Pulsed terahertz bi-directional reflection distribution function (BRDF) measurements of materials and obscurants

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ABSTRACT

We report a pulsed method to measure reflection and scattering from several samples with different degrees of surface roughness and material properties at terahertz frequencies. Reflection from a flat gold mirror shows that the full width half maximum (FWHM) of the terahertz beam angular spread is < 4° for frequency range 0.2 THz to 3 THz with signal-to-noise of 65 dB. Measurement of a paper index card, used as a low scattering sample, shows that the reflection/scattering properties are essentially similar to the system signature response except for multiple reflections between the front and back surfaces of the sample. Sixty-grit sandpaper shows multiple scattering events with almost no signal reflected from the flat backing paper surface. Corduroy cloth shows periodic reflections in the time domain, which correspond to diffraction lobes in the spectral domain.

Keywords: terahertz imaging, BRDF, concealed threat detection, terahertz reflection and scattering

1. INTRODUCTION

Pulsed terahertz technologies, conventionally referring to the generation and detection of picosecond pulsed electromagnetic waves using ultrafast lasers, have found various applications in spectroscopy and imaging. In most cases, pulsed terahertz systems are designed to measure the transmission or reflection of a single cycle terahertz pulse through and from the surface of the sample at a fixed angle. While measurements at a fixed angle do not affect the results for most laboratory samples, samples that are inhomogeneous, porous or composite could yield ambiguous information due to diffraction and scattering. These materials are common and found in everyday use such as cardboard, cloth and plastic foam. Hence, understanding the scattering properties of these and other materials is important for security and related imaging applications.

Several groups have reported simulations and measurements of the scattering of terahertz frequencies by inhomogeneous, rough media. Pearce et al., reported terahertz scattering of a material composed of randomly distributed Teflon spheres and found that by calculating the angular correlation function, it is possible to identify the occurrence of a single scattering event. Cheville et al. measured reflection/scattering from a single dielectric sphere and found that it is possible to separate different scattering events occurring at different locations on the sphere. Shen et al., showed that chemical fingerprint could be distorted and attenuated inside an inhomogeneous medium if the particle size is comparable to the incident wavelength. By averaging different points in the terahertz image, they show that it is possible to reduce the effect of scattering from the chemical fingerprint. An improved method of convergence based on wavelet transform has been proposed by Arbab et al., studying the effect of humidity on scattering from cloth filaments of incident terahertz signals. Bjanarson et al., measured the extinction of cloth samples made from different materials. Zuck et al., conducted finite-difference time-domain (FDTD) simulations and experiments on the effects of diffraction and scattering of reflected terahertz pulses from coated sandpaper and the plastic explosive C4. Dean et al., constructed a quantum cascade laser based imaging system that shows that powders of different sizes exhibit distinctive scattering intensities in comparison to the corresponding specular reflection images.

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The bidirectional reflectance distribution function (BRDF) has been used to minimize instrument-related contributions to reflectance measurements of surface roughness in the visible and near-infrared spectral ranges.\textsuperscript{11,12} In computerized modeling applications,\textsuperscript{13} BRDF measurements permit accurate simulation of reflection properties of a variety of materials, and hence a realistic virtual presentation. While the BRDF technique has been used in radio-frequency, visible, near- and mid-infrared spectral ranges, similar measurements are lacking in the terahertz/millimeter wave regime due to the lack of powerful sources and sensitive detectors. Hence computerized simulation of terahertz/millimeter wave imaging systems often have to resort to using material properties measured in homogeneous medium or by extrapolation using parameters obtained in different frequency ranges. Such methods often do not yield accurate predