A review of terahertz sources

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Abstract
Bibliometric data set the scene by illustrating the growth of terahertz work and the present interest in terahertz science and technology. After locating terahertz sources within the broader context of terahertz systems, an overview is given of the range of available sources, emphasizing recent developments. The focus then narrows to terahertz sources that rely on surface phenomena. Three are highlighted. Optical rectification, usually thought of as a bulk process, may in addition exhibit a surface contribution, which, in some cases, predominates. Transient surface currents, for convenience often separated into drift and diffusion currents, are well understood according to Monte Carlo modelling. Finally, terahertz surface emission by mechanical means—in the absence of photoexcitation—is described.

Keywords: terahertz, from, surfaces

1. Introduction

1.1. Terahertz bibliometrics
Before turning to the topic of terahertz sources, a brief survey of the growth of the terahertz field as a whole will be made from a bibliometric perspective.

1.1.1. Terahertz outputs have grown exponentially. For decades the field of terahertz science and technology has grown exponentially. Literally. Figure 1 displays the number of documents published in the years 1975–2013 that contain ‘terahertz’ in the abstract, title or keyword field [1]. Figure 1(a) is a linear-linear display and figure 1(b) is a log-linear display of the same data. The data from 1975 to 2010 (inclusive) have been fitted with an equation for exponential growth,

\[ D = D_0 + \alpha \exp \left( \frac{Y - Y_0}{\tau} \right) \]  

(1)

Here \( D \) is the number of documents, \( D_0 = 0 \) is the base number of documents, \( Y \) is the year, \( Y_0 = 1975 \) is the base year, \( \alpha = 1.55 \pm 0.42 \) is the initial rate—meaning approximately one or two documents per year to begin with—and \( \tau = 4.66 \pm 0.18 \) is the time constant—meaning an increase by a factor of e approximately every five years. This time constant implies that the annual output of documents doubles approximately every three years (precisely, every 3.23 years). A simple and memorable model of one document in 1975 and the annual production doubling every three years,

\[ D = 2^{(Y-1975)/3} \]  

(2)

furnishes a reasonable approximation to the output of terahertz literature between 1975 and 2013, as indicated by the crosses in figure 1(b).

1.1.2. The most-cited articles concern technology. In each of the last three full years (2011, 2012 and 2013), more than 3000 documents have appeared with ‘terahertz’ in either their title or abstract or as a keyword. The most-cited documents involve terahertz technology. For example, the articles ‘Terahertz semiconductor-heterostructure laser’ [2], ‘Cutting-edge terahertz technology’ [3], ‘Terahertz technology’ [4] and ‘Materials for terahertz science and technology’ [5] have each been cited more than 1000 times.

1.1.3. Monographs are now emerging. The field has sufficiently matured such that monographs are now appearing. At first, these were edited works, with contributions from numerous authors, on topics such as terahertz sensing [6], terahertz optoelectronics [7] and terahertz spectroscopy [8].
Figure 1. Number of documents with ‘terahertz’ appearing in the abstract, title or keyword field as a function of time. (a) Linear-linear scale. The full line is given by $y = 1.55 \exp((x - 1975)/4.66)$. (b) Log-linear scale. The crosses correspond to a starting value of 1 in 1975, doubling every third year.

As the field matures further, more cohesive books are appearing, with only two or three authors [9–11] and, at last, sole authors [12, 13], on topics such as terahertz excitations [9], terahertz photonics [10], terahertz techniques [11], terahertz principles [12] and terahertz physics [13].

1.2. Terahertz apparatus

A typical terahertz experimental station, such as a developed for imaging [14], tomography [15] or spectroscopy [16], comprises the three principal parts of the source, which produces the terahertz radiation, the components, which manipulate the radiation, and the detector, which senses the radiation. Practical arrangements may contain multiple sources, components and detectors (figure 2).

1.2.1. Terahertz sources. The focus of this article is on terahertz sources, which will be discussed in greater detail shortly (section 2). To give some context for the discussion of sources, a few brief remarks will be made first about terahertz components and detectors.

1.2.2. Terahertz components. Optical components are such things as mirrors, lenses and polarizers.

In contrast to visible optical systems, where lenses and similar transmitting elements predominate, terahertz systems tend to employ reflecting elements, which have minimal loss and no dispersion. Terahertz mirrors have conventionally been made of metal. Other materials have been recently trialled, for example, doped and undoped GaAs [17] and a hybrid of polypropylene and high-resistivity silicon [18]. Tunable mirrors, based on one-dimensional photonic crystals, have also been developed [19–22].

Lenses are typically made of plastics. It is advantageous if the lens transmits visible radiation, as this facilitates optical alignment. Traditionally, plastic terahertz lenses were made by machining on a lathe. Recently, lenses have been manufactured by compressing various micropowders in metal moulds using a tabletop hydraulic press [23]. Lenses with an adjustable focal length have been realized by introducing medical white oil into the lens cavity [24]. Less conventional lens designs include grooved-dielectric Fresnel zone plates, thin and lightweight [25]; plasmonic-resonance lenses, fabricated from 100 nm gold foil on a 500 µm silicon wafer [26]; and even lenses made from paper [27].

A precise method of fabricating terahertz optical components is femtosecond laser machining of LiNbO3 single crystals [28]. Diffractive elements, cavities and waveguides have been so constructed. On the other hand, temporary, or reconfigurable, components may be formed by optical modulation, using visible light projected onto a silicon chip; aperture arrays and polarizers have been made in this manner [29].

1.2.3. Terahertz detectors. The earliest terahertz detectors were based on a thermometric property of a material, such as a change in electrical resistance with temperature (the bolometer) or a change in size with temperature (the Golay cell). More sophisticated examples of thermal detectors are under development, such as microelectromechanical systems (MEMS) incorporating a tuned metamaterial absorber and a bi-material (differential thermal expansion) [30–32]. In principle, arrays of such sensors will facilitate imaging, but they are presently optimized only for specific frequencies. Moreover, thermo-mechanical systems are inherently slow. Electronic transitions are faster. Electronic sensors were originally based on bulk semiconductor materials, but more sophisticated designs have been demonstrated, for example, impurity bound-to-unbound transitions in beryllium and silicon delta-doped GaAs/AIAs multiple quantum wells [33]. On a finer scale still, quantum dots have been employed as terahertz sensors [34]. Single electron transistor read out is employed. A limitation is that the operating temperature is close to zero, 0.3–1.5 K [35].
With the development of time-domain spectroscopy (TDS), the distinct identity of the sensor has become blurred. The sensor functions only within the system as a whole, requiring for its operation the ultrashort laser pulse that also is essential to the operation of the source. The two broad TDS detector types are electro-optical [36] and photo-conductive [37]. The use of asynchronous optical sampling [38] obviates the need for a mechanical delay stage in TDS and has been used to characterize surface sensors based on split-ring resonators [39].

### 2. Terahertz sources

There are many sources of terahertz radiation. A small sampling is given in table 1.

**Extraterrestrial thermal sources** include the sun and the cosmic background radiation [40]. Common laboratory thermal source are the mercury lamp [41] and the globar, an electrically heated rod of carborundum (SiC). Various schemes are available to mount the globar [42, 43]. It typically operates at 1650 K with emissivity in the range 0.5–0.8 [44], so may be thought of as a ‘grey-body’. It provides more power than the Nernst glower in the terahertz region [45].

**Vacuum electronic sources** include the backward-wave oscillator [46–50], extended-interaction klystrons [51–55], travelling-wave tubes [56], gyrotrons [57–62], free-electron lasers [63–66] and synchrotrons [67–76]. These sources are typically of high power [77]. **Solid-state electronic sources** include the Gunn diode [78–81] and high-frequency transistors [82–86]. Frequency multipliers are used to shift fundamentally sub-terahertz electronic oscillations into the terahertz range [87–89]. Josephson junctions serve as sources of terahertz and sub-terahertz radiation in superconductors [90, 91].

**Terahertz lasers** have been built from the archetypical elemental semiconductors, germanium and silicon [92, 93]. Semiconductor lasers include electrically pumped photonic-crystal lasers of low angular divergence [94]. Much interest is currently in the quantum cascade laser, a challenge being to raise the temperature of operation [95–97]. Gas lasers preceded the solid state lasers [98].

Visible or near-infrared lasers, either operating continuously or in pulsed mode, are widely-employed in various schemes to generate terahertz radiation by **optical pumping**. Two **continuous** laser sources may be mixed and the difference frequency lie in the terahertz range [99]. The laser sources exploited include diode lasers [100], dual-mode lasers [101], multi-mode lasers [101] and a dual-colour-VECSEL [102]. Photomixers are typically based on low-temperature grown GaAs. Difference frequency mixing occurs in a variety of materials, notably DAST [103]. Periodically inverted electro-optic crystals [104] and tilted fields increase efficiency [105]. Much work is at 780 nm, but there is also great interest at communications wavelengths of about 1.5 µm [106, 107]. These in practice use pulsed sources, but do not depend on them. Continuous stimulation of mesocrystal microspheres by a single laser produces mechanical resonance accompanied by terahertz emission [108, 109]. **Pulsed** laser sources, the basis of time-domain spectroscopy, have been reviewed by Davies et al [110] and Kitaeva [111]. Pulsed lasers are used to excite photoconductive switches [112] or antennas [113]. Terahertz radiation also arises when a laser pulse pumps nothing more than air—the photoionization of the gas producing a plasma [114]. Other gases, including noble gases, may be used [115]. The radiation is very broad in its frequency range [116, 117]. As a diagnostic method, gas photoionization allows the measurement of the carrier-envelope phase of short laser pulses [118]. Solid targets may be used [119]. The polarization may be coherently controlled [120], and is enhanced by electric fields [121, 122]. In ferromagnetic films, magnetic, rather than electric, dipoles are employed [123]. Terahertz parametric oscillation in crystals such as LiNbO₃ pumped by ns-Nd : YAG lasers produces coherent, tunable and unidirectional radiation; efficient output coupling is critical in realizing high power [124–126].

### 3. Terahertz surface emission—optical rectification

Experimentally, the principal signature of optical rectification is the strong geometrical dependence. As the emitting crystal is rotated around its surface normal perpendicular to the propagation direction of the pump beam an increase and decrease in the emitted terahertz radiation is observed. This phenomenon is usually referred to as ‘azimuthal angle dependence’. Nanoporous InP (1 1 1) membranes, for example, show a marked azimuthal angle dependence, indicating optical rectification plays a major role [127]. Secondly, the terahertz emission remains directly proportional to the pump power, without evidencing saturation [127]. Thirdly, subjecting the crystal to a magnetic field has little or no effect.

#### 3.1. Bulk effects

Optical rectification can occur in the bulk of a material. The bulk effect has been studied in detail in various crystals. Complete expressions have been given for zinc-blende (43m) crystal faces of arbitrary orientation [128]. The terahertz generation in uniaxial birefringent crystals is similar in principle, but differs in the important respect that the polarization of the pump beam rotates as it traverses the crystal, as is illustrated in the case of ZnGeP₂ (chalcopyrite) [129]. For ZnGeP₂, it is found that {1 1 4} planes are more efficient than {0 1 2} and {1 1 0} planes for terahertz generation [129].
3.2. Surface effects

Not only the bulk of a crystal contributes to optical rectification. A surface contribution, induced by the electric field, may be important, and even dominate the bulk effect, at least at high excitation fluences. The surface electric-field-induced effect has been studied in detail in InAs, for (1 0 0), (1 1 0) and (1 1 1) faces, with second-harmonic (sum frequency) measurements made to supplement the terahertz (difference frequency) data, and the polarization of both measured [130]. It was found that bulk optical rectification was inadequate to explain the experimental results; an additional surface electric-field-induced contribution was also present. In fact, the surface field contribution was greater than the bulk contribution. Rotating the sample relative to the pump beam (azimuthal angle dependence of the terahertz emission) clarified this.

A surface electric-field-induced optical rectification has since been found in other materials. For example, the azimuthal angle dependence of terahertz emission from (1 0 0), (1 1 0) and (1 1 1) faces of Ge is consistent with a surface field effect and leads to an estimation of the third-order nonlinear optical susceptibility of Ge [131]. In the case of (1 1 2) planes of InSb, both bulk and surface field-induced optical rectification are observed, with the latter approximately twice the strength of the former [132].

General expressions for surface optical rectification for arbitrary planes have been calculated [128]. In the accompanying experimental study of high-index planes of GaAs it was found that, while the bulk expressions gave a good agreement with the data, including the surface expressions gave an excellent agreement. Only a single parameter was needed to fit the data for the (1 1 2)A, (1 1 3)A, (1 1 4)A, (1 1 5)A, (1 1 2)B, (1 1 3)B, (1 1 4)B and (1 1 5)B faces of GaAs; moreover, the surface fields on the faces were estimated [128]. Those results applied to transmission geometry. The work has been extended to the more complicated situation of quasi-reflection geometry. Here transient currents, bulk optical rectification, and surface optical rectification all play a role, but the various contributions can be untangled [133].

The contribution of surface field-induced optical rectification is essential in explaining the terahertz emission from GaAsBi (3 1 1)B faces [134]. To explicate the mechanism, the effect of increasing optical fluence on the
terahertz emission was investigated. The effect was linear, with no saturation (such as would indicate a transient current effect) being observed. Moreover, rotation of an in-plane magnetic field also had no effect on the produced terahertz radiation, again suggesting no role of transient currents. The azimuthal angle dependence of the substrate, with three peaks per rotation, was quite different to that of the GaBi0.035As0.965 epilayer, which exhibited only one peak per rotation. While bulk rectification could not account for the epilayer result, inclusion of the surface optical rectification term could [134]. More recently, a similar account has been given of terahertz emission from nanostructured (3 1 1) GaAs [135].

4. Terahertz surface emission—transient currents

Terahertz emission can result from a changing dipole. The terms current surge, surge current or transient current are alternative ways of describing this. The transient current itself may arise because of carrier drift, due to the surface electric field, or due to diffusion; these currents are respectively referred to as drift currents (section 4.2) and diffusion currents (section 4.1).

Experimentally, there are several signatures of a transient-current emitter [136]. Rotating a magnetic field in the plane of the sample surface will change the direction of the Lorenz force on the charge carriers. This will either add to or subtract from the motion leading to terahertz emission and consequently an increase and decrease in the terahertz emission is observed. So, for example, bulk InP (1 1 1) crystals exhibit a transient current effect, whereas nanoporous InP (1 1 1) crystals do not [127]. Likewise, the dependence of the terahertz generated on the excitation fluence can give insight into the emission mechanism. Transient current emitters tend to saturate, as a result of charge screening, as the optical fluence increases. Charge-carrier screening, as well as terahertz absorption in the material, imply heavily doped semiconductors in general are not good emitters. For example, in measurements of InP doped in the range n-type \( 7 \times 10^{18} \text{cm}^{-3} \) to \( p \)-type \( (8-10) \times 10^{19} \text{cm}^{-3} \), the highest signal was from nominally undoped \( (\leq 1 \times 10^{16} \text{cm}^{-3}) \) InP [136].

4.1. Diffusion currents

Diffusion currents arise when charge carriers of opposite sign diffuse at different rates. Typically, diffusion currents dominate for high carrier energies [137]. A dipole is formed if the positive and negative charge carriers diffuse at different rates. This is known as the Dember effect (or the photo-Dember effect) [138–140]. It is usual that electrons are more mobile than holes. It is often a good approximation to think of the holes as stationary. Usually, diffusion is away from (perpendicular to) the surface. The case of diffusion along (parallel to) the surface is termed the lateral effect. This direction of the current is optimal for coupling out of the surface. The lateral photo-Dember effect was first reported by pumping (1 0 0) GaAs and In0.53Ga0.47 on InP at 825 nm [141]. Subsequently the effect was observed under pumping with an Er: fibre (1.55 \( \mu \text{m} \)) laser [142]. The effect has also been observed in semi-insulating and low-temperature grown GaAs [143]. A cylindrical micro-lens array was found to increase the output power five-fold [144]. The effect of the spot position and size and an external bias have been investigated [145], as have the dependence on fluence and polarization [143].

4.2. Drift currents

Typically, drift currents dominate for high electric fields [137]. Drift currents are influenced strongly by the local surface field, as illustrated by experiments involving passivated surfaces [146]. For this reason, emission due to drift currents is sometimes termed surface-field emission.

4.3. Modelling

Monte Carlo modelling has been used to simulate the motion of photoexcited charge carriers and so clarify the mechanisms of terahertz emission. The landmark work is that of Johnston et al [147]. This demonstrated that InAs is predominantly a photo-Dember, or diffusion, emitter and GaAs is predominantly a surface-field, or drift, emitter. Moreover, the improvement of emission with a magnetic field is largely due to geometric reorientation of the radiating dipole (rather than a change in the dipole strength). Magnetic-field enhancement of terahertz emission had been reported for InSb, InAs, InP GaSb and GaAs in modest magnetic fields (up to 1.2 T) [148] and attributed to increased radiation from transient currents in the surface plane [149]. Other Monte Carlo modelling work has identified the roles of hot and cold charge carriers [150] and explored the roles of the photon energy of the pump beam [151, 152], large electric fields [153], and the cross-over from drift to diffusion emission [154]. InSb [155] and InAs [155, 156], GaAs \( p-i-n \) structures [157], and \( \delta \)-doped and other heterostructures [158–162] have been investigated. Similar modelling has also been carried out on terahertz photoconductive emitters [163] and detectors [164].

Modelling allows parameters that are difficult or perhaps impossible to vary in practice to be varied in the simulation, with a view to understanding how terahertz emission might be improved. Consider the case of InAs [165]. Scattering is seen to play a minor role. The terahertz emission with all scattering mechanisms included in the simulation (polar optical phonon, carrier–carrier, intervalley, and ionized impurity) differs little from the emission with no scattering at all. The transport, on the sub-picosecond timescale, is almost collisionless. Likewise, varying the dielectric constant or the surface field has little effect. Varying the band gap or the effective mass have a noticeable effect, which can be understood in mechanical terms—if the electrons or holes are given greater kinetic energy, the dipole resulting from the motion of the charge they carrier will change more rapidly. It is also clear from the simulation that, during typical pump pulse of 100 fs, there is sufficient time for the initially-created electrons to move away from the surface and so create an opposing potential even before the peak of the pump pulse arrives. The existence of such a vanguard counterpotential implies that shorter (<100 fs) excitation pulses not
only improve the bandwidth but also improve the strength of the terahertz emission [156, 165].

The case of InAs may be contrasted with that of GaAs [166]. There are significant differences. Polar optical phonon scattering plays a large role in reducing the output of terahertz radiation from GaAs surfaces. The effect of the surface potential is great; the terahertz field not only reduces as the surface potential does, but reverses in direction as the sign of the surface potential changes. There is a vanguard counter-potential in GaAs, but it is weaker than in InAs. Overall, InAs is the stronger emitter [166].

