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# Potential for detection of explosive and biological hazards with electronic terahertz systems

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The terahertz (THz) regime (0.1–10 THz) is rich with emerging possibilities in sensing, imaging and communications, with unique applications to screening for weapons, explosives and biohazards, imaging of concealed objects, water content and skin. Here we present initial surveys to evaluate the possibility of sensing plastic explosives and bacterial spores using field-deployable electronic THz techniques based on short-pulse generation and coherent detection using nonlinear transmission lines and diode sampling bridges. We also review the barriers and approaches to achieving greater sensing-at-a-distance (stand-off) capabilities for THz sensing systems. We have made several reflection measurements of metallic and non-metallic targets in our laboratory, and have observed high contrast relative to reflection from skin. In particular, we have taken small quantities of energetic materials such as plastic explosives and a variety of *Bacillus* spores, and measured them in transmission and in reflection using a broadband pulsed electronic THz reflectometer. The pattern of reflection versus frequency gives rise to signatures that are remarkably specific to the composition of the target, even though the target's morphology and position is varied. Although more work needs to be done to reduce the effects of standing waves through time-gating or attenuators, the possibility of mapping out this contrast for imaging and detection is very attractive.

**Keywords: electronic terahertz systems; bacterial spores; explosives**

## 1. Introduction

The heightened sense of vulnerability to concealed threats—whether non-metallic weapons, explosives or even biological agents such as anthrax—has motivated a wide-ranging response from the scientific community. Terahertz (THz) technology, with its ability to 'see through' many types of non-metallic enclosures (and to distinguish metal from plastics), is a promising candidate to address the problem of detecting such concealed threats.

One contribution of 16 to a Discussion Meeting 'The terahertz gap: the generation of far-infrared radiation and its applications'.

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Many researchers with backgrounds in optics explore the regime by generating THz power with mode-locked femtosecond lasers driving photoconductive switches or nonlinear crystals, taking the 'top-down' photon-energy approach to this regime. Here we take the energetic 'bottom-up' angle by multiplying the frequency of microwave or millimetre-wave oscillators using nonlinear transmission lines and integrated antennas. Fundamental sources of THz power, such as backward-wave oscillators, free-electron and quantum cascade lasers, novel heterojunction diode and transistor circuits, and, most recently, micromachined vacuum electronic devices, are now beginning to round out the THz toolset.

Nonlinear transmission lines (NLTLs) are integrated circuits on GaAs that consist of high-impedance waveguides periodically loaded with Schottky diodes in reverse bias (Rodwell *et al.* 1991). These act like voltage-variable capacitors that modulate the wave propagation velocity according to the instantaneous voltage on the line. Voltages at one extreme (e.g. 0 V) travel slowly due to the shallow depletion region (hence large capacitance) beneath the Schottky contact, while voltages at the other extreme (strongly negative biased) cause deep depletion and thus low capacitance, speeding up that portion of the wave. What results is a sawtooth waveform evolving from a sinusoidal input whose frequency ranges from 1 to 10 GHz, depending on the design of the NLTL. These circuits have exhibited sub-picosecond transition times, making them some of the fastest circuits to date (van der Weide 1994).

This set of sources is complemented by a growing set of detectors, coherent samplers and electro-optic sensors that measure the amplitude of the THz field, and hence the real and imaginary components of the dielectric response. Our detectors consist of diode sampling bridges strobed by NLTLs identical to those used for the emitters. If the source of THz is incoherent, thermal (bolometric) or Schottky mixer diodes are used. For example, atmospheric and space-based THz spectroscopy seeks out natural sources of radiation such as the cosmic background or emission from molecular species, and takes advantage of cryogenic detector environments to sense weak photon energies.

For terrestrial applications, thermal detection of THz becomes much more difficult than in space, so time-domain THz systems that can perform coherent generation and detection dominate the scene when it is inconvenient to use cooled bolometers as detectors. These systems synchronize the detector with the emitter so that thermal noise power not related to the emitter's frequencies is largely ignored. The energy at a cryogenic temperature of 5 K corresponds to a 0.1 THz photon, while room temperature (300 K) corresponds to 6.25 THz. Between these two frequencies, the total thermal radiation from a 10 mm square blackbody (an ideal radiator) whose temperature is 300 K is *ca.* 46 mW—overwhelming the sub-milliwatt powers of most THz sources—while at 5 K it is only 350 μW. Thus thermal background strongly delineates space-based and cryogenic THz technology from its room-temperature counterparts. In order to detect THz with room-temperature antennas, we require a technique to limit the input noise power, such as coherent sampling of the THz wave, the technique employed here.

Whether Earth- or space-based, the frequency range bounded by these 5 and 300 K photon energies brackets the majority of THz systems, and defines one of the least explored and most promising regions of the spectrum for a growing range of applications, primarily for sensing and spectroscopy, but perhaps ultimately for communications. Such opportunities in sensing, like detecting explosives or bioweapons, spectroscopic imaging for medical applications, and communications, like exploiting the unlicensed spectrum above 0.3 THz, could become significant and growing military and commercial markets for THz technology.

## 2. THz free-space propagation

The boundaries of the THz frequency range and the technologies associated with it have almost as many definitions as the number of laboratories pursuing THz studies. What unifies most researchers in THz technology is their work with quasi-optical propagation. Although important work has been done to probe and map THz frequencies on planar waveguides and circuits, the overwhelming majority of publications in this field discuss free-space propagation. This naturally involves both antennas borrowed from scaled-down microwave concepts and optics adapted from infrared and visible light. Propagation over distances greater than *ca.* 1 m is envisioned to be necessary to achieve stand-off detection of concealed threats.

Scientific curiosity has motivated a substantial amount of narrowband or continuous-wave work in the THz regime for space research and cosmology, where atmospheric absorption cannot interfere with observations. Back on Earth, however, pressure-broadened absorption lines of water vapour place severe limitations on the distance many THz waves can propagate.

Although sending THz frequencies through gases and measuring selective absorption has been useful for spectroscopy of narrow absorption lines (van der Weide *et al.* 2000), atmospheric absorption due to water vapour is largely a parasitic effect in THz sensing, imaging and communications unless it is used for calibration. Water vapour absorption at 50% relative humidity at frequencies above 0.5 THz rises to between 0.1 and 10 dB $\mathrm{m}^{-1}$ in broad peaks that severely limit the distance of detectable propagation, effectively rendering the atmosphere black. This, in turn, limits stand-off distances when using greater than 0.5 THz frequencies for sensing and imaging the dielectric contrast of remote targets. It also challenges communications systems attempting to use these frequencies for long distances. Conversely, THz communications within rooms or buildings will naturally be quite secure, and THz sensors will exhibit little interference with other systems or with each other. Liquid water in living tissue limits depths of THz penetration to millimetres, limiting THz medical imaging to applications involving skin conditions and teeth.

One approach to gaining greater stand-off detection ability is to develop higher-power sources. Both solid-state two-terminal sources like Gunn or IMPATT diodes and vacuum electronic devices like klystrons, magnetrons and backward-wave oscillators have continued their steady march toward higher power and higher frequencies within the microwave regime over the past several decades. One of the most prominent gaps in the THz toolkit, however, is the lack of suitable broadband and high-power amplifiers (to say nothing of low-noise amplifiers). While numerous source concepts, both broadband and narrowband tunable, are under current investigation, the quest to find the analogy of a broadband laser's gain medium in the THz regime has been challenging. The problem is one of fundamental physics: it gets increasingly difficult with high frequency to move carriers in semiconductor crystals, while, from the visible working towards the THz, inverting carrier populations becomes harder as level separations approach a few times $kT$. One promising approach warranting research is the use of powerful THz vacuum electronic amplifiers, such as travelling-wave tubes, both for narrowband and wideband signals. Large stand-off radar applications would benefit from the high power available from THz-regime gyrotrons, but important research challenges would include finding ways to significantly reduce their size and weight.

Thus atmospheric absorption can make it largely impractical to do spectroscopy or imaging above 0.5 THz (what we will call the 'upper THz' regime) beyond a metre or so of distance between emitter and detector. While this limits the range of applications to mostly laboratory studies and close-in spectroscopic imaging for industrial purposes, the majority of THz systems and results to date have been achieved in this upper THz regime.

## 3. Application of pulsed electronic spectrometer

Even with atmospheric absorption, there are windows of transmission that potentially enable large stand-off detection of concealed threats. By illuminating a sample with a pico- or sub-picosecond electrical stimulus and coherently detecting its response, we can obtain reflection and transmission spectra of several substances and compare them as a starting point for distinguishing biological, chemical and energetic threats. We use an all-electronic THz spectrometer with phase-locked microwave sources to drive GaAs nonlinear transmission lines, enabling measurement of both broadband spectra and single lines with high precision. Our use of stable and frequency-offset synthesizers gives a user-selectable beat frequency that is low enough for processing by conventional 100 kHz instrumentation. We have also implemented a frequency offset generator (single-sideband modulator) that can enable the system to be produced using integrated circuits (Akkaraekthalin *et al.* 1998).

Increasingly sophisticated bioweapons and explosives require increasingly sophisticated detection technologies. Non-metallic threats (e.g. explosives) have motivated a multi-pronged approach to detection, including residue sniffing and computerized tomography. These techniques, however, suffer from invasiveness, slowness, unfamiliarity to the public and significant potential for false negatives and positives. Broadband radiation of this range is normally difficult to achieve, but has great potential for screening; many compounds show specific absorption and dispersion in the 1–500 GHz range. These absorptions can arise both from intrinsic chemical responses (such as long-chain polymer vibrational or librational modes) and from the aperiodic crystalline-air matrix that could 'trap' short wavelength radiation. Because of the difficulty in deconvolving these effects from those of standing waves, it is still challenging to implement this technology in the field. Ways around these issues include variable rate pulsing to time-gate the received pulse and remove the effects of multiple reflections.

Ultrawideband (UWB), carrier-free, impulse or baseband radar has been rapidly gaining popularity in applications where complex and elusive targets are the norm. While UWB radar works up to *ca.* 10 GHz, our circuits generate extremely short electrical impulses (*ca.* 1 ps) with correspondingly millimetre-wave spectra. Because we employ coherent detection, we reject noise outside the frequencies of interest. These systems can be deployed for spectroscopic imaging, as shown in figure 1. With the appropriate optics, output power and receiver sensitivity, the potential exists for a new type of security screening tool.

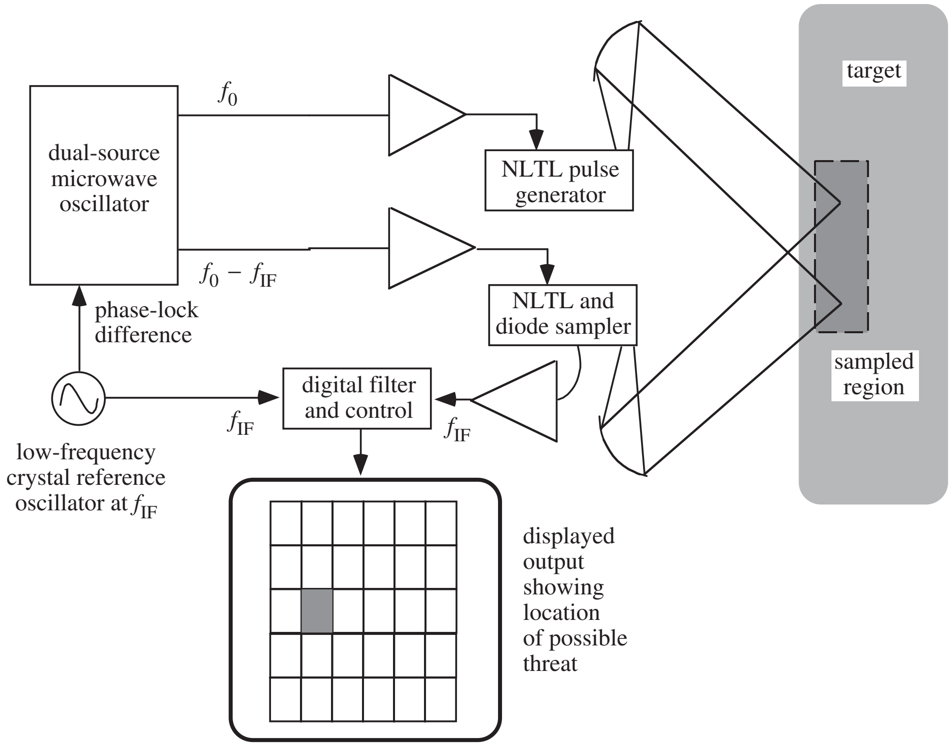

Figure 1. Electronic THz spectroscopic imaging system showing optical arrangement using off-axis paraboloidal mirrors for focusing and collimating the THz beams.

Using this spectrometer in our laboratory, we have measured broadband reflections from non-metallic targets that indicate specific spectroscopic signatures associated with threats such as plastic explosives and spore-forming bacteria. Since this system is based on integrated circuits, covers an unprecedented broad range of non-ionizing, safe and low-power frequencies (10 to greater than 500 GHz) and can distinguish among a large variety of concealed threats on personnel (van der Weide 2001), it could prove useful as a field-deployable early-warning system for detecting hidden explosives or for the release of biological agents (Choi *et al*. 2002).

## 4. THz optics

Since most applications for THz technology use free-space propagation, managing THz 'light' is critical to success, especially because the power of THz sources is low, even though the dynamic range of THz systems can peak at 60 dB. Far-field imaging and sensing constitute the majority of current THz activity, though researchers have also employed near-field techniques such as using sharp, conductive probes as near-field antennas to localize THz energy, in some cases to sub-micrometre extents. It is also possible to generate and detect near-field THz light with sub-wavelength-sized crystals and excitation beams.

Far-field optics in the visible spectrum are much larger than the wavelength of light; this is not true in most THz systems, and the situation is further complicated by the greater than 10:1 range of wavelengths common to pulsed THz technology.

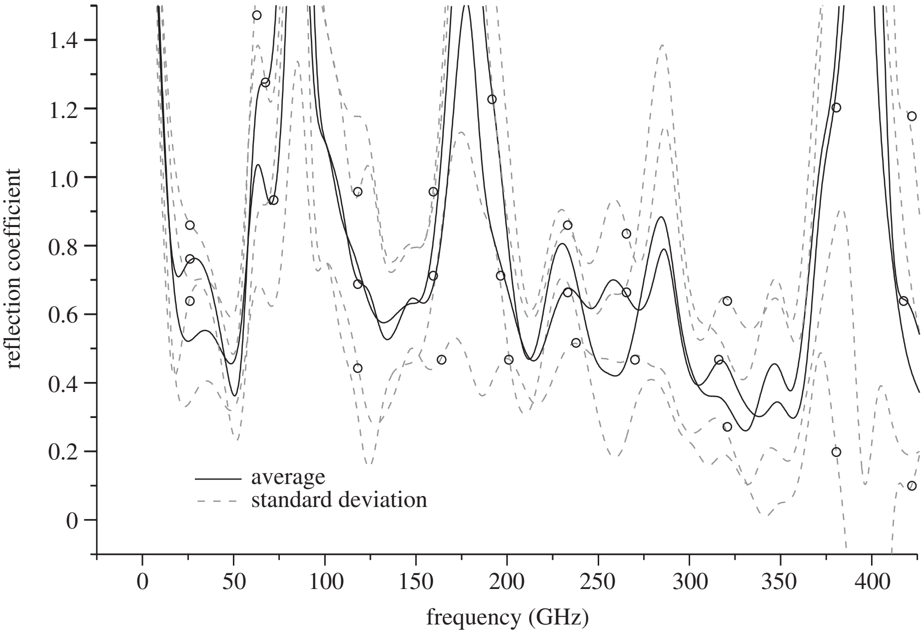

Figure 2. Overlay of C-4 (solid lines) and RDX (solid lines with circles) signatures in a 1 mm$^2$ sample from 5 to 450 GHz shows similarity between the reflection coefficient magnitudes, even though their morphology is distinct and there are standing-wave effects that cause the reflection coefficient to exceed unity. Dashed lines are standard deviations; solid lines are the average of eight trials with repositioning the sample between each trial. ($P_{\text{in}} = -7.8$ dBm, fundamental frequency = 7.75 GHz, 60 harmonics, 8 datasets.)

Transducing, guiding and focusing this broad range of wavelengths with high efficiency down to the diffraction limit is difficult: metallic losses and modal dispersion in the source and detector substrates tend to attenuate the higher frequencies, while the limited spatial extent of reflective optics permits diffraction of longer wavelengths. New techniques such as photonic bandgap structures and ultrabroadband antennas are being incorporated into THz systems to improve optical management. By controlling the dimensions of the THz beam (i.e. the antenna pattern), THz imaging and sensing systems will realize considerable gain, even with their current power limitations. This, in turn, will create new opportunities for greater stand-off distances in remote sensing.

## 5. Explosives detection

In contrast to the more common hybrid optoelectronic techniques that use lasers for generating broadband radiation (Fattinger & Grischkowsky 1989; Grischkowsky *et al.* 1990), our technology is based on integrated circuits. Whether optoelectronic or purely electronic, broadband (as opposed to single-wavelength) imaging has the chief advantage of flexibility: if threats change composition over the years, a single-wavelength or narrowband source may no longer detect the new composition, but having a broad range of frequencies maximizes the opportunity to detect the new threat's signature. For example, figure 2 shows the broadband reflection spectra we measure from 7 to 450 GHz, which highlights the similarity between plastic explosives C-4 and RDX; both spectral signatures are distinct from HMX, demonstrating specificity in the dielectric response of these targets (van der Weide *et al.* 1999). While these results are clearly complicated by standing-wave effects, the ease of distinguishing these signatures from those of skin makes for a potentially powerful new detection technique.

## 6. Detection possibilities for biological agents

Biological macromolecules are expected to exhibit weak absorptions, if any, in the microwave and THz regimes. Proteins are weakly dielectric and exhibit absorption due to orientational relaxation at radio frequencies (Pethig 1979). Double-helical DNA does not exhibit such orientational relaxation, since the dipole moments of the two helices nullify each other. Normal mode analysis predicts that both proteins (Tama *et al.* 2000) and DNA (Bykhovskaia *et al.* 2001) should exhibit multiple absorptions in the THz range due to a variety of collective, vibrational, twisting and librational modes.

All biological macromolecules, however, are surrounded by one or more shells of bound water (Pethig 1979); molecules in solution are also bounded by bulk solution. Water is a strongly dielectric molecule, with absorptions throughout the microwave and THz regimes (Segelstein 1981). Normal mode analysis predicts multiple absorptions for water in the microwave and THz ranges (Kindt & Schmuttenmaer 1997). Hence the primary contributor to the absorption of any solvated biological macromolecule is water, either in bound or bulk form. The observed spectra will be a combination of absorptions from bound and/or bulk water, the biological molecule(s) and interfacial dielectric phenomena. Several authors have described responses of DNA and proteins at microwave and THz frequencies (Pethig 1992; Markelz *et al.* 2000; Nandi *et al.* 2000; Taylor & van der Weide 2001*a*, *b*; Wichaidit *et al.* 2001).

Cells and tissues are complex mixtures of biological macromolecules and water. Spectra from such samples are not amenable to simple analysis, but differences in hydration may be used for imaging and/or identification purposes (Han & Zhang 2000; Smye *et al.* 2001; Fitzgerald *et al.* 2002). Thus, as the complexity of the sample increases, the potential for unique identification decreases, unless multiple frequencies can be used to correlate the spectral signature unambiguously to the response of the sample.

### (*a*) *Bacterial spores*

#### (i) *Sensing in transmission*

To examine the content of envelopes with an eye toward distinguishing common powders from potentially dangerous ones, we prepared samples of sugar, starch, flour and talcum powder. We performed transmission measurements through envelopes with the same four powders and with *B. cereus* (BC), a simulant of *B. anthracis.* We were able to see a negative dispersion in the transmitted phase of the BC in contrast to other common powders (Choi *et al.* 2002).

The transmission measurement set-up is shown in figure 3. Transmission through samples was normalized by the detected signal using empty containers (e.g. envelopes) in the beam path.

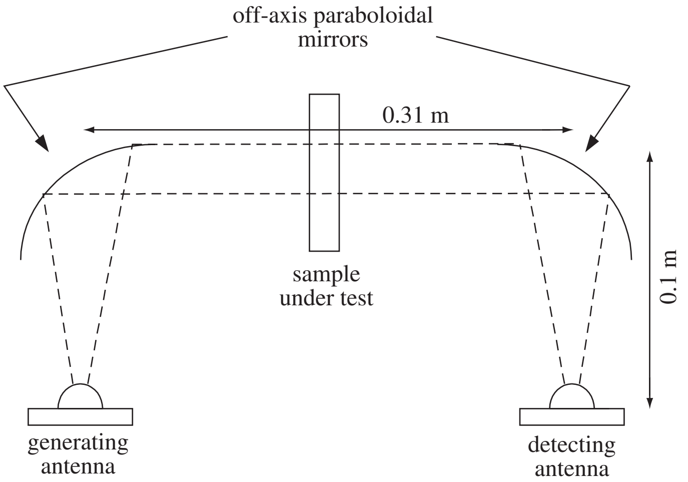

Figure 3. Transmission measurement set-up diagrams: without sample.

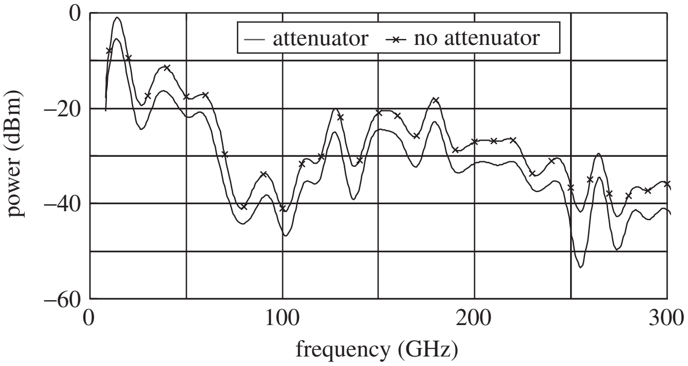

Figure 4. Transmission with and without a 300 Ω per square attenuating sheet.

As we have seen, standing-wave effects cause the ratios of reflection coefficient magnitudes to have magnitudes greater than 1 at some frequencies. One approach to reducing the effects of standing waves is to perform time-gating of the pulse, which is to measure only the initial transmitted (or reflected) pulse, then shutting off the receiver. An alternative to this is inserting attenuators into the beam path, at the expense of signal-to-noise. To check for uniform attenuation, we inserted a Mylar resistive sheet (300 Ω per square) in the beam path (figure 4). The attenuation was 5 dB at most frequencies, but small variations are clear, and these are magnified when taking the ratio of two measurements.

With this configuration, we examined samples both in petri dishes and in sealed envelopes. The normalization for the petri dishes was done by using an empty petri dish and, for the envelope measurement, the reference values were taken with an empty envelope at the sample position as in figure 3. While we observed inconclusive magnitude spectra for flour, starch and powders of BC, the phase signal showed a lower dispersion for the bacterial spores than for the other common powders (figure 5). This highlights the potential of using coherent measurements that can distinguish both components of the dielectric function. This signal, in conjunction

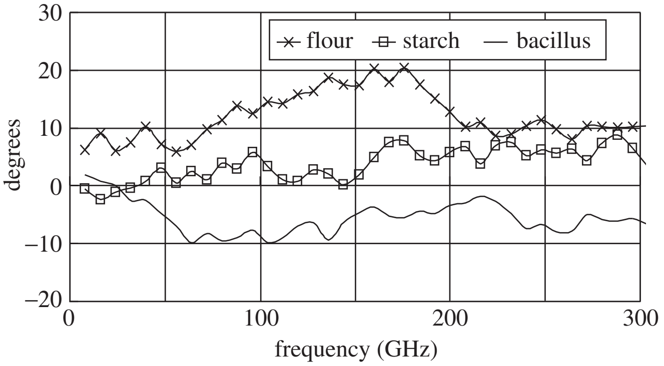

Figure 5. Normalized and unwrapped transmission phase for samples in envelopes (Choi *et al.* 2002).

