

Frontiers in terahertz sources and plasmonics

Daniel M. Mittleman

The development of innovative tools and techniques is vital for improving research capabilities and opening up new research directions in the terahertz regime. Terahertz sources and plasmonics are just two examples of current exciting advances.

Terahertz (THz) radiation offers researchers many intriguing possibilities, ranging from fundamental science through to applications in communications, non-invasive imaging, and other areas. The THz region (~ 0.1 THz to ~ 10 THz) is often described as the last frontier of the electromagnetic spectrum because of the relatively low maturity level of components and systems that operate in this region. Although the well-known 'THz gap' has begun to be filled through advances in many areas, much of its exciting potential still remains untapped. As a result, the THz community continues to be active and growing, as manifested by the record-setting attendances at relevant conferences¹ and the special issues of journals devoted to the topic (such as this one).

From a spectroscopist's perspective, the excitement surrounding THz science can be easily understood. This spectral region corresponds to photon energies in the range 0.4–40 meV (roughly 3.3 – 330 cm^{-1}), a span that includes the energy scale of many fundamental excitations in condensed matter, including lattice vibrations (phonons), superconducting energy gaps, spin quasiparticles and many other excitations. In disordered materials and conductive media, broadband THz spectroscopy can reveal the microscopic nature of incoherent processes; furthermore, many rotational transitions for molecules in the gas phase fall in the THz range. This spectral range is thus fertile ground for spectroscopy.

THz technologies have the potential to realize new capabilities in a variety of applications. Many of these applications rely on a unique combination of factors: the transparency of many common packaging materials such as paper, cardboard and most plastics; diffraction-limited, submillimetre spatial resolutions

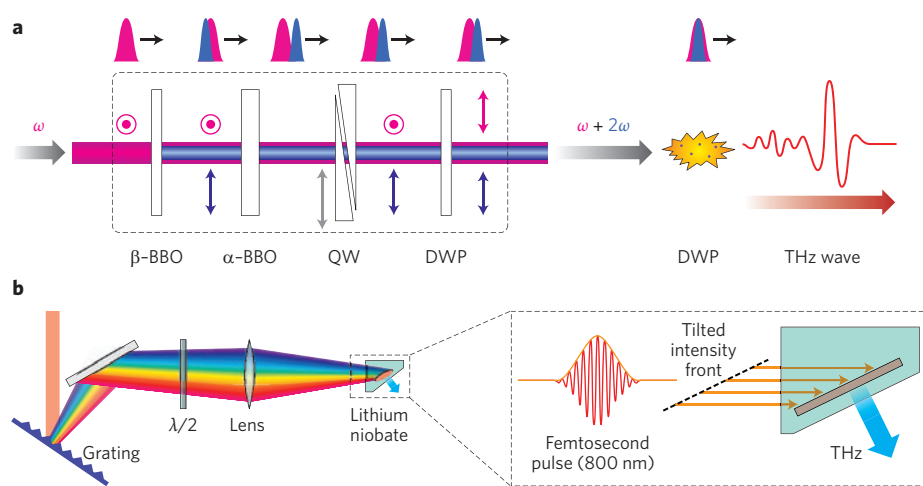


Figure 1 | Two methods for generating high-intensity THz pulses. **a**, Schematic of the experimental set-up for generating THz pulses in air. The dashed rectangle indicates an inline phase compensator. β -BBO, beta-barium borate crystal; α -BBO, alpha-barium borate crystal; QW, quartz wedges; DWP, dual-wavelength waveplate. The red and blue arrows indicate the polarization of the fundamental (ω) and second-harmonic (2ω) beams, respectively. **b**, Schematic of the experimental set-up for the tilted-pulse-front method of high-intensity THz generation in LiNbO_3 (ref. 7). Figure **a** reproduced with permission from ref. 8, © 2011 IEEE.

comparable to that of human vision; and the possibility of identifying unknown materials (especially gases, molecular crystals and polycrystalline powders) based on spectral 'fingerprints'. Researchers have envisioned a wide variety of uses, ranging from security to non-destructive testing. Other ideas exploit the large bandwidth available for short-range wireless communication or the sensitivity to moisture content (as water is a strong absorber in the THz region) for non-contact inspection of food products.

Of course, a great deal of effort is still required to make progress in these areas. The development of innovative tools and techniques continues to be critical for improving research capabilities and opening up new avenues of exploration.