Finally, by way of synthesis, it might be remarked that both optical rectification (section 3) and transient currents (section 4) may simultaneously produce terahertz emission. For example, in both InSb and InAs it is observed that the azimuthal-angle varying difference frequency mixing is superimposed on a considerable surge current; moreover, as the temperature is varied, so is the terahertz emission, very markedly for InSb, indicating the presence of the photo-Dember effect [167]. On the other hand, in InP bulk difference frequency mixing is observed to be comparable to the effects of transient currents [168].

5. Terahertz emission from mechanical excitation

Is it possible to generate terahertz radiation from surfaces without any photoexcitation? Recent experimental [169] and theoretical [170] work would suggest so.

5.1. Peeling tape

The process of peeling (sometimes referred to as unpeeling) adhesive tape from the roll it is packaged in, or from another surface, has been long known to produce visible radiation [171]. Recently, peeling tape has been found to generate, in addition to visible light, radio-frequency radiation, as well as x-rays [172]. Low pressure ($< 10^{-2}$ mbar) is required for the x-ray emission to be observed; by examining the pressure dependence of the x-ray fluence the mechanism of emission may be determined [173]—the acceleration of free electrons in the surrounding gas by the potential built up on the tape. Refinements to the x-ray production have been described recently [174–177] and the phenomenon used to study charge localization [178]. Given that peeling tape has been shown to produce visible light and radio-frequency radiation, it is of interest to ask if unwinding tape also produces radiation in the space in the electromagnetic spectrum between these two—in the ‘terahertz gap’.

Peeling adhesive tape has indeed been shown to emit electromagnetic radiation at terahertz frequencies [169]. Over the measured range of 2–32 cm s$^{-1}$, the radiation emitted is independent of the speed at which the tape is unwound. The radiation extends across a broad range of terahertz frequencies, from 1–20 THz. The intensity is rather small, amounting to only a few per cent increase over the blackbody radiation from the tape at room temperature before it is unwound. The radiation was therefore investigated with a high-sensitivity sensor, a helium-cooled bolometer. In contrast to TDS, the emission is incoherent. A comparison of emission from double-sided tape (which will show a heating effect, but no charge separation) and the more usual single-sided tape (in which charge separation occurs on unwinding) establishes that the effect has its origin in charge separation. The radiation shows peaks in intensity at around 2 THz and around 18 THz, as measured by a Fourier-transform spectrometer. The origin of these peaks is unknown. These peaks sit on a background of emission that increases in intensity with frequency. This dependence is the opposite to the usual bremsstrahlung from a plasma, which decreases in intensity with frequency, but is consistent with bremsstrahlung from a plasma with absorption. Also consistent with this mechanism is the observation that the radiation is unpolarized.

5.2. Surface formation

Just as peeling tape emits visible radiation, so does fracturing sugar [179, 180]. This sort of mechanoluminescence generally is understood to involve charge transfer between two newly created surfaces.

There may also be motion of charges within newly formed surfaces. The transient dipoles so formed will then emit radiation at terahertz frequencies (figure 3). The intensity of the radiation will be increased as the speed of surface formation is increased, as the surface potential is increased and as the charge-carrier concentration is increased. This proposed mechanism of radiation has been dubbed ‘terahertz surfoluminescence’ [170].

The intensity of the terahertz field produced by terahertz surfoluminescence may be estimated using the same Monte Carlo calculations as when a charge distribution is produced by photoexcitation. It is found that the terahertz electric field due to surface creation is of the same order as the terahertz electric field produced by a 1 nJ laser strike. This holds true both near room temperature (300 K) and at cryogenic temperature (70 K) [170].
There has been no unambiguous experimental demonstration of terahertz surfoluminescence, although the mechanism may provide a partial explanation for the observation of terahertz radiation from peeling adhesive tape (section 5.1). Previously, cleavage luminescence has been observed in silicon and other semiconductors, including GaAs, InP, Ge and Ge-Si$_{1-x}$-As$_x$. However, that emission was on a far different timescale ($\mu$s rather than ps) and energy scale (eV rather than meV) than terahertz surfoluminescence.

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