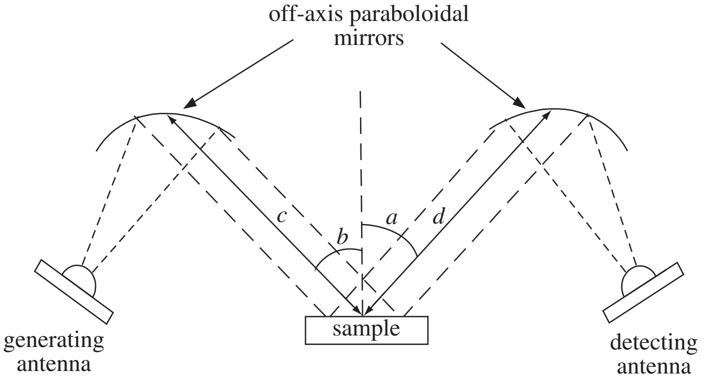

Figure 6. Reflection measurement set-up: $a = b = 45^\circ$, $c = d = 0.254$ m.

with measuring transmission characteristics of neighbouring pixels, would provide additional information about potential threats in envelopes.

### (ii) *Sensing in reflection*

For both lossy and highly conducting materials, reflection measurements are preferable, since transmission measurements may have a signal-to-noise ratio that is too low. As can be seen in figure 2, the reflection magnitude shows some differences in spectral patterns, part of which can be attributed to standing waves when normalized (hence the greater than 1 reflection coefficient magnitude). In addition, the phase information can potentially be used to identify the sample powders.

We measured *B. globigii* (BG) and *B. thurengiensis* (BT) on glass cover slips, and we found that at several frequencies we could distinguish among them. The reflection measurement set-up we used for this experiment is shown in figure 6. First, we used a blank glass slide to support the samples and to obtain a magnitude and phase reference, then we tested a series of cover slips, some empty and some with very light coatings of spores. We collected five sets each of magnitude data for the empty slip, as well as for slips with small quantities of BC, BG and BT (see figure 7). At many frequencies there were clear distinctions between the empty cover slips and

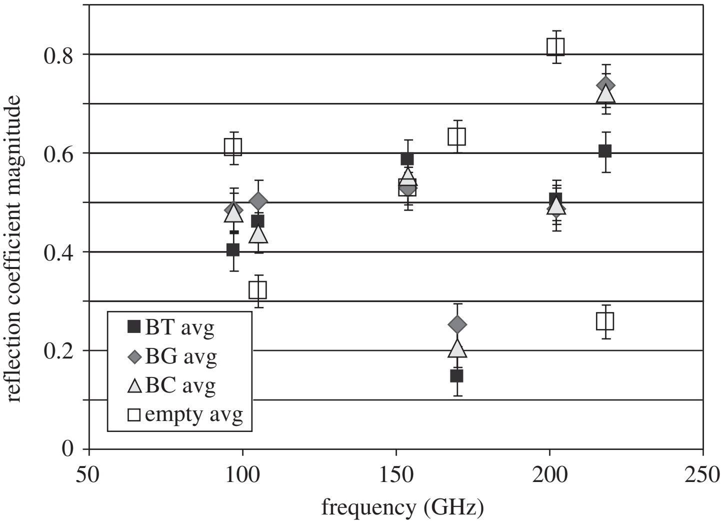

Figure 7. Representative reflection data for a variety of spore-forming bacterial powders on glass cover slips versus an empty cover slip, all normalized to the reflection from a glass slide support. Line at 150 GHz indicates ambiguity between these conditions, while other lines show clear distinction between spores and an empty cover slip.

those with spores, and distinctions among the spores were also visible to a much lesser extent. This provides evidence that a spectrometer of this type could perform point detection of collected spores. Variations in reflection coefficient of the cover slips versus frequency will arise from Fabry–Perot effects.

## 7. Discussion

We have surveyed reflection and transmission of radiation from a pulsed electronic spectrometer whose output falls in the 'lower THz' regime where free-space propagation for large stand-off detection would be achievable. We have found spectral signatures both in the magnitude and in the phase of these measurements that suggest the possibility of remote or point detection of dielectric contrast from concealed threats that could be distinguished from the background.