Indeed, two of the most active areas in this field are the development of novel high-intensity THz sources and plasmonics for providing new routes to control and manipulate THz beams. This Commentary focuses on these two forefront research areas.

THz sources

One of the key challenges in the THz field is the low energy or power provided by typical THz sources. THz gas lasers, which have been around since the late 1960s, can provide watts of power in a continuous-wave single-frequency beam, but they are often bulky, expensive and inconvenient for many applications. THz time-domain spectroscopy systems, which are based on nonlinear frequency

conversion of short optical pulses, typically produce average powers of the order of only nanowatts and pulse energies in the femtojoule range, although they offer the possibility of subpicosecond temporal resolution. Other sources have been explored, including vacuum devices such as backward-wave oscillators, travelling-wave tubes, gyrotrons, nonlinear frequency multiplication of powerful microwave sources and nonlinear optical sources based on parametric generation. These various options offer some interesting compromises between power, bandwidth and tunability, as well as cost, complexity and availability.

Clearly, the needs of a given application dictate the best choice of THz source. For example, broadband THz transmission spectroscopy has simplified dramatically with the increasing refinement of THz time-domain spectroscopy. This is because it provides broad spectral coverage, reasonable spectral resolution and a high dynamic range, and because this application does not generally require high power². Sophisticated and flexible spectrometers based on the time-domain approach are now widely available, with several vendors offering a variety of options. Similarly, numerous applications (such as short-range communication) can now rely on sources that provide milliwatt or higher outputs with limited tunability, based on compact vacuum electronics³. Active imaging applications that demand higher spatial resolutions can exploit quantum cascade lasers⁴, which provide milliwatts of power at somewhat higher frequencies than can typically be achieved using vacuum electronics (however, these lasers generally require aggressive cooling).

Given this situation, it is not surprising that research into sources and systems continues to be actively pursued. As time-domain systems have amply demonstrated, power is not the only important consideration. One of the most exciting developments in THz science in the past few years has been the demonstration of new techniques for producing high peak intensities, rather than merely high average powers. The optimization of the efficiency of nonlinear frequency conversion from short optical pulses to short THz pulses, combined with the efficient scaling of the power in the input optical beam, has led to growing excitement regarding THz nonlinear optics.

Previously, driving optical nonlinearity in the THz range required the use of large facilities. Free-electron lasers (for example, those at the University of California in Santa Barbara, USA, and DESY in Hamburg, Germany) have been used to

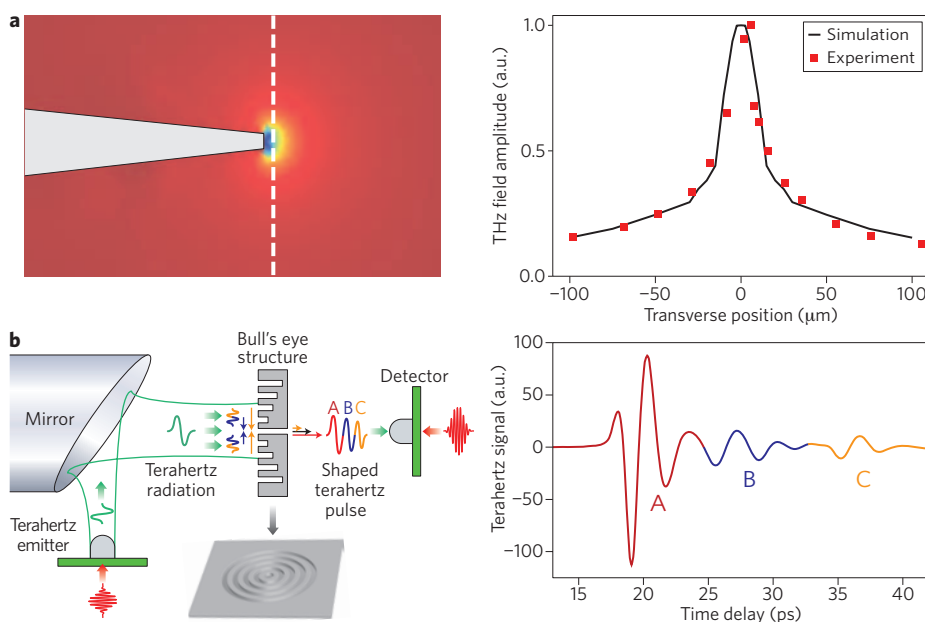


Figure 2 | Illustrations of THz plasmonic effects. **a**, Simulation and measurement of the longitudinal component of the THz electric field at the tip of a smooth tapered wire waveguide along an axis transverse to the waveguide (indicated by the dashed line). The field is confined to a volume of smaller than 1/100th of the wavelength. **b**, Experimental arrangement for studying extraordinary transmission through a subwavelength hole. A series of concentric grooves on the input facet of the opaque mask gives rise to delayed pulses ('B' and 'C') transmitted through the subwavelength aperture, which emerge after the primary transmission ('A'). Figure **a** (left) redrawn with permission from ref. 21, © 2009 AIP; figure **a** (right) reproduced with permission from ref. 21, © 2009 AIP; figure **b** reproduced with permission from ref. 25, © 2005 OSA.

induce and study nonlinear interactions for several years. In addition, intense synchrotron sources may also soon come online⁵. More recently, nonlinearity has become accessible using tabletop sources driven by amplified femtosecond amplifiers. This last development has resulted in a large increase in the number of research groups working in the field of nonlinear THz science.

Two general strategies have been developed for producing subpicosecond THz pulses with energies in the microjoule range (corresponding to peak electric fields in the MV cm^{-1} range or higher). Both strategies employ amplified femtosecond pulses. The first approach relies on efficient nonlinear conversion in a crystalline material with a large second-order nonlinearity. This phase-matched process can be either collinear⁶ (for example, parametric generation in GaSe) or non-collinear⁷ (for example, optical rectification in LiNbO_3 , as shown in Fig. 1b). These methods are conceptually fairly similar to traditional methods for nonlinear $\chi^{(2)}$ frequency conversion, although they have been optimized for the generation of very broadband pulses in the THz range by employing

high-energy (typically millijoule) input pulses. In contrast, the second approach employs non-phase-matched rectification by focusing an intense optical pulse and its second harmonic in a gas (often air). Above the ionization threshold, the laser-generated plasma produces a burst of intense THz radiation with a remarkably broad bandwidth and high power. The second-order nonlinearity, which requires symmetry breaking of the nominally centrosymmetric (gaseous) medium, originates from the superposition of the input fundamental and second-harmonic fields with adjustable relative phase⁸, as shown in Fig. 1a.

These two techniques each have their own advantages and disadvantages. The rectification or parametric generation schemes typically produce greater THz pulse energies, but with more restricted bandwidth as a result of the phase-matching constraints. The THz generation technique in air is somewhat more challenging to align because of the need for careful phase control of the overlap between the fundamental and second-harmonic fields, but it produces very broad (~ 30 THz) smooth and continuous spectra, and moreover may be more readily scalable

to higher energies because there are no damage threshold issues. Both techniques have rapidly gained a foothold in research laboratories around the world, opening up new possibilities for ultrafast nonlinear optics in the THz region. It is now possible to drive condensed matter systems into a non-perturbative regime using intense coherent few-cycle THz pulses^{9,10}, and even to induce irreversible material damage with a THz beam¹¹.

As an aside, an issue that is becoming increasingly relevant with the proliferation of high-power THz sources is that of exposure limits. Currently, no regulatory agency offers specific guidance for the THz region. It is generally thought that the extension of existing exposure limits for the microwave range should be adequate. This assumption has been justified on the basis that THz radiation should have the same biological damage mechanism as that of microwaves, namely heating. As there have been no reliable observations of any well-resolved underdamped absorption resonances in realistic (that is, not dried or frozen) biological samples, it seems reasonable to discuss the absorption of radiation in such systems in the context of a dense and highly overdamped collection of vibrational resonances¹². In this case, it is difficult to imagine any mechanism besides simple heating by which THz radiation could influence biological molecules or systems in aqueous environments. Nevertheless, a few recent experiments have revealed what appear to be non-thermal THz-induced changes, for example in gene expression¹³ and cell membrane potential¹⁴. Because of the lack of an experimentally testable hypothesis regarding the interaction mechanism, the community remains sceptical, but clearly more studies on this issue are required.

THz plasmonics

The second area in which there has been a great deal of interest and activity involves the extension of the ideas of plasmonics to the THz range. Just as in the more familiar near-infrared and visible regions of the spectrum, plasmonics has the potential to enable new capabilities in the THz regime¹⁵.

