of homogeneous bulk systems under thermodynamic equilibrium conditions. It is intriguing to see how local solvation phenomena in nanoheterogeneous environments determine the reactivity and selectivity in solution, receptors, and enzymes, and even the function of electrocatalysts [83, 84]. While macroscopic solvation can be tracked by traditional calorimetry approaches, local solvation and processes in non-equilibrium cannot. THz spectroscopy uncovers when local mutation serves as a game changer for protein dynamics. These measurements allow conductivity measurements in micelles [85], and probe hydration within supramolecular nanocages, even in the vicinity of

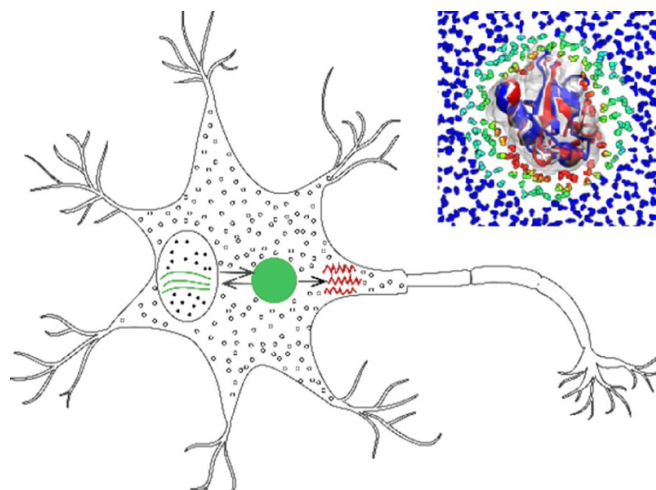


Figure 8. 'Local solvation hotspots' in cells are suspected to trigger neurotoxic protein aggregation. The hydration shell of a single protein (inset) is inhomogeneous, and contains areas of entropically highly unfavourable (red) and unfavourable (green) zones, which facilitate protein aggregation.

an electrified interface [86]. Local 'solvation hotspots', such as the so-called liquid–liquid phase separation membraneless compartments in a cell are suspected to trigger neurotoxic protein aggregation [87] (see figure 8). There are indications that the phase separation is entropy driven, with the solvent playing a decisive role. Development of new experimental tools, such as THz nanoscopes, would allow study of local hydration changes in living cells in real time.

Current challenges for understanding high pulsed field cellular response are robust studies of the exposure dependence of the response. Care must also be given to the choice of biological system to study. Cellular systems that are responsible for different regulatory roles have structural and functional differences that affect the set of possible responses available when introduced to external stimuli, and so different cell or tissue types may respond differently to a given THz exposure condition.

Advances in science and technology to meet challenges

A considerable focus on attaining high resolution terahertz nanoscopy has yielded imaging of solvated single biomolecules [88], however spectroscopic imaging with these systems needs to be developed.

2D-IR (IR pump-IR probe) spectroscopy techniques have been highly successful for condensed phase and allow to deduce structural information such as vibrational mode coupling, anharmonicities, as well unravelling chemical dynamics, including energy transfer rates and chemical reactions with femtosecond time resolution. These experiments have only become possible with the development of ultrafast (fs), high power IR laser pulses. Thus, further progress in THz technology is coupled to the availability of laser sources, which

generate high single cycle laser power as needed for an extension of the non-linear spectroscopy methods into the THz range. Especially in the chemical relevant frequency range (between 3 and 10 THz), adequate radiation sources are still lacking. Recent advances are based on the tilted pulse front method in lithium niobate, driven at an unprecedented high average power of more than 100 W and at a 13.3 MHz repetition rate, provided by a compact amplifier-free mode-locked thin-disk oscillator. The generation of a maximum THz average power of 66 mW, the highest reported to date from a laser-driven, few-cycle THz source, puts 2D THz spectroscopy in reach [89].

Interpretation of THz spectra require new theory and simulation methods. While *ab initio* molecular dynamics is able now to decipher the experimental THz spectra of small solutes, these cannot be extended yet to model more complex systems, such as a proteins or enzymes. For more complex systems, classical force field models have been successfully applied. While up to 1 THz non-polarizable force fields could be successfully used to describe energy flow, but these failed to reproduce even the absorption spectrum of bulk water, i.e. the maximum at 200 cm^{-1} or 7 THz. The recent developments of polarizable force fields are encouraging and will open new avenues for accurate description even of complex biochemical processes [90]. However more must be done to reach consensus for experimental needs. For example estimations are needed for the required sensitivity levels in THz

ellipsometry instruments to measure circular dichroism from protein solutions [91].

While a variety of THz-induced effects on cells and tissues have been reported, the differences in the exposure conditions and sample types investigated across studies have limited the general conclusions that can be made. The ‘reproducibility crisis’ is a serious issue that requires a concerted response from the research community. The primary reason for reproducibility errors has been a lack of rigorous dosimetry reporting [92, 93]. In 2011, Wilmink & Grundt recommended a rigorous dosimetry framework for THz exposure studies to mitigate the dilution of research findings due to spurious results from insufficient reporting practices [94]. Studies investigating THz-induced biological effects should only be formulated with an established dosimetry framework.

Concluding remarks

Synergistic combination of experimental THz techniques, establishment of best practices along with advanced computational approaches will provide needed insight into the role of biomolecular dynamics in biology, solvation properties site-specifically down to the level of individual molecules, and possibly novel control of cellular function. This will in turn impact innovative energy conversion and storage device development, quantitative predictions of biomolecular recognition and in the long run in areas such as sensor technology and drug design.

7. Terahertz spectroscopy of emerging materials

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Status

Since the inception of terahertz time-domain spectroscopy approximately 30 years ago, THz spectroscopy has evolved from an academic novelty into an indispensable tool for materials characterization, for which there are several user-friendly commercial systems available on the market. Owing to its frequency range, THz spectroscopy is ideal for probing intraband electrical transport and low-energy excitations in emerging materials. As a free-space optical technique, THz spectroscopy is inherently contact-free. This circumvents the need to make and optimize electrical contacts and avoids processing steps that could potentially contaminate or modify the material. The ultrashort nature of (sub)picosecond THz pulses lends THz spectroscopy to time-resolved studies of ultrafast processes, for example using optical pump–THz probe (OPTP) spectroscopy or terahertz pump–terahertz probe spectroscopy. A further advantage is that THz spectroscopy measures the dielectric response of the material, rather than its luminescence. Therefore, unlike alternative time-resolved tools such as time-resolved photoluminescence (PL), THz spectroscopy is suitable for probing weakly-luminescent or non-luminescent materials, including those with undetermined band structures or low quantum efficiencies—properties that are common for materials in the earliest stages of their development while synthesis techniques are still being optimized.

In recent years, enormous strides have been made in the development of new materials, ranging from low-dimensional materials, to designer metamaterials. Advances in THz spectroscopy have kept pace with the development of these materials and enabled the study of their exceptional properties. For example, highly efficient THz high harmonic generation has recently been observed in electrically gated graphene probed by THz spectroscopy [57]. Optical pump–infrared push–THz probe spectroscopy—which is an extension of OPTP spectroscopy—has been implemented to reveal intraband relaxation processes and stimulated THz emission in tin iodide perovskites [95]. The Mott transition between an electron–hole plasma and excitons in WSe₂ van der Waals bilayers has been observed using ultrafast THz nanoscopy [96]. InAs plasmonic disk arrays have demonstrated nonlinear perfect absorption—implemented both as optical limiters and saturable absorbers [97]. These observations were made possible by advances such as high field THz pulse generation, THz-transparent polymer-electrolyte gates,

broadband THz generation techniques, and near-field THz techniques.

Current and future challenges

Although each type of material will pose its own specific challenges, there are some general challenges:

- (i) Spatial resolution—Many emerging materials, particularly low-dimensional materials, are characterised by nanoscale features orders of magnitude smaller than the diffraction-limited spot size of the far-field THz probe ($\sim 300\ \mu\text{m}$). This limits far-field THz spectroscopy to spatially-averaged measurements, across inhomogeneous regions or ensembles. Even with existing near-field techniques (figure 9(a)), it remains challenging to probe individual nanoparticles and the internal structure of materials with nanometer-scale resolution.
- (ii) Broad bandwidth—The need for broad bandwidth (0.1–>30 THz) measurements is apparent in figure 9(b), which plots the immense frequency range spanned by typical spectral signatures in emerging materials. Spectral features can also shift depending on experimental conditions and the geometry of the material [98]. THz conductivity spectra of emerging materials are often broadened by high charge carrier scattering rates, compounding the need for broadband sensitivity.
- (iii) Signal-to-noise ratio—Many novel materials feature weak absorption of THz radiation, necessitating improvements to measurement sensitivity. Moreover, THz measurements often involve compromises between bandwidth, signal-to-noise ratio, acquisition speed and spatial resolution. The obvious way to improve signal-to-noise ratio is to increase the intensity of the THz pulse, but intense THz fields can induce non-linear and non-equilibrium effects so this probe is no longer minimally-invasive. New techniques are needed for improving signal-to-noise ratio without compromising other parameters.
- (iv) Polarization—The ability to probe polarization-sensitive THz-range phenomena will reveal new insights into bandstructure and selection rules, spin states and intrinsic chirality in emerging materials.
- (v) Non-linear phenomena—Intense THz fields are predicted to drive non-linear phenomena in some novel materials, as already demonstrated in graphene [57]. These phenomena are promising for high frequency information and communication techniques, and worthy of detailed investigation.
- (vi) *In-situ, operando*, cross-correlated and environmentally-controlled studies—As THz pulses are a minimally invasive probe of equilibrium and excited states, THz spectroscopy is well-suited to *in-situ* or *operando* measurements, particularly of electrochemically-active materials, but so far there are few studies in this area [99]. In cross-correlated studies, THz spectroscopy is performed

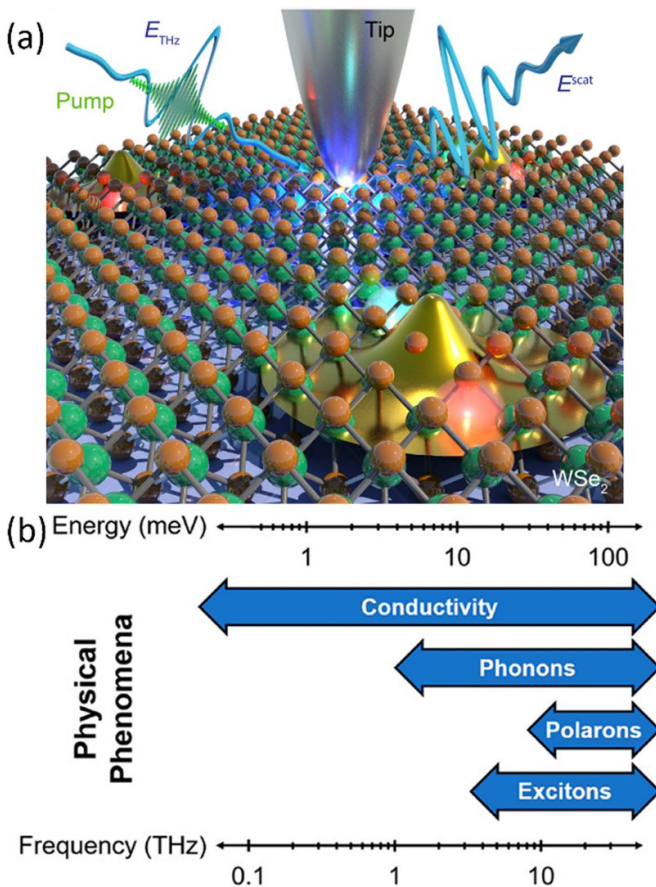


Figure 9. Near-field THz techniques are required to elucidate the properties of emerging materials featuring spatial inhomogeneity such as WSe₂ van der Waals bilayers. Reprinted with permission from [96]. Copyright 2022 American Chemical Society. Emerging materials exhibit signatures of electronic and quasiparticle phenomena across the THz frequency range. Reprinted with permission from [102]. Copyright 2020 American Chemical Society.

in concert with a complementary technique (e.g. PL) to yield fuller insight and disentangle competing processes such as recombination from charge carrier separation across interfaces [100]. Ideally, cross-correlated measurements would be undertaken simultaneously to maintain rigorously identical experimental conditions. Some emerging materials—especially quantum materials such as topological insulators, Weyl semimetals, etc—exhibit exceptional properties in extreme environments such as at cryogenic temperatures and under high magnetic fields. These types of studies are challenging experimentally due to constraints on the THz beampath and transparency of the media.

- (vii) **Theory**—Robust theories are needed to interpret THz spectra and yield insight into the physical mechanisms at play. The use of phenomenological models of THz conductivity is only justified under specific conditions [101]. In future, the increasing availability of artefact-free and highly broadband experimental data may support the validity of some of the models currently used, but call other models into question.

Advances in science and technology to meet challenges

Advances in both instrumentation and theory will allow THz spectroscopy to address the existing and forthcoming challenges posed by emerging materials. Key areas for advancement include:

- (i) **THz near-field techniques**—As discussed in section 13, scattering-type near-field scanning probe systems have entered a phase of rapid development. Further improvements to their sensitivity, resolution, throughput and user-accessibility will enable the study of inter-nanoparticle variability, enable the mapping of processes across interfaces and grain boundaries, and eliminate inhomogeneous broadening. Miniaturization of THz emitters and detectors, and the integration of these miniaturised components with samples and waveguides, is another promising route towards near-field THz spectroscopy.
- (ii) **THz waveguides** are needed for the delivery of THz pulses to and from samples placed in an optically inaccessible space, such as a dilution fridge. Ideally, THz waveguides which allow multimodal or broadband wave guiding will be achieved.
- (iii) Creative alignment strategies are required to achieve simultaneous and cross-correlated measurements of material properties.
- (iv) **THz metamaterials** permit access to the full range of responses possible at the subwavelength scale in accord with Maxwell's equations. They possess the capability to enhance every functional terahertz system component [103]. Implementation of high Q-factor metamaterials [104] would enable dichroic filters necessary for higher precision Raman and PL measurements in concert with THz spectroscopy.
- (v) **Broadband THz generation and detection**—Further development, especially of spintronic (section 3) and air-based (section 8) THz generation and detection schemes, are needed to extend bandwidth and achieve adequate signal strength up to at least 30 THz.
- (vi) **THz polarizers and polarization-sensitive detection systems**, such as the nanowire-based system recently developed [39] will allow anisotropic and chiral material properties to be investigated. Implementation of THz vector beams could expedite investigations of polarization sensitive phenomena. Section 4 maps out potential advances in THz polarimetry.
- (vii) **Intense THz sources**—Continued development of THz sources delivering field strengths greater than 1 MV cm^{-1} is required to drive and probe non-linear effects. A detailed roadmap of non-linear THz spectroscopy is provided in section 5.
- (viii) **High repetition rate lasers**—As discussed in section 5, the advent of femtosecond lasers with millijoule pulse energies and repetition rates over 50 kHz will in principle improve signal-to-noise ratios by an order of magnitude compared to current mJ-class regenerative

amplifier-based systems that are limited to 1–10 kHz repetition rates. The average power from such a high repetition rate laser would be of the order of hundreds of Watts, so would cause substantial heating of the experimental system and of any photoexcited sample. Strategies to mitigate heating would be needed.

- (ix) Reduction of laser noise—Improvements to laser pointing stability and pulse-to-pulse energy stability, along with improved detection schemes that filter laser noise, are expected to yield a significant improvement in signal-to-noise ratio [105].
- (x) Theory and models—Data driven techniques such as deep learning will allow more sophisticated terahertz

devices and responses than what has been shown before, while enabling physics informed neural networks to uncover the underlying terahertz science [106, 107].

Concluding remarks

With time, further materials will emerge, each with new and specific challenges. Exciting developments in THz spectroscopy, many already underway, will propel our understanding of emerging materials. Reciprocally, many emerging materials, particularly metamaterials, exhibit exotic properties in the THz domain that will make them indispensable building blocks for future THz components.

8. THz air photonics

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Status

The efficient generation of THz-frequency radiation from laser-induced plasmas in air was demonstrated by Cook and Hochstrasser at the turn of the century [108]. The term *THz air photonics* refers to the generation and detection of ultrashort THz transients by a femtosecond laser-induced plasma in air. In the air plasma, the acceleration of free electrons in the plasma by the laser field $E(t)$ generates a THz transient under the right conditions. In contrast to THz generation in nonlinear crystals the plasma-driven THz generation is extremely broadband, resulting in THz transients with a duration as short as the driving laser pulse duration and a spectral bandwidth that approaches or even exceeds that of the driving laser if more than one harmonic of the laser is used in the process [109]. The short duration combined with the smooth and broad spectrum of THz air photonics signals makes appealing for broadband spectroscopy across the 1–30 THz range [110], also in ultrafast transient pump-probe studies [111].

Figure 10 shows a generic experimental setup for generation and detection of THz waveforms in air or other gases. THz air photonics is often conducted in combination with coherent detection in the same gas (air). This process is referred to as air-biased-coherent-detection (ABCD) [112] and is based on upconversion of a femtosecond probe beam focused in air just below the plasma formation threshold. The third-order nonlinearity of the detection gas is generally responsible for the field-induced second harmonic generation (SHG) proportional to the instantaneous field strength of the THz field [112]. Dispersion in both the generation and detection medium is minimal compared to solid-state materials, and hence the THz transient can be resolved with a bandwidth determined by the femtosecond probe pulse. THz air photonics thus offers ultrashort THz pulses with extreme spectral coverages up to 200 THz [113] and high peak field strengths [114] comparable to virtually any other THz generation/detection techniques. Figure 11 shows an example of THz generation from a two-color laser-induced plasma with the fundamental pump wavelength tuned from 1200 to 2200 nm at 1 kHz repetition rate. It provides a peak conversion efficiency of 0.34% (3.4×10^{-3}) at 1980 nm. The resulting THz pulse has a peak field strength of at least 8 MV cm^{-1}

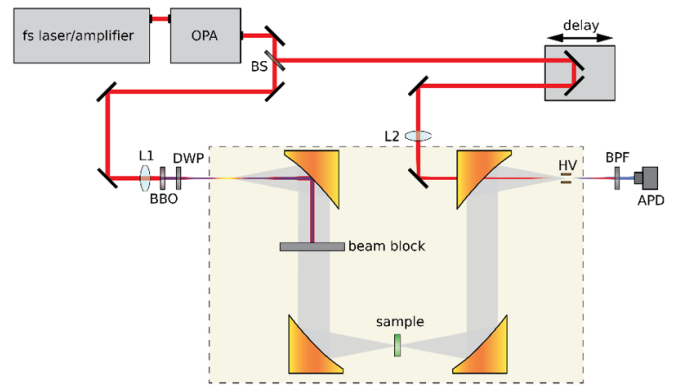


Figure 10. Experimental setup for THz air photonics. A laser beam from a femtosecond amplifier, possibly combined with an optical parametric amplifier (OPA) is split in two arms. One arm generates THz pulses via two-color laser mixing in air by focusing the split beam through a BBO crystal for second-harmonic generation and a dual-wavelength half-wave plate (DWP) for alignment of polarization. Residual pump light is blocked by a Si wafer. After passing the sample, the THz temporal waveform is detected by ABCD using a modulated high voltage (HV) and lock-in detection of the THz-induced frequency-doubled probe light with an avalanche photodiode (APD) after filtering with a band pass filter (BPF). The THz beam path is enclosed in a compartment purged with dry air or nitrogen to suppress water vapor absorption.

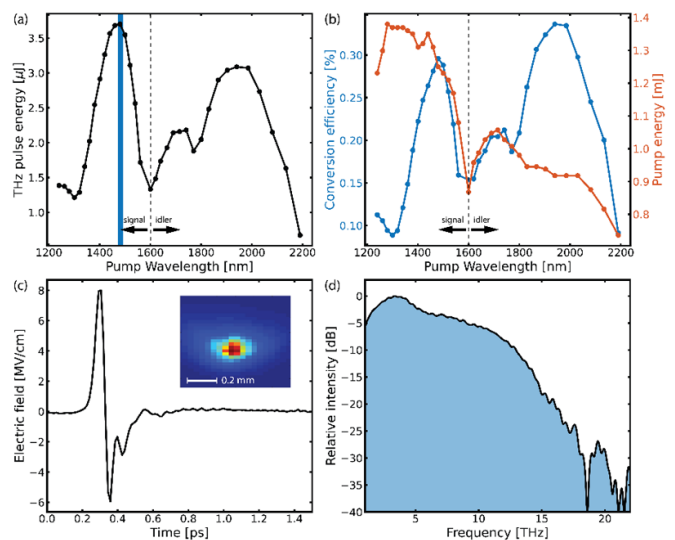


Figure 11. Some general properties of THz signals generated by two-color femtosecond air plasma. (a) THz pulse energy, (b) conversion efficiency (blue curve), and pump energy (red curve) as function of the fundamental pump wavelength. (c) Electric field of the THz waveform, calibrated using the focused spot size (inset) and pulse energy (position of blue, vertical bar in (a)). (d) Spectral intensity of the THz pulse in (c).

and detected spectral coverage 1–20 THz. These properties make THz air photonics highly relevant for spectroscopy [110]. The available pulse energies are comparable to those from free-electron lasers (FELs) that can provide ps to sub-ps pulses tunable across the THz range [115]. THz air photonics

delivers pulses with a duration of a few tens of femtoseconds, together with high peak field strengths, allows nonlinear studies on time scales below the typical electron scattering time in solid-state materials. Hence THz air photonics techniques are of great importance for lightwave-driven electronics in the future.

Current and future challenges

Compared to other THz sources, the generation mechanism in THz air photonics with more than one pump harmonic is relatively complex to describe and model. As shown in figure 10, the fundamental pump wavelength significantly influences the THz yield, which tends to scale strongly with the wavelength [116]. Many experimental parameters influence this scaling, including focusing of the pump beam, pump duration and phase matching conditions in the SHG crystal for harmonic generation, polarization of optical beams, as well as the phase offset between the fundamental and harmonic [117].

A significant challenge is the low repetition rate of current THz air photonics systems. The laser pulse energy must typically be in the millijoule (mJ) regime to exceed the ionization threshold of the gas and form a plasma. This practically limits the repetition rate to the kHz range for many laser systems. The initial demonstration of THz generation in air using strong focusing of sub-microjoule pulses and generation of microplasmas [118] and theoretical investigations [119] has shown the potential using a single harmonic. There are major technical challenges to overcome to further advance this technique, including focusing with high numerical aperture of more than one harmonic to the same position in the microplasma, with a controllable phase between the different harmonics. The development of high-repetition-rate THz air photonics is particularly important for its future use in near-field microscopy. There is already strong interest in combining THz sources with scattering-type scanning near-field optical microscopy (THz-SNOM) [120] since this technique allows for deep sub-wavelength resolution (<20 nm), thousands of times below the diffraction limit. THz-SNOM operates with a modified atomic force microscope in tapping mode, and so relevant light sources must have repetition rates high enough to allow for high-order demodulation of the scattered signal, up to hundreds of kHz. Hence, MHz repetition rates are required to bring THz air photonics onto a SNOM platform and enable its use in THz nanoscopy. THz-SNOM is currently mainly performed with tabletop light sources, but the technique has also been combined successfully with light sources at large user facilities such as FELs [121].

Advances in science and technology to meet challenges

To further enhance the capabilities of THz air photonics systems, advances in the understanding and control of the phase offset between the different harmonics of the pump laser are necessary. Also, the development of optical parametric amplifier (OPA) systems with increased control over the pulse wavefront and duration is required. Additionally, given the preference for long-wavelength pumping for high conversion efficiencies, high-power femtosecond laser systems operating in the mid-infrared (MIR), such as thulium-doped fiber lasers near 2000 nm, and possibly novel MIR OPA schemes, are strongly demanded for the advancement of THz air photonics.

Ongoing and future developments within high-average-power fiber-based femtosecond laser systems and external pulse compression techniques will also be important for the future growth of THz air photonics, not the least in relation to integration with THz-SNOM technology. Laser systems with mJ pulse energy at MHz repetition rate (kW average power) are already commercially available, with the first reports on high-average-power THz radiation approaching the watt level generated by mJ-level pulses focused in an argon jet at a 0.5 MHz repetition rate [122].

Apart from laser technology, the control of the THz field itself can be developed further. Advanced studies of the interaction between THz fields and matter on the femtosecond time scale benefits from complete control over the polarization of the ultrafast THz wave. As an example, circularly polarized light will enable access to a preferred spin polarization of carriers in a material, and subsequent emission of light from the accelerated charges provides signature information about the spin properties. Further development of techniques for the generation of other polarization states than the standard linear one, such as circular polarization, therefore, represents a significant challenge for THz signals with extreme bandwidth.

Concluding remarks

In summary, THz air photonics has over the past two decades developed into a mature and versatile technology for spectroscopy and sensing. The focus has been on understanding the properties of femtosecond laser-driven plasmas as THz sources and on finding their limits in the generation efficiency, power and bandwidth. In the future it can be expected that the sources will find their use in new research directions, including lightwave electronics and extreme THz science, thanks to their inherent unique qualities compared to other pulsed THz sources and their scalability in power and energy, given the right femtosecond laser sources are developed.

9. Recent progress in terahertz intersubband devices

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Status

Intersubband (ISB) transitions [123] are electronic excitations between the quantized energy levels within the conduction band of a semiconductor quantum well heterostructure, and unlike interband transitions, only involves one carrier type (see figure 12). A key property of ISB transitions is the possibility to design the energy separation between two electronic levels by simply changing the dimensions of the quantum well, without any limitation induced by the constituting material. For example, a 40 nm-thick quantum well based on AlGaAs/GaAs will lead to a transition of ~ 10 meV (~ 2.5 THz) between the first two confined states (see figure 13). The importance of such an approach is that it has opened up the possibility of artificial bandstructures to realize semiconductor devices in hard-to-reach spectral regions such as the terahertz (THz) frequency range. This has strongly impacted the THz domain over the last two decades, with the development of the THz quantum cascade laser (QCL) [124], one of the only practical and compact laser systems to date at such frequencies. Here sophisticated band structure engineering of GaAs-based quantum wells has enabled the centre wavelength of these THz lasers to be tailored flexibly, simultaneously enabling output powers of several watts [125], large gain bandwidths facilitating frequency comb (FC) operation [126] and ultrashort pulse generation [127], with operation up to 250 K recently demonstrated (2020) [128]. Band structure engineering has also been exploited in the design of intersubband detectors such as THz quantum well photodetector (QWP) where mid-infrared (MIR) concepts have been translated to the THz region [129]. QWPs possess an inherently fast (picosecond) response time and can be engineered in both patch antenna and meta-material architectures [130], taking advantage of both electronic and photonic approaches to light confinement, and resulting in relatively high temperature operation of 60 K [131].

ISBs also display atomic-like absorption from the subbands possessing the same parabolicity, which results in giant optical nonlinearities. This has led to room-temperature THz operation using intra-cavity difference-frequency mixing in MIR QCLs [132], FCs through four-wave mixing [126], nonlinear detectors [133], as well as recent demonstrations of high-order wave mixing in QCLs [134] and intersubband

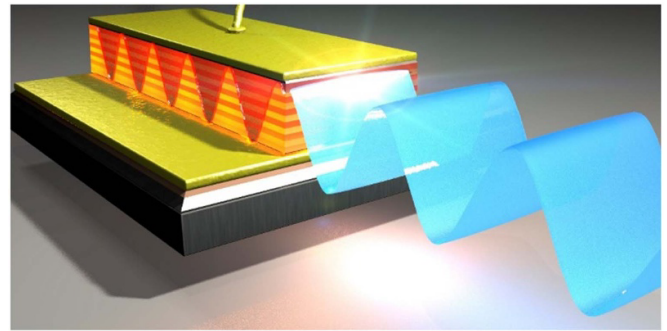


Figure 12. Artistic impression of an intersubband device—a THz quantum cascade laser emitting a THz beam (blue wave) from the end of the laser cavity. Image credit: D Darson.

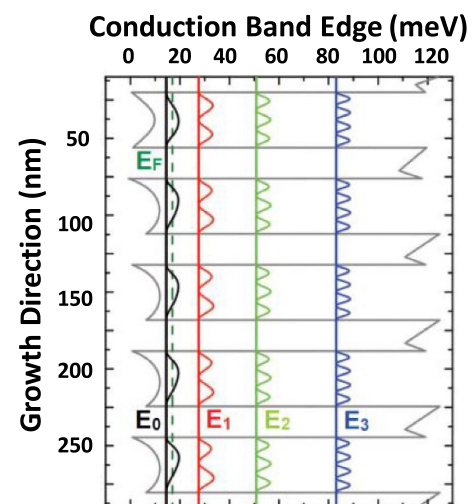


Figure 13. Bandstructure simulation of five quantum wells showing the first four subbands. The energy separation between the first two ($E_1 - E_0$) corresponds to an intersubband transition of ~ 10 meV.

polaritons [135] (coupling between a ISB and a cavity mode). Further, ISB transitions have extremely short lifetimes, owing to interactions of electrons with scattering mechanisms of the materials, resulting in ultrafast dynamics that is of importance for active modelocking for THz pulse generation [127] and ultrafast detection using the QCL itself as a detector [136]. These advances in ISB devices have strongly impacted applications and are nowadays successfully exploited for spectroscopy/metrology uses [137], and for non-destructive 3D imaging [138] and microscopy [139].

Current and future challenges

Despite unlocking new opportunities for both fundamental and applied research owing to their ability to be quantum engineered to operate in previously inaccessible spectral regions, the translation of ISB devices to widescale use outside the research environment has still to be realised. Immediate challenges include the development of turn-key compact, cheap

and portable THz imaging, spectroscopy and security systems of benefit to both academia and industry. Longer-term, the use of ISB devices in, for example, satellite instrumentation would be beneficial, capitalizing on the unique opportunities for THz QCL local oscillators in Earth observation and planetary sciences, targeting key atmospheric spectral signatures. There is also the potential to address the urgent need for both terrestrial and satellite-borne high data-rate wireless communication systems, utilising the high power of THz QCL sources and exploiting the broad unallocated spectral bandwidth available over 2–5 THz. Underpinning these objectives is the need to address the challenges presented by the necessary cryogenic cooling required for both source and detector, and the efficient handling of power consumption (especially for satellite implementations). Beyond THz detectors and lasers, there is a requirement for complementary devices, including THz amplifiers, modulators and saturable absorbers, that can also benefit from exploitation of ISB transitions. There is then an accompanying need to move to sub-system and fully integrated system developments, with rugged user-friendly interfaces for widescale uptake, rather than a focus on individual component development in a research environment.

An interesting challenge is to broaden the spectral coverage of ISB devices to address the 5–12 THz range, and more specifically the Reststrahlen band, which lacks the compact sources and detectors available to lower (THz) and higher (MIR) frequencies owing to the strong photon absorption of the constituent III–V based materials systems. This could open interesting perspectives for a number of applications such as sensing complex hydrocarbons in the petroleum-industry and in astronomy, or quantum optics, for example in the manipulation of Rydberg artificial atoms for quantum computation architectures.

Approaches to develop high-power (>10 mW) broadband THz QCL FCs, operating across the entire dynamic range of the laser, remains a topic of considerable current interest, but the power per mode remains limited. Complementary to this has been ultrashort pulse generation from QCLs, where progress have been made down to the generation of few-picosecond pulses in an active, cumbersome configuration, but neither passive mode locking in the THz nor the femtosecond regime has been reached yet. The simultaneous generation of much shorter pulses with high powers through new integrated modelocking geometries remains therefore a challenge. Finally, harmonic modelocking by engineering the bandstructure, waveguide and group velocity could be a valuable approach to achieve both goals simultaneously. This would also open up opportunities in THz quantum optics where the nonlinear nature of frequency comb and pulse generation could potentially permit the observation of quantum correlations between the comb modes. Here, the parallel developments of sensitive, high-speed and high-temperature operation ISB-based detectors would also considerably advance the field targeting sensitive and shot noise level sensitivity.

Advances in science and technology to meet challenges

Although there has been improvement in the THz QCL operation temperature, with recent demonstrations of Peltier-cooled operation, room temperature operation, or at least high power and continuous wave operation at Peltier temperatures, would be advantageous, with new theoretical designs and materials beginning to address this [140]. The temperature performance of THz ISB detectors has been limited in contrast to their MIR counterparts, although recent work has shown an important improvement in operation temperature when the device is coupled to metamaterials, which reduce the effect of parasitic dark currents [131]. This can be improved by bringing similar concepts to quantum cascade detectors or even interlevel transitions in quantum dots with increases in temperature beyond liquid nitrogen temperatures, permitting the use of accessible cryocoolers. There is also potential to realise ISB detector arrays, offering a sensitive and fast alternative to microbolometer arrays for real time imaging applications.

ISB devices operating in the 5–12 THz region can be engineered in less conventional material systems as Si-SiGe, where lasing and detection have yet to be demonstrated in compact devices. However, the first demonstrations of THz ISB absorption in SiGe and in ZnO have been recently shown below 5 THz [141] and 8.5 THz [142], respectively, that hold promise for the future as the materials develops. The giant nonlinearities from these ISB materials could also be used in conjunction with existing THz and MIR QCLs as efficient nonlinear optical pump sources to realise emission in the 5–12 THz region through sum or difference frequency generation. Unforeseen opportunities for THz ISB-based devices could also be realised in new heterostructures based on 2D systems such as graphene, transition-metal dichalcogenides or topological insulators that possess distinct dynamics and phonon spectra when compared to traditional semiconductor materials.

Beyond laser and detectors, THz ISB amplifiers based on metasurfaces offer a promising solution (especially in a multi-pass geometry) as they do not depend on anti-reflection coatings and offer amplification of, for example, THz pulses where the frequency response drops off rapidly at high frequencies. High frequency THz modulators could also be realisable by combining adapted radio frequency waveguides with Stark effect modulators for high bandwidth telecommunications. New types of ISB devices could also be realised by leveraging advances in light-matter interactions, where metamaterial-based materials have enabled the manipulation of quantum states in intersubband devices in the strong coupling regime.

THz ISB devices would benefit from new types of resonators and waveguides. The current state-of-the-art is typically based on metal-metal geometries, permitting strong modal confinement, but usually at the cost of optical losses resulting in low Q factors (~ 10). Cavities based on dielectric confinement would potentially realise high Q-factor cavities but are challenging to grow using existing growth technology owing

to long growth times. Alternatively, advances in processing as in silicon THz photonics could be leveraged to realise, for example, suspended structures and photonic crystals that would enable high Q factors to, for example, enhance light-matter interactions.

Concluding remarks

Bandstructure engineering in THz ISB devices has permitted the development of new types of powerful sources and sensitive detectors in the 2–5 THz range, with unique nonlinear and ultrafast properties. Through the potential advances highlighted above, further applications and functionalities of these devices will be found, including their use as compact systems

for applications ranging from imaging, spectroscopy, trace gas analysis, and quantum technologies. Over the next decade, however, the real excitement of the applications of ISB devices in the THz range, may arise from the fundamental science that can be unlocked through non-linear optical, pump-probe, and multi-dimensional spectroscopy, for which both the techniques and applications are still in their infancy, promising an enticing future for the control and manipulation of states in materials. Further, the energies, length scales and timescales at THz frequencies are ideally placed for exploiting the interaction of 2D, topological and magnetic materials with ISB devices. The opportunities arising from their integration with existing ISB promise a step change in designs of electronic and opto-electronic devices.

10. Laser-based terahertz sources

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Status

Since the laser first emerged six decades ago, laser technology has been applied systematically to the problem of producing terahertz radiation [143] (see figure 14). In the 1960s the non-linear phenomenon of optical rectification (OR) developed as a way of down converting visible or near-infrared pulses to the terahertz regime. In the 1970s optically-pumped far-infrared gas lasers flourished. With the advent of the femtosecond laser, photoconductive antennas (PCA) [144] and time-domain spectroscopy (TDS) [145] were developed in the 1980s. The 1990s witnessed the first quantum cascade laser (QCL) (see section 9). OR, PCA and QCL sources typically produce polarised radiation, advantageous in many applications (see section 4). This century has seen multiple advances in such areas as fibre-based systems, improved optical rectification schemes [146], air-based emission and detection (see section 8), and nanostructured antennas.

The status of laser-based relative to other sources is as follows. Non-laser-based terahertz sources include conventional thermal sources and various electronic schemes. Thermal sources tend to be inefficient and are, fundamentally, incoherent. Electronic schemes are realised in both the vacuum, such as free electron lasers (FELs) and gyrotrons and in the solid state, by high-electron mobility transistors. The former are large and expensive instruments. While they offer high performance, they are not practical for widespread or field use. The latter still struggle to move from the sub-terahertz to terahertz frequency, in spite of much research.

Importantly, pulsed laser sources have opened up the possibility of TDS, with a tremendous range of fundamental and practical applications. The pump-probe scheme of coherent emission and detection synchronously links emitter and detector, with tremendous gain in signal-to-noise, and permits the further enhancement of time-resolved TDS.

Future advances are on the horizon, even with mature laser-based systems, and there are active research programs pursuing these. An exciting development in FELs is the development of purpose-built terahertz-specific devices at user facilities providing easy access. QCLs are a very active area of research, with the promise of the ‘holy grail’ of a source of electromagnetic radiation—a compact, robust, inexpensive, monochromatic emitter. New approaches to harnessing intense optical pulses to produce terahertz radiation by either optical rectification or photoconductive antennas [147] include optimising the coupling of the pump radiation in and the terahertz radiation out; the conversion efficiency; and the lifetime of the emitter.

Current and future challenges

Photoconductive antennas are the mainstay of THz time domain spectroscopy techniques. These devices typically fail if either too high a bias is applied, resulting in too large a current (electrical failure) or if too much laser power is supplied (optical failure). Optimum operation without failure involves balancing these factors. Techniques such as pulsed operation, cooling, and innovative antenna design have addressed these challenges, but more development in this area would be beneficial.

For high peak power THz pulse generation, optical rectification is often the method of choice as conversion efficiency scales linearly with pump pulse energy. The search continues for new and better OR materials, that simultaneously have a high damage threshold, large nonlinear coefficients and broad phase matching for wide spectral content. Excessive optical pumping of OR crystals can lead to their failure, either through multi-photon absorption or thermal issues. Use of a gas as the medium avoids some of these [148].

Coupling of the pump IR beam with the resulting THz beam is another area where future advances are possible, as well as combining new materials with novel noncollinear phase matching geometries. A clear challenge is the ability to produce pulses in the 5–15 THz range with high conversion efficiency due to the presence of optical phonons and other absorption bands in most materials at these frequencies.

The intersubband lasers have shown rapid development. The challenge remains to have these operating at higher temperatures, specifically at room temperature, rather than at the low temperatures where they operate now. Higher-temperature operation is possible with an applied magnetic field, but this introduces additional technical issues. Ideally, room-temperature, zero-field application is desirable.

Difference frequency generation offers compact, tunable systems, and advances in pump laser technology, quite separate from advances in terahertz technology, offer more compact, stable, and reliable operation [149]. Further advances in photomixers might be expected to increase bandwidth.

Advances in science and technology to meet challenges

Laser-based THz generation depends critically on available pump laser sources. A current challenge for THz generation based on lasers is to either provide a low-cost source or provide a unique application justifying the cost. This is of particular importance to industrial applications (see section 20). Femtosecond Ti:sapphire lasers have long been the workhorse for pulsed THz generation but have remained comparatively expensive and maintenance intensive. Fibre laser-based sources have taken over for low-power TDS sensing applications due to their robustness and low cost. Femtosecond fibre lasers are available at communications wavelength of 1550 nm and 1030 nm. These wavelengths require engineered semiconductor materials with fast rise times, rivalling those of GaAs, which has traditionally been used at 800 nm wavelengths

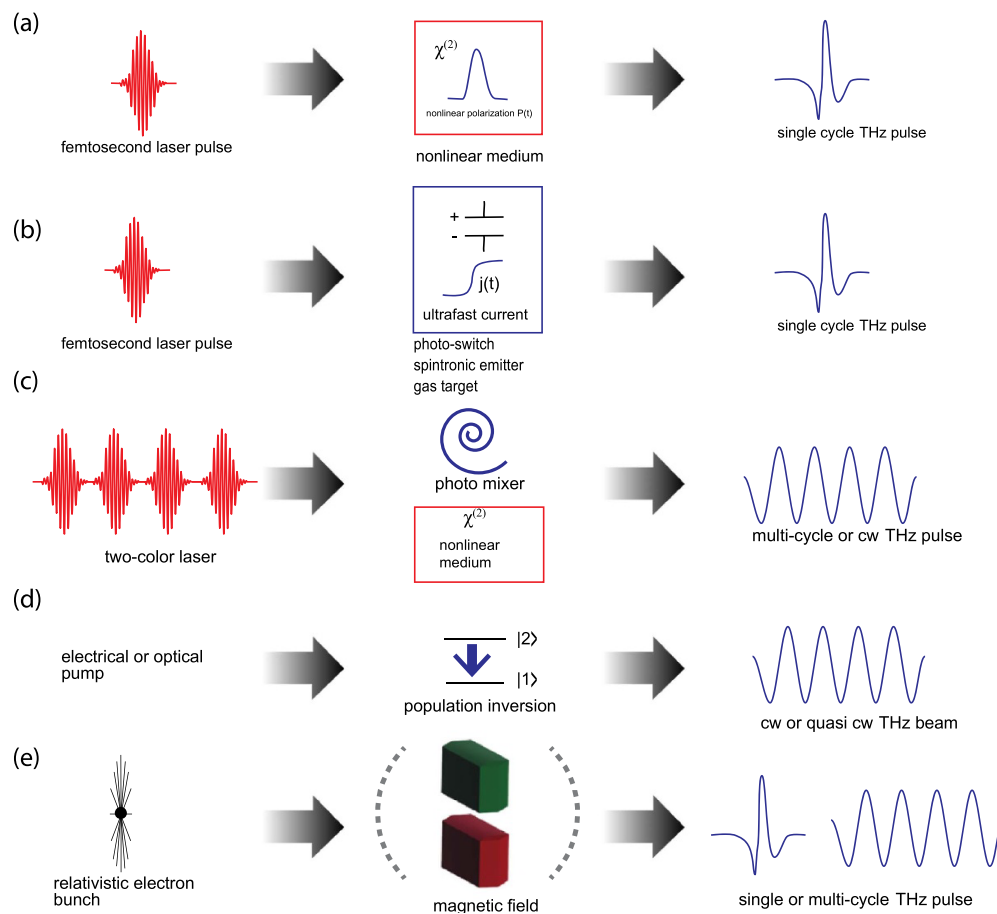


Figure 14. Overview of laser-based THz generation methods. Femtosecond laser pulses produce single cycle THz pulses via nonlinear optical processes like optical rectification (OR) (a) or THz driven ultrafast currents in photoconductive antenna (PCA), plasma or spintronic target (b). Coherent mixing of two-color lasers in photomixers or nonlinear media yields a cw or multi-cycle THz output through difference-frequency generation (DFG). (c). Direct THz emission from population inversion is achieved in intersubband and gas lasers or the quantum cascade laser (QCL) (d). THz emission from relativistic electron bunches in magnetic fields may be viewed as the analog of optical rectification (e). True lasing can be achieved when a magnetic undulator device is placed inside a cavity (dashed lines), resulting in a free electron laser (FEL).

[150]. Lasers equipped with nonlinear optical fibres enable pulse durations of just a few femtoseconds that can be used to generate ultra-broadband THz radiation.

In recent years great progress has been made with high average power high pulse energy laser systems. These are based on optical parametric chirped pulse amplification or spectral broadening and recompression and use picosecond pump lasers based on solid-state slab or disc laser technology. While record values for average THz powers and peak fields have been achieved [68], these laser systems remain at the forefront of technology. For practical applications of such THz sources the cost and complexity of high repetition rate, high pulse energy laser systems need to be significantly reduced.

On the materials front there has been renewed interest in nonlinear organic crystals for optical rectification. These materials were originally developed in the 1990s and exhibit extremely high nonlinear coefficients and broad phase matching. In combination with femtosecond pulses in the wavelength range of 1200–1500 nm some of the highest

conversion efficiencies for OR have been recorded. However, these crystals are expensive to manufacture and susceptible to thermal damage at high repetition rate. Improved thermal management through bonding to suitable substrates as well as active cooling may mitigate this problem. Another promising approach for broadband intense THz generation are spintronic THz emitters that are based on thin metallic films in an external magnetic field (see section 3). Magnetic, rather than electrical, dipoles offer a novel approach [151].

Concluding remarks

Lasers provide one solution to the problem of making terahertz radiation. *Direct laser emission* is well established but further development is now occurring: THz FELs being made more compact; QCLs operating at higher temperatures and lower frequencies; new emission schemes emerging for p-Ge and cognate devices; gas lasers continuing to be refined for niche applications. *Difference frequency mixing*

combines two visible or near-infrared CW lasers to construct a pseudo-monochromatic, tunable terahertz-frequency source. Advances are increasing the tuning range and power while further reducing the already small size and cost. *Optical pulse pumping* produces terahertz radiation through two mechanisms. OR has been demonstrated in a wide variety of targets,

notably recently in sundry gases, with power and bandwidth increasing year-by-year. PCAs continue to improve in reliability, efficiency, durability and bandwidth. Given this continuous development, we may confidently predict a future that is brighter and brighter—literally—for laser-based terahertz sources.

11. Terahertz emission spectroscopy and imaging

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Status

Terahertz (THz) emission spectroscopy (TES) relies on macroscopic photocurrents that emit a measurable THz pulse encoding the current dynamics (see figure 15(a)). It has become an essential tool to explore photocarrier dynamics upon femto-second (fs) laser illumination in advanced material systems. TES complements THz time-domain spectroscopy (TDS), which is often used to measure the complex THz refractive index of materials. By spatially scanning the pump-laser spot, one can gain local insights into the photocarrier dynamics with a spatial resolution that is only limited by the pump-laser-beam diameter. This technique is called laser THz emission microscopy (LTEM).

TES of novel materials. TES has been used to study semiconductors [153] and extended to various other materials [154, 155]. Figure 15 gives an example of TES on aligned semiconducting carbon nanotubes, where the amplitude of the THz emission follows that of the deposited pump energy and discloses the ultrafast dissociation dynamics of excitons [152]. Another important example is TES of spintronic heterostructures, which has recently received considerable interest [24]. It typically relies on ultrafast heating of a magnetically-ordered thin film that, thereupon, emits a spin current into an adjacent metallic thin film with strong spin–orbit coupling (see section 3). Spin-to-charge-current conversion, e.g. due to the inverse spin Hall effect, inside this second layer, converts the ultrafast spin into an in-plane charge current that emits THz waves. These examples demonstrate the large potential of TES to study even more advanced material systems in the future.

Spatially resolved insights. LTEM provides the opportunity to visualize an ultrafast photo-response spatially resolved. Examples are the impurity distribution of wide-bandgap semiconductors, quantitative supercurrent distributions in high- T_c superconductors, the domains of spontaneous polarization in ferroelectrics, and localization of the defects in large-scale integrated circuits [156].

Recent LTEM developments aim at nanoscale imaging with similar approaches as used in a terahertz scanning tunneling microscope (see section 12). In 2017, a nanoparticle was imaged by nanoscale LTEM (nano-LTEM) with

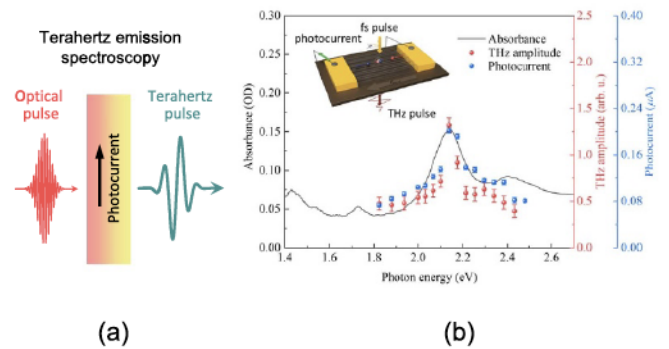


Figure 15. (a) Principle of TES. An optical pump pulse generates a photocurrent, which gives rise to the emission of a THz electromagnetic pulse. (b) The absorbance spectrum (black solid line) for an ordered carbon nanotube film plotted along with the THz-emission amplitude (red spheres) and photocurrent amplitude (blue spheres) as a function of photon energy. An external electric field was applied along the nanotubes. Inset show the schematics of the CNT-based photoconductive antenna. Reproduced from [152]. CC BY 4.0.

a record spatial resolution of approximately 20 nm using a scattering-type scanning near-field optical microscope (s-SNOM) [157]. Until now, the use of nano-LTEM has been very limited, including a recent study that provided a detailed description of how the probe geometry affects the waveform emitted from the sample [158].

Current and future challenges

Retrieving the photocurrent dynamics. Why are TES and LTEM not as prevalent as THz-TDS is in general? A major reason could be the lack of a straightforward inversion procedure that allows one to relate the recorded TES or LTEM data to the photocurrent dynamics inside the materials of interest. One possible route is, therefore, to widen the studied sample systems to build a comprehensive model toolbox. A robust understanding of how local currents act as sources of THz pulses in homogeneous samples including a separation of bulk vs surface contributions is needed. Thereby, advancements in LTEM to study inhomogeneous samples locally would come into reach.

Enhancing spatial resolution using (spintronic) LTEM. Very thin THz emitters such as a spintronic THz emitter could potentially realize a spatial resolution far below the THz wavelength via a spintronic LTEM or a near-field imaging approach [159]. A spintronic LTEM could allow one to study the local spintronic THz-emission dynamics that encodes the static and dynamic magnetic ordering such as magnetic domains [160] or skyrmions, the nature of the spin current (electrons or spin waves) [161], or spin–orbit-interaction strength.

The nano-LTEM should be applied to study local information in a wider range of materials such as 2D materials and

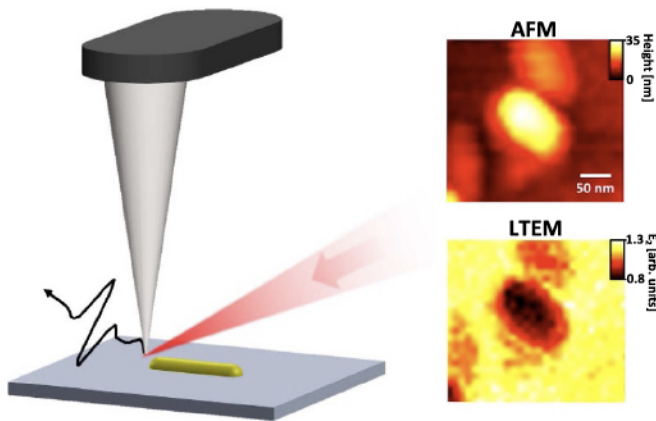


Figure 16. Illustration of nanoscale LTEM where a near-infrared laser beam is focused below an atomic force microscopy (AFM) probe approached to a sample. Here, the sample is a gold nanorod on an InAs substrate, where the AFM probe facilitates the nanoresolved THz emission originating from the substrate. By raster-scanning the sample, AFM and LTEM images are simultaneously obtained. The spatial resolution is defined by the curvature of the used AFM-tip apex, which is ~ 20 nm for the images shown here. Reprinted with permission from [157]. Copyright (2017) American Chemical Society.

semiconducting nanostructures. The main limiting factor for nano-LTEM is the currently still poor detection sensitivity preventing the ability to measure weaker signals. Strong coupling between the radiated THz field and a resonant probe has until now been critical for detecting a THz near field signal in an s-SNOM experiment, as shown in figure 16. The nano-LTEM would advance with the development of more sensitive THz-TDS detection methods.

Theory development. One of the challenges for TES, LTEM, and nano-LTEM is the development of useful models covering a wide range of scales and materials. The photoexcited carriers travel on atomic scales, the photons excite the carriers in the electronvolt range, and the THz waves have photon energies in the millielectronvolt range. This multitude of relevant scales makes it very difficult to capture the entire system response even using a supercomputer [162]. Thus, we need new approaches that combine precise, yet expensive models and potentially cheaper and, ideally, more insightful phenomenological models.

Envisioned novel applications. The latter theory developments could allow for fast data pre-processing that would be key to on-site industrial-scale applications of TES and LTEM. In this area, a prime envisioned application of TES or LTEM is to quickly monitor various parameters of semiconductors, such as surface potential, spintronic properties, impurity doping density, defects in passivation layers, or surface states by tracking the emitted THz radiation [163].

Advances in science and technology to meet challenges

TES and LTEM are emerging methodologies and much progress is still needed. In terms of ultrafast photo-response dynamics, a dynamic LTEM that relies on an optical-pump/TES-probe technique can provide spatially local information (see examples above) following an ultrafast pump-pulse excitation.

Foreseen advances in (spintronic) LTEM. For the proposed spintronic LTEM technique, structured pump light [161] promises a massive parallelization and thus greatly reduced acquisition times as well as making the need for actively scanning across the studied sample obsolete. Regarding nano-LTEM, many efforts are being devoted to improve the sensitivity of the required s-SNOM THz detection, but the exploration of fundamentally new concepts seems necessary. Moreover, nano-LTEM would benefit from the development of ultra-stable high-repetition-rate femtosecond lasers increasing the number of the detected near-field photons, and, thereby, the signal-to-noise ratio. Furthermore, the development and better accessibility of geometrically well-controlled near-field probes and/or photoconductive-detector-integrated probes are needed for a more reliable quantitative extraction of local sample parameters at THz frequencies. In general, TES and LTEM will greatly benefit from advances in the field of THz-TDS.

Foreseen advances in theory. The above-mentioned advances cannot be reached without a deeper understanding of the underlying physics over different time scales from femto- towards microseconds as well as different length scales from nano- to millimetres. Tackling this challenge requires strongly enhanced theoretical methodologies and presumably the use of supercomputers. Such developments would enable innovations in a wide range of fields far beyond the THz community and eventually deepen our understanding of ultrafast light-matter interaction.

Foreseen impact on novel applications. To realize large-scale real-world applications of TES and LTEM, the recorded data and images need to be massively pre-processed to allow for an easy user interpretation. Such simplification strongly depends on the material properties and parameters that are of interest. Possible examples are diverse and include picosecond anisotropic carrier transport, acoustic-phonon propagation, ultrafast dynamics of spintronic devices, and ultrafast local optical heat dissipation in metals.

Concluding remarks

TES encodes the ultrafast photo-carrier dynamics upon fs laser illumination in the THz-emission waveforms. LTEM provides the images of such photo-response at the resolution of the

optical pump beam diameter, which enables new advances in material science and device development. For instance, using spintronic THz emission to obtain local information of the emitting material is still largely unexplored although it promises a manifold of insights into complex magnetic systems. With technological advances in the femtosecond laser technology, sensitive THz detection, and resonant s-SNOM probes, nano-LTEM is a promising technique for obtaining

carrier information on the nanoscale with femtosecond temporal resolution. Models that integrate the precise ultrafast carrier dynamics at the atomic scale with phenomenological approximations covering larger temporal and spatial scales will support such progress. With these advances, large-scale applications such as in the field of semiconductor R&D or in spintronic memory technology [24] will eventually come into reach.

12. Terahertz scanning tunnelling microscopy

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Status

Electrons, atoms and molecules are the elementary building blocks of all condensed matter; they move and interact according to quantum mechanics and form complex systems that bring out a boundless variety of functions. Directly observing this vibrant nanocosmos in true microscopic videography and relating it with macroscopic functionalities has been an ultimate dream of modern sciences as well as nano-, quantum- and bio-technologies. However, the bar is set high, with intrinsic dynamics occurring at length and time scales as short as angstroms and femtoseconds or less.

The recent combination of THz technology with scanning tunnelling microscopy (STM) [164–173] has resulted in the first-ever—and currently only—approach allowing for atomic-scale single-molecule ultrafast movies [165, 166]. To this end, the usual static electric bias of the tip-sample junction has been replaced with the transient voltage created by the oscillating carrier wave of a free-space THz pulse coupled into the tip [164]. The locally enhanced THz field can be regarded as a strong-field quasi-static bias producing sub-ps current spikes. The nonlinear current–voltage characteristic causes a rectified net current, which can be read out using conventional STM electronics. This idea has been successfully employed with large numbers of electrons per laser shot, to obtain sub-ps temporal and sub-nm and even atomic spatial resolution [164, 167, 171].

For actual videography of single-electrons, another key ingredient is required: state selective tunnelling. As demonstrated with low-temperature STM of a single molecule [165], the junction can be tuned such that only the peak of a THz waveform opens the sequential tunnelling channel through a select molecular orbital (figure 17(b)). Thereby the field controllably removes (adds) a single electron from (to) a specific orbital facilitating snapshot images of the orbital with a strongly subcycle temporal resolution of ~ 100 fs. Pump-probe experiments further reveal coherent molecular vibrations at THz frequencies directly in the time domain and with sub-angstrom precision [165]. Furthermore, the THz near-field in the junction can be used as a femtosecond atomic force to control structural dynamics while leaving the system in its electronic ground state [166]. First experiments with optical excitation of electrons in molecular

films have produced an electron movie featuring the picosecond spreading of photoexcited carriers in a potential landscape [172].

Current and future challenges

THz-STM is a very young field that has inspired a rapidly growing community to explore an increasing variety of systems—from single molecules to quantum materials. New challenges have arisen in matching the latest breakthroughs to material-specific questions. (see figure 18). *Optical pumping* is desirable, as it enables selective driving of electronic excitations, collective modes (phonons, magnons etc), and phase transitions. THz-STM can then image the motion of individual atoms, charges and spins, or the dynamics of symmetry breaking and temporally evolving hidden or light-driven metastable phases. Another frontier is to *push the temporal resolution* of THz-STM to few femtoseconds or better, to follow yet faster dynamics, ranging from lattice vibrations to electronic wave-packet motion. This goal may be achievable by transient biasing with multi-THz and infrared pulses, but rigorous studies quantifying the role of tip thermal expansion are required. Controlling spin-selective tunnelling with *external magnetic fields* could help tackling long-standing problems in ultrafast magnetism (section 3). Besides microscopy, THz biasing could also extend *tunnelling spectroscopy* to ultrashort time scales. The derivative of the THz-induced current with respect to the THz peak field can be used to extract a snapshot of the sample's density of states, enabling ultrafast energy tracking of quantum states and their occupation.

Furthermore, the uniquely direct access of THz-STM to the quantum dynamics of individual particles opens conceptually new opportunities: While THz-STM has hitherto concentrated on mean numbers of transferred electrons, the technique is inherently capable of exploring their number fluctuations or charge fractionality. This novel approach may sensitively probe non-classical and topological states in matter (section 7). Even the quantum nature of the electromagnetic field in the strongly enhanced near-field region of the tip may become relevant (section 13). Furthermore, electronic quantum *correlations in the tunnelling dynamics* could become observable. On ultrashort timescales, two sequentially tunnelling electrons are no longer independent and affect each other's tunnelling probability via quantum interference and back action. Positioning another STM tip several nanometres away could ultimately give access to complete spatio-temporal correlations, which could be used as a unique quantum spectroscopic tool. Finally, *many-body states* such as excitons or Cooper pairs may be accessed with THz photon-assisted or dynamically driven tunnelling. For instance, consecutive THz half cycles of opposite polarities could inject one electron and one hole, respectively, creating an exciton. Precise control over THz near-fields and quantized electron tunnelling could lead to a unique spectroscopic tool for quantum materials.

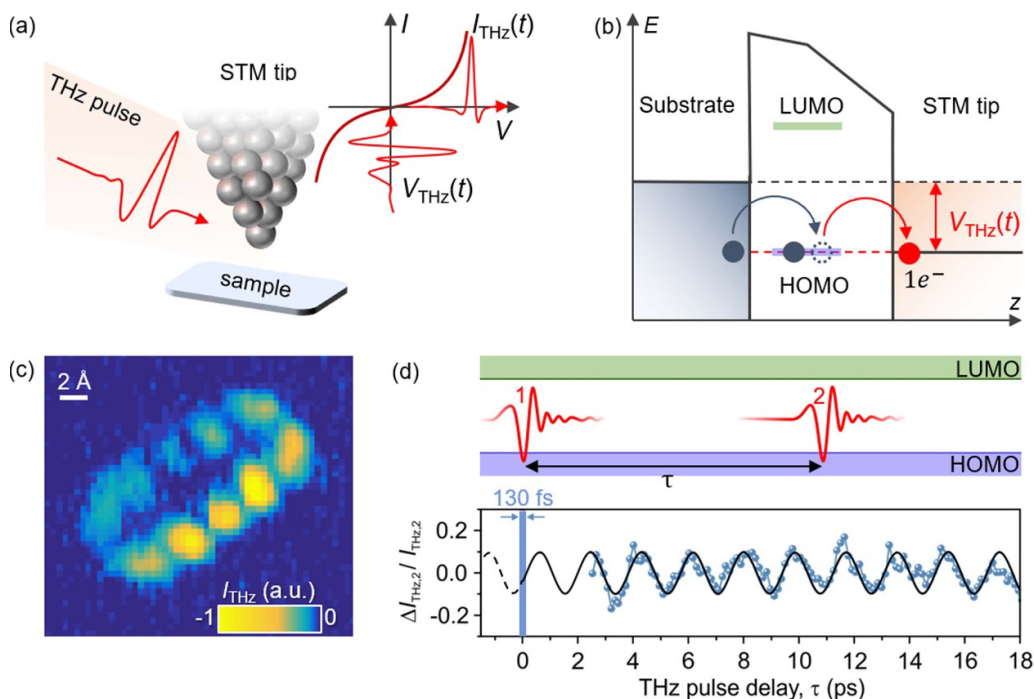


Figure 17. (a) Schematic of THz-STM: an intense THz pulse is coupled to an atomically-sharp tip, resulting in an ultrafast transient voltage $V_{\text{THz}}(t)$. Inset: Ultrashort THz current $I_{\text{THz}}(t)$ pulses produced by $V_{\text{THz}}(t)$ with the rectifying nonlinear I - V curve of the tunnelling junction. (b) State selective sequential tunnelling in a molecule: $V_{\text{THz}}(t)$ transiently biases the tip and removes a single electron from the doubly occupied molecular orbital (red arrow). The circuit is closed by an electron from the substrate, filling the transiently single-occupied orbital (blue arrow) once the THz pulse is gone. (c) Sub-angstrom THz-STM image of the HOMO of a single pentacene molecule adsorbed on few NaCl layers on Au(110) [165]. (d) Pump-probe THz-STM. Top: a first THz pulse charges the molecule by removing an electron from the HOMO, changing its adsorption distance. A second pulse probes its instantaneous vertical position. Bottom: time-resolved variations of the THz-induced tunnelling current reveal the oscillations of the pentacene molecule in its adsorption potential [165]. Reproduced from [165], with permission from Springer Nature.

Advances in science and technology to meet challenges

For most of these challenges, knowing the exact THz near-field in the tunnelling junction is of paramount importance. This goal has indeed been achieved using hot-electron field emission [169, 170] or single-molecule voltage gauges [168]. Also, laser development continues to be in focus: with just one electron transferred per THz pulse, the tunnelling current is limited by the laser repetition rate, and stable THz sources with repetition rates in the MHz range [173] have been in high demand. New large bandwidth sources [174], such as quantum well (section 9) or spintronic emitters [169] alongside tips supporting ultrabroadband near-field coupling will pave the way towards yet higher temporal resolution. It will be interesting to see how increasing driving frequencies morph quasi-static tunnelling into dynamic wave packet motion and, ultimately, perturbative multiphoton excitation (section 5). In such THz-STM studies, thermal tip expansion caused by optical absorption deserves particular attention because sub-angstrom changes of the tunnelling gap can translate into large parasitic currents. This problem can be effectively mitigated by keeping the optical power constant as evidenced by recent optical pump—THz probe experiments [172].

Pure THz-STM may not always sensitively discern competing orders in quantum materials, since tunnelling inherently

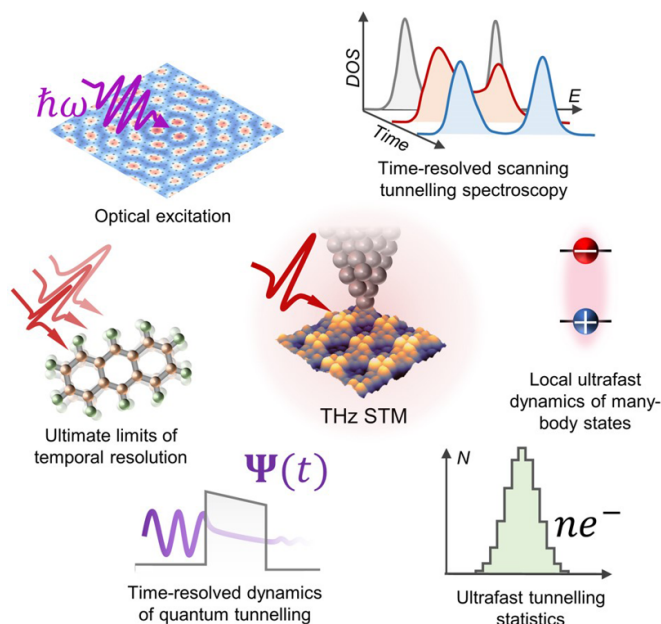


Figure 18. A kaleidoscope of exemplary challenges illustrating the roadmap for THz-STM.

discriminates between single-particle rather many-body states. Employing recent advances in STM like magnetic, superconducting, molecule-terminated tips or tips brought in

close proximity [175], THz-STM could resolve some of these issues, but data interpretation would require theoretical advances. An attractive alternative is to complement the measurement of the tunnelling current with information carried by photons emitted in tip-induced electroluminescence [176] or the THz near-field scattered from the atom-scale tunnelling junction, converting THz-STM into a *multi-messenger microscopy* platform. Scattering-type near-field microscopy (section 13) can probe the local polarizability; both techniques combined could image the local density of states and the dielectric response simultaneously and thus, e.g. dynamically distinguish itinerant and localized states on the nanoscale. THz-STM and its newly emerging, ambitious varieties stands on the shoulders of a giant, namely STM itself. It is encouraging to see that employing these novel concepts in the investigation of quantum materials can often benefit from the extensive experience gained in conventional STM—only adding a decisive variable that characterizes the nanoworld in motion: time.

Concluding remarks

THz-STM has transitioned from a breakthrough technological achievement, into a vibrant and sustainable field. While it is still the only technique capable of taking single-electron ultrafast movies with atomic resolution, it is set out to expand by exploring the ultrafast dimension of conventional STM techniques and the spatial dimension of emergent ultrafast phenomena. Advances in laser and THz source development will determine the ultimate time and energy resolution. Improved signal-to-noise ratio will eventually enable the detection of individual ultrafast tunnelling events, thus uncovering the emergent capacity of THz-STM as a quantum sensing technique. The stunningly direct access of THz-STM to the atomic world may enable us to steer (bio)chemical reactions, and ultrafast phase transitions and to eventually gain full control of the fundamental building blocks of physics, chemistry and biology.

13. Near field THz imaging

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Status

Combining terahertz (THz) radiation with scanning near-field optical microscopy (SNOM) has opened new opportunities to explore THz phenomena at length scales below the diffraction limit down to the nanoscale. Recent years have seen growing success in the application of SNOM for probing evanescent fields confined to surfaces and the local material properties of a range of condensed matter systems, including spatial mapping of ultrafast plasmon phenomena in graphene heterostructures [177], interlayer charge transport and exciton–polariton propagation [178], and spectroscopy of single THz resonators [179]. Whereas far-field techniques are restricted in spatial resolution by the diffraction limit, SNOM overcomes this limit by detecting evanescent fields localised around sub-wavelength structures and therefore can access deeply sub-wavelength length scales. Several THz SNOM configurations have been developed since the first demonstrations over 20 years ago. For the purpose of the Roadmap, we will group them by near-field probe type: SNOM with direct near-field detection (d-SNOM) and SNOM with scattered wave detection (s-SNOM).

d-SNOM is realised by introducing a subwavelength size THz detector or aperture into the near-field region of the sample. A range of THz detectors, compatible with CW and pulsed THz sources and covering practically the entire THz band, have now been integrated into the near-field probes for improved sensitivity, spatial resolution, and phase-sensitive detection [180]. These techniques enabled THz imaging with an intermediate level of spatial resolution (2–30 μm) and have been reliably applied for mapping and spectroscopy of THz fields in THz waveguides, plasmonic and dielectric resonators, and THz metasurfaces. In a slightly different approach, the detector in proximity of the sample can be replaced with a subwavelength-size source of THz radiation, and a high-resolution THz image of the object can be formed by scanning the source with respect to the sample. Furthermore, the use of spatial light modulators instead of localised THz sources recently eliminated the need for raster scanning [181].

s-SNOM achieves subwavelength spatial resolution by introducing a sharp tip of a probe within tens of nanometres of the sample surface. The spatial resolution is determined by

the radius of curvature of the probe, typically on the order of ~ 10 nm, which has enabled spatial mapping of the local dielectric function of individual nanostructures on nanoscale length scales. This approach has been combined with both emission microscopy (section 11) and scanning tunnelling microscopy to enable imaging of coherent molecular vibrations within single molecules (section 12). It has also been employed with free-space electro-optic sampling and photoconductive antennae for time-resolved electric field detection on ultrafast timescales (~ 10 fs) [182]. Phase-resolved near-field detection has also been demonstrated via self-mixing in a THz quantum cascade laser (THz-QCL) coupled to s-SNOM [183], allowing narrowband operation with high-field sources at frequencies beyond 2 THz for selective excitation of THz resonant modes [139].

Together, these techniques form a powerful toolkit for near-field THz characterisation, providing access to length regimes spanning over four orders of magnitude and specific probing functionalities. For example, d-SNOM enables vectorial field mapping with minimal probe invasiveness and allow fast scanning speeds of large areas. Conversely, s-SNOM typically scans smaller areas ($10\mu\text{m}^2$) and requires more elaborate signal processing to suppress background artefacts, yet it offers superior spatial resolution and surface sensitivity. There are also important differences between the two approaches: while s-SNOM signals can be correlated to the dielectric function of the sample, d-SNOM measurements tend to correlate with THz fields.

Current and future challenges

Many of the challenges facing near-field THz imaging and spectroscopy are inherent to the long wavelength of THz radiation. For example, confining THz radiation to deeply sub-wavelength scales leads to extremely low light coupling into aperture probes and much lower scattering efficiencies for scattering probes compared to the MIR range. This poses challenges for realising all the opportunities THz-SNOM can offer.

Nonlinear near-field spectroscopy has yet to be achieved in the THz and MIR range, despite its enormous potential for opening up a pathway for observation of nonlinear THz phenomena and temporal dynamics, such as light-induced lattice vibrations, phase transitions and high harmonic generation, at the nanoscale. The difficulty stems in part from challenges associated with the operation of high-power THz sources but also due to probe heating and weak sample-probe THz interactions. Nonlinear microscopy would also benefit from pump-probe configurations and field-resolved detection, which is challenging due to the low scattering efficiency for THz radiation. Furthermore, high-field THz sources usually operate at repetition rates much lower than the probe tapping frequency, so more complicated sampling techniques are required.

Polarisation-resolved near-field spectroscopy is also an essential tool for identifying modes in THz resonators and waveguides, probing surface waves and mapping material anisotropy. While d-SNOM techniques allow vector-field

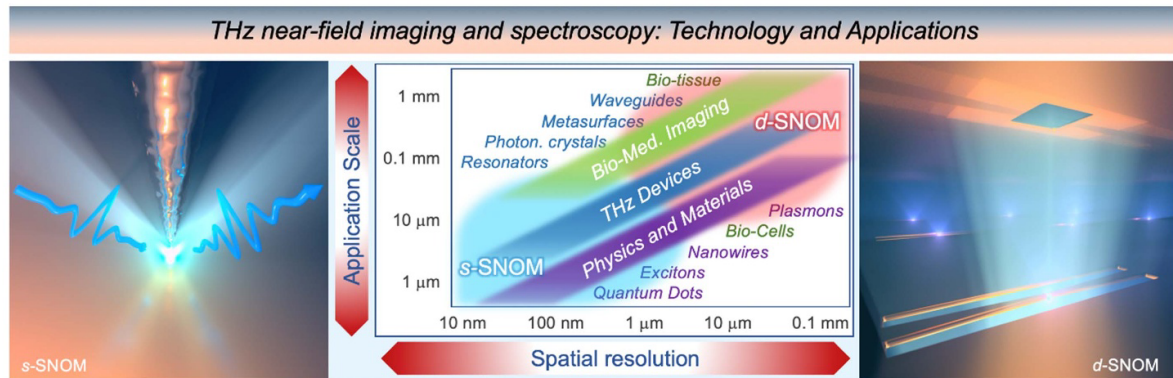


Figure 19. Application space of THz near-field imaging and spectroscopy techniques highlighting the achievable spatial resolution and application scale with example applications (Centre). Artistic illustration of s-SNOM (left) and d-SNOM (right) techniques.

mapping, in some cases the different field components can be mixed (e.g. within the near-field probe), so their decomposition requires post-measurement analysis. In contrast, s-SNOM usually employs excitation with vertical polarisation for more efficient coupling with the tip. While it is sensitive to both in-plane and out-of-plane components, the in-plane components are significantly suppressed.

Another key challenge for pursuing new studies of THz phenomena on the micro- and nanoscale is the lack of accessibility to THz-based SNOM systems. While commercial systems are now available, modular ‘turn-key’ options in the THz range are in their infancy. Current systems are expensive and often require bespoke design for THz and pump-probe operation. While significant progress has been made towards cryogenic near-field studies [177, 184], cryogenic systems are still only offered by one company and suffer from issues with vibration isolation. As a result, the THz community tends to either use home-built THz microscopes and near-field probes; or couple THz sources and detectors to commercial SNOM or atomic force microscope units, restricting their widespread use.

Advances in science and technology to meet challenges

To address these challenges, an increase in detection sensitivity of near-field THz microscopy is desirable. Utilising high-field THz sources could increase the signal-to-noise ratio (SNR) and enable nonlinear spectroscopy, yet they currently operate at low repetition rates and are limited by Nyquist-Shannon sampling. New sampling techniques, such as phase-domain sampling [185], can expand THz-SNOM to the high-field THz regime. Development of high-field, high-repetition THz sources would also improve the SNR and enable field-resolved, phase-sensitive detection for pump-probe spectroscopy. Alternative sources, such as THz QCLs (section 9) could also contribute to these respects.

Further structural near-field probe engineering is essential. Integration of nanoscale THz detectors with aperture probes has already provided gains in detection sensitivity. Resonant THz antenna tips have also been shown to increase scattering

efficiency and field enhancement compared to commercial AFM tips [186–188]. However, commercial scattering probes are currently not designed to support $\lambda/2$ resonances in the THz range. In the MIR range, tips coated by metallic nanoparticle aggregates that support localised surface plasmon resonances have been introduced to improve both sensitivity and spatial resolution (nexttip). A similar approach could be applied in the THz range and lessons in tip design learnt from STM.

Underpinning both these future developments is a quantitative understanding of sample-probe THz interactions. The probe geometry, conductivity of the probe, sample topography, resistivity and electrostatic environment can all affect the coupling between the probe and sample [189, 190]. This can provide artefacts in the observed near-field contrast and poses challenges for studying nanoscale nonlinear THz light–matter interactions. Several analytical models have been employed, however, they do not always completely replicate experimentally-measured response—in particular, tip-induced phase shifts of the scattered electric field measured in the far-field. FDTD simulations have provided further insight yet are time-consuming. An extensive literature collection comparing complimentary SNOM-based techniques (i.e. emission and pump-probe spectroscopy) and models (i.e. finite-dipole, point-dipole, FDTD) would therefore be useful for providing a reference for home-built systems.