Spectroscopic detection or imaging with integrated-circuit broadband micrometre- and millimetre-wave sources and detectors could be used to measure dielectric reflection spectra of plastic weapons and explosives. With advances in coherent generation and amplification, it will be possible to develop arrays of these sources and detectors in an imaging system to offer a new screening technology. Furthermore, the potential advantages of this new approach to security over other microwave imaging techniques include smaller size, lower cost and potential for integration directly into existing security portals, all resulting from an integrated circuit approach that uses no moving parts.

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## *Discussion*

A. Crompton (*Cybernetic Machine Group of British Computer Society, East Harptree, UK*). I have two questions on biological applications. What is the potential for the direct observation of biological membranes and biological processes, and how do your spectra compare with NMR spectra?

D. W. van der Weide. We are looking directly at membranes and membrane proteins, and believe there is considerable potential. Obviously, NMR is a more advanced technique, with a higher signal-to-noise ratio as it has been commercially developed. We are looking for localized interactions and ones that do not require a magnetic field, which NMR cannot do. We are mostly exploring this for work in the field, which is very hard to undertake with a 400 MHz NMR spectrometer.

H. Roskos (*Physikalisches Institut, Johann Wolfgang Goethe-Universität, Frankfurt-am-Main, Germany*). Could you comment on the reproducibility of measurements on explosives and other hazardous materials?

D. W. van der Weide. We attempted to quantify repeatability by making several measurements and plotting not only the average but also the standard deviation. There's definitely something there even as we reposition the sample. We were confident enough that we chose to publish our data. However, very few people can get hold of these hazardous samples, and the community really needs to make measurements of these kinds of materials on different systems.

K. Unterrainer (*Institute of Solid-State Electronics, Vienna University of Technology, Austria*). How sensitive are you to standing waves in your reflectometer?

D. W. van der Weide. We're quite sensitive. This is a very important point—even though we make great efforts to damp out reflections by using absorbing boundary conditions, etc., there still is a cavity set up between the reflecting surfaces of the receiver, that of the transmitter and that of the sample holders. So there is a lot of potential for standing waves, which, of course, are always going to contaminate any measurement unless you do time-gating, which we have not yet implemented.

S. Withington (*Cavendish Laboratory, University of Cambridge, UK*). On the issue of standing waves, you get a similar problem between mixers and local oscillators, and in an attempt to understand this we've set up full models of the situation where you've got a horn facing a horn. So you use a modal matching technique to do the modal scattering down a horn, then free space modes, and then modal matching again down the next horn. And, of course, what you get then is a full scattering matrix which you can diagonalize, you can find what the eigenmodes are, and, in actual fact, what you find is that the frequency dependence of these is not simple. You don't just get a simple sinusoidal behaviour, but very sharp discontinuous resonant features which could easily be mistaken as spectral lines. The different modes have different cross-sectional forms and therefore you can, for example, put in absorbing apertures at the sample and even design the shape of the horn such that you prevent these resonant modes.

D. W. van der Weide. I agree with that entirely and I would say that the challenge thus far has been to create a low enough pulse repetition rate source that we can do time-gating, which would obviate the whole standing wave issue providing the ring-down was short enough.

G. Smith (*Department of Physics and Astronomy, University of St Andrews, UK*). I have a comment and a question. On the issue of standing waves, in our laboratory we use free-space quasi-optical isolators. They certainly work up to 300 GHz, I'm pretty sure they are extendable up to 500 GHz, and they've been very successful in removing a lot of these problems. How much power is typically going out in your nonlinear transmission line, and what are the typical efficiencies in generating power?

D. W. van der Weide. One concern is that using diffuse waves, rather than standing waves, makes a lot of difference too. If there was the possibility of a photonic bandgap THz diffuser, for example, that could obviate some of these issues. The nonlinear transmission lines that we build are pumped with roughly 0.5 W input power and have a total integrated power at the output of a few milliwatts. They're not really efficient generators but they're very broadband and that seems to be the trade-off. Our approach has been to survey the spectrum, see where the 'sweet spots' are, and design a frequency multiplier that can give us power in those harmonics and nowhere else. We can thus put the power into the spectral lines that are important to us.