A surface plasmon polariton (SPP) is an electromagnetic wave that propagates along the interface between a metal and a dielectric; it is coupled to a charge density oscillation in the metal. The imaginary part of the wave's transverse wave vector attenuates the wave amplitude perpendicular to the interface, so that the wave is confined near the interface. The wave also extends a finite distance into the metal, so that it propagates with

both loss (ohmic losses in the metal) and dispersion. Thus, the most strongly bound SPPs are also the most lossy ones. In the visible and near-infrared regions, where many common metals have relatively large resistivities, the interaction between the wave and the electron plasma is very strong, and hence the propagation distance is relatively short. At the very low frequencies of the microwave and radiofrequency regions, where metallic resistivity is typically extremely low, the interaction is very weak. In this case, such waves are often referred to as 'surface waves', essentially plane waves propagating parallel to the interface with very little plasmonic character.

One still frequently encounters the claim that the THz regime is equivalent to the low-frequency regime, so that there is no such thing as a THz surface plasmon at a smooth metal interface. However, this statement ignores many experimental observations of plasmonic behaviour at THz frequencies^{16–19}. In fact, THz surface plasmons operate in an intermediate regime, where the interaction between the electromagnetic mode and the electrons in the metal are stronger than in the microwave range, but weaker than in the infrared region. This gives rise to many unique possibilities for both physics and engineering. For example, if a metal surface curves, a significant fraction of the propagating THz wave will follow the curving metal. This is in contrast to the case in the microwave region where almost none of the electromagnetic energy of a surface wave follows a curved surface. Furthermore, THz SPPs can have propagation distances of hundreds or thousands of wavelengths, whereas SPPs at higher frequencies typically have propagation lengths of only a few tens of wavelengths.

The applications of plasmonic concepts in the THz region are very diverse. As mentioned above, one can take advantage of the relatively low propagation loss and dispersion to build effective THz waveguides²⁰. By tapering waveguide structures, plasmonic effects can be exploited to obtain deep subwavelength field confinement, while maintaining the broadband and nearly dispersionless nature of the propagation^{21,22} (Fig. 2a). Alternatively, one can employ the concept of a spoof surface plasmon, which is a surface wave propagating on a structured metallic surface, giving it an enhanced plasmonic character²³. By engineering the structure, the interaction between the electromagnetic wave and the metal can be enhanced, although generally at the

expense of a higher propagation loss and a lower bandwidth. This can also be valuable for subwavelength field confinement²⁴, as well as controlling the transmission^{25,26} and beam collimation²⁷ at subwavelength apertures (Fig. 2b).

Other plasmonic concepts have also become prominent in the THz community. For example, metamaterials, which are composite metal/dielectric structures with subwavelength elements, have been studied as the basis for new THz devices. In most cases, these are planar metal patterns on a dielectric substrate, but emerging techniques for out-of-plane fabrication continue to open up new possibilities²⁸. Numerous technologies such as electrical modulation²⁹, polarization control³⁰ and cloaking³¹ have been explored through the careful design of THz metamaterial structures. A second example is the use of plasmonics to create large enhancements in the peak electromagnetic field near a metal structure. This idea, which is the basis of sensitive sensing strategies in the visible and near-infrared regions, is now beginning to be explored in the THz region as well. Nanoscale slits in a metal screen can produce giant field enhancements³², which can then improve the ability to detect an analyte of interest³³ through a mechanism analogous to surface-enhanced infrared absorption.

Outlook

For many years, the THz community faced a sort of catch-22 situation — there was no strong incentive to develop new technologies without first identifying a 'killer application', but there was no motivation to explore applications without a viable technology base. Of course, this situation is not unique to THz technologies. It was noted as early as 1970 for the THz field³⁴, and was still true in the early part of this century². In such a situation, one can anticipate slow steady growth until a certain threshold is reached, at which point technological feasibility stimulates rapid progress. Within the last few years, the field appears to have reached that threshold, as we are now witnessing rapid growth in both fundamental and applied research in THz science and technology, as well as in the number of companies manufacturing THz components and systems. THz technologies are anticipated to have an enormous societal impact as proposed applications continue to be commercialized. It is now also becoming clear that the impact of nonlinear THz science on the THz community may be as large as that of nonlinear optics on the optics community.