Ensuring wide access to THz SNOM systems would further strengthen understanding of sample-probe interactions and development of nonlinear and polarisation-sensitive THz near-field microscopy. Offering AFM parts, aperture probes and near-field nanoscale detectors that can already integrate with home-built systems could reduce system complexity and cost. At the same time, access to more expensive cryogenic systems could be provided via (and in conjunction) with national facilities (e.g. FELIX) to facilitate near-field studies at new extremes.

Concluding remarks

Near-field THz microscopy has become a powerful tool for hyperspectral imaging of THz light-matter interactions

beyond the diffraction limit. In recent years, significant step-changes have occurred, including availability of commercial cryogenic systems and time-resolved and phase-sensitive detection, that are enabling THz investigation at three extremes: cryogenic temperatures, ultrafast timescales

and nanoscale length scales. One next challenge is exploration of the nonlinear regime. By employing structural probe engineering and high-field THz sources, nonlinear near-field THz microscopy could also be realised, paving a way to investigate nonlinear THz phenomena on nanometre length scales.

14. Terahertz instrumentation for satellites

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Status

Sensing terahertz (THz) radiation from space platforms has been predominately directed towards underpinning research associated with astrophysics and atmospheric remote sensing. In complementing and extending ground-based and airborne observations, the scientific information gained plays a pivotal role in expanding knowledge and understanding of the cold and dark interstellar medium (ISM) that harbours early star formation [191], delivers a unique view (figure 20) of the cosmic microwave background [192], enhances awareness and understanding of the impact of anthropogenic activities on Earth's climate [193], and provides *in-situ* observations of solar system objects.

Free from the Earth's atmospheric attenuation, space provides a vantage point to access regions of the THz spectral range that are otherwise inaccessible. It allows chemical species to be measured and characterised within the ISM's giant dust and molecular clouds, and aspects of Earth's atmosphere that strongly influence weather and climate. Passive THz instruments, for example, supply critical information on global atmospheric pressure, temperature and humidity for numerical weather prediction and there is a trend from large multi-instrument platforms, e.g. the MetOp Second Generation satellite [195] series, towards smaller, lower cost, shorter development schedule, missions such as TEMPEST-D [196] and TROPICS. Thus, whilst the Earth-to-orbit transition is a difficult, costly and risky procedure, with instruments sometimes spending many years in preparation, the returns are considerable and the data gained is of great importance in furthering scientific knowledge. In the case of instruments providing data for weather forecasting, there is also significant societal and economic benefit. Additionally, there exists increasing interest in, and potential, for ultra-high data rate intersatellite communication that will allow rapid data transfer between constellations performing, for example, Earth observation or delivering future global telecommunication services.

Spaceborne THz deployments have been relatively few and instruments operating above 1 THz are even rarer [193]. This primarily arises from the relatively immature status of payload technology, though advancements are being made that increase the potential for THz observation in relation to both Earth observation and astronomy [197, 198]. When compared with optical counterparts, THz structures are large and spatial imaging capabilities are inferior. Nevertheless, the case for science exploration missions is strong and the spectral range provides commercial exploitation potential with direct benefit to society at large through, for example, weather prediction services and future high-data-rate communications [199]. Substantial motivation therefore

exists for new THz space instrument development in the form of both passive and active (radar) detection instruments. These will require payloads located on individual satellites and free-flying constellations located in low Earth orbit (LEO) [196], planetary probes and observatories located in deep space and, in the case of high-speed data links, technical evolution that provides source and detector technologies with necessary power output and sensitivity capabilities.

Current and future challenges

Developing satellite THz instruments provides technical challenges that are often mission specific. For instance, a scientific explorer with objectives of sensing weak signals emanating from within our galaxy or from extragalactic origins, requires cryogenic systems to minimise device and background noise [192, 200, 201]. To achieve adequate angular resolution and/or flux collection, large scale (>1 m) primary antenna apertures are often necessary. Conversely, payloads that are destined to traverse our solar system en route to giant planets may require neither cryogenics nor large antennas. However, the journey time necessitates long periods of instrument hibernation followed by reliable operation within a different and potentially hostile environment, e.g. Jupiter's radiation field [202]. Similarly, high-speed data relays in LEO will require reliable source and detector instrumentation in configurations that operate at or near to room temperature in order to minimise deployment costs.

However, and as a generality, all face a common challenge of minimising the use of scarce resources of volume, mass and power provided by a specific satellite platform, i.e. satellite bus. It matters not if the mission is located in low Earth orbit, orbiting a more distant Lagrange point, nor traversing the interplanetary regions of our solar system; all must work reliability within stringent payload constraints. Added to this is consideration of mission cost, which encompasses phases of development, build, qualification, launch and ground segment, i.e. operations and data acquisition, preparation and delivery. Current and future THz satellite instruments must therefore address all of these issues very effectively if they are to progress.

Unfortunately, THz payloads are relatively large, massive and often power hungry. Power may be needed to operate coolers for cryogenic detectors for astronomy, or scanning antennas and higher resolution spectrometers needed for improving Earth observation, or beam steering and signal processing electronics necessary for telecommunications. There have been notable enhancements with respect to these attributes, but, and paradoxically, the gains made are offset by more ambitious user objectives associated with next generation missions and services. Thus, respective areas of THz component and systems integration technology will need to advance further by achieving greater compactness, increased component power efficiency, and lower implementation cost. As an example, future constellation systems providing rapid global response THz weather data services will use

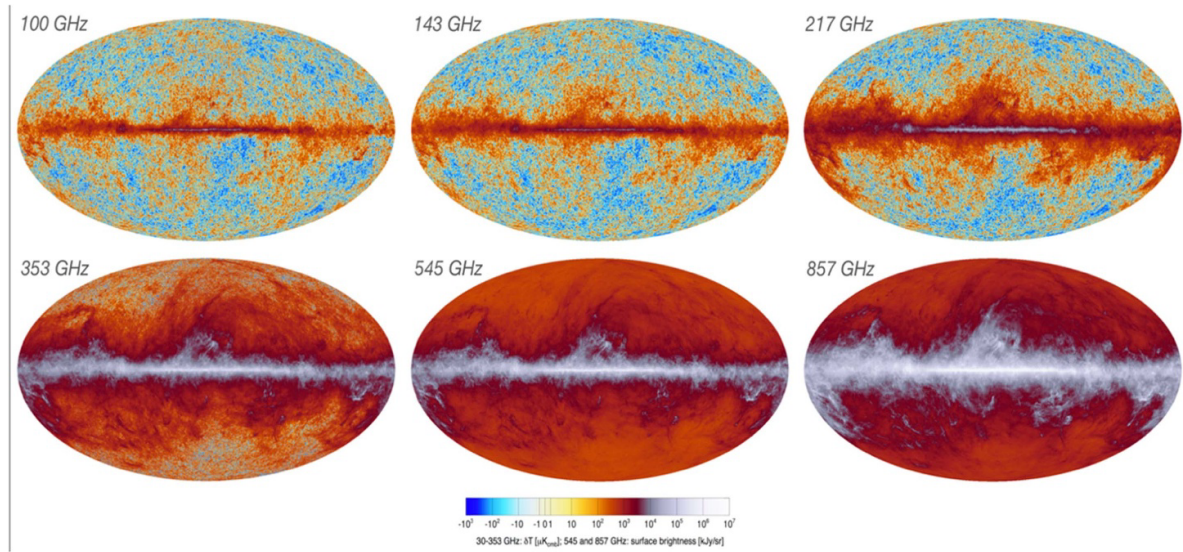


Figure 20. ESA Planck payload observations at different THz frequencies of the cosmic microwave background. The images reveal small temperature anisotropies related to the origins of the observable universe. Image credit: ESA and the Planck Collaboration [194]. Reproduced with permission from [194]. F R Bouchet, A&A, 641, 4, 2020, reproduced with permission © ESO.

microsatellite (CubeSat scale) buses that possess highly constrained resources, e.g. <100 W of power and typically cm^3 volumes [196].

Advances in science and technology to meet challenges

Improving performance, while lowering volume, power consumption and cost, presents a rich area for advancement. This encompasses not only the core THz solid-state semiconducting, superconducting and vacuum devices needed for passive/active sensing and telecommunication purposes, but also system engineering architectures.

THz payload size is often dominated by the need for a large primary antenna that impacts satellite bus and launch vehicle selection and costs. Advances in THz antenna reflective surface materials, mechanical structural supports and deployment mechanisms allow stowage during launch and in-orbit deployment [203]. Related developments are relevant not only to LEO microsats, but also largescale systems where multi-metre class apertures are required. Alternative approaches that utilise satellite formation flying, or single satellite sparse arrays to form interferometers, also require advancement in THz signal distribution to ensure receiver phase coherence and positional control.

The components that form THz systems are selected in relation to the mission's observational objectives. They may take the form of a single pixel, phased or multi-pixel coherent/incoherent focal plane arrays. For instance, THz high-resolution spectroscopy, radar and telecommunication systems employ coherent detection methodologies that require a range of THz components including low noise and power amplifiers, frequency mixers, and local oscillators (figure 21). These devices utilise semiconductor materials such as GaAs, GaN and InP. Continued refinement is needed to support

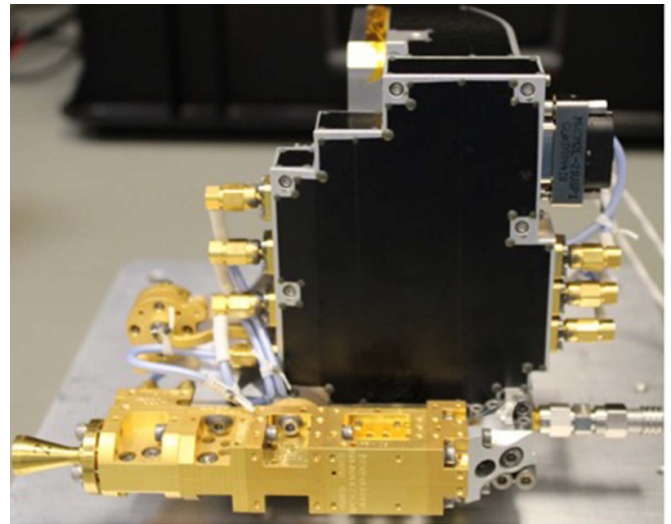


Figure 21. Spaceflight qualified 166 GHz heterodyne radiometer for the MetOp-second generation series of operational meteorology satellites. The volume of $120 \times 120 \times 106 \text{ mm}^3$ incorporates a low noise amplifier, sub harmonic Schottky diode mixer and local oscillator system.

better detection sensitivity at increasing frequencies, efficient THz signal generation, and reduced power consumption. Furthermore, advances in superconducting materials will substantially benefit coherent and incoherent receiver systems required for astronomy [200, 201, 204]. Vacuum tube amplifiers, e.g. extended interaction klystrons, are at present the only technology offering adequate transmitter power for downward looking space radars: these are expensive and electrical power hungry and require replacement with semiconductor equivalents. Similarly, semiconductor device advancements will be required for intersatellite link transmitters to deliver necessary power levels at THz frequencies.