The recent developments highlighted here represent just a few examples of the exciting advances that are contributing to the vibrancy of THz science and technology. □

Daniel M. Mittleman is at the Department of Electrical and Computer Engineering, Rice University, MS 378, Houston, Texas 77251-1892, USA.
e-mail: daniel@rice.edu

References

1. International Society of Infrared, Millimeter and Terahertz Waves; available at <http://www.irmmw-thz.org/>.
2. Mittleman, D. (ed.) *Sensing with THz Radiation* (Springer Series in Optical Sciences 85, Springer, 2003).
3. Booske, J. H. *et al. IEEE Trans. THz Sci. Tech.* **1**, 54–75 (2011).
4. Williams, B. S., Kumar, S., Hu, Q. & Reno, J. L. *Electron. Lett.* **42**, 89–91 (2006).
5. Carr, G. L. *et al. Nature* **420**, 153–156 (2002).
6. Junginger, F. *et al. Opt. Lett.* **35**, 2645–2647 (2010).
7. Hebling, J., Yeh, K.-L., Hoffmann, M. C., Bartal, B. & Nelson, K. A. *J. Opt. Soc. Am. B* **25**, B6–B19 (2008).
8. Dai, J., Liu, J. & Zhang, X.-C. *IEEE J. Sel. Top. Quant. Electron.* **17**, 183–190 (2011).
9. Hirori, H. *et al. Nat. Commun.* **2**, 594 (2011).
10. Junginger, F. *et al. Phys. Rev. Lett.* **109**, 147403 (2012).
11. Liu, M. *et al. Nature* **487**, 345–348 (2012).
12. Markelz, A. G. *IEEE J. Sel. Top. Quant. Electron.* **14**, 180–190 (2008).
13. Wilmink, G. J. & Grundt, J. E. *J. Infrared Millimeter THz Waves* **32**, 1074–1122 (2011).
14. Siegel, P. H. & Pikov, V. *Electron. Lett.* **46**, S70–S72 (2010).
15. Yu, N. *et al. Electron. Lett.* **46**, S52–S57 (2010).
16. O'Hara, J. F., Averitt, R. D. & Taylor, A. J. *Opt. Express* **13**, 6117–6126 (2005).
17. Jeon, T.-I. & Grischkowsky, D. *Appl. Phys. Lett.* **88**, 061113 (2006).
18. Astley, V., Scheiman, J., Mendis, R. & Mittleman, D. M. *Opt. Lett.* **35**, 553–555 (2010).
19. Liu, J., Mendis, R. & Mittleman, D. M. *Appl. Phys. Lett.* **98**, 231113 (2011).
20. Wang, K. & Mittleman, D. M. *Nature* **432**, 376–379 (2004).
21. Astley, V., Mendis, R. & Mittleman, D. M. *Appl. Phys. Lett.* **95**, 031104 (2009).
22. Zhan, H., Mendis, R. & Mittleman, D. M. *Opt. Express* **18**, 9643–9650 (2010).
23. Pendry, J. B., Martín-Moreno, L. & García-Vidal, F. J. *Science* **305**, 847–848 (2004).
24. Maier, S. A., Andrews, S. R., Martín-Moreno, L. & García-Vidal, F. J. *Phys. Rev. Lett.* **97**, 176805 (2006).
25. Agrawal, A., Cao, H. & Nahata, A. *Opt. Express* **13**, 3535–3542 (2005).
26. Liu, J., Mendis, R. & Mittleman, D. M. *Phys. Rev. B* **86**, 241405(R) (2012).
27. Yu, N. *et al. Nature Mater.* **9**, 730–735 (2010).
28. Tao, H. *et al. Phys. Rev. Lett.* **103**, 147401 (2009).
29. Chen, H.-T. *et al. Nature* **444**, 597–600 (2006).
30. Zhang, S. *et al. Nat. Commun.* **3**, 942 (2012).
31. Iwaszczuk, K. *et al. Opt. Express* **20**, 635–643 (2012).
32. Seo, M. A. *et al. Nature Photon.* **3**, 152–156 (2009).
33. Park, H.-R. *et al. Nano Lett.* **13**, 1782–1786 (2013).
34. Senitzky, B. & Oliner, A. A. in *Proc. Sym. Submillimeter Waves* (ed. Fox, J.) (Polytechnic Inst. of Brooklyn, 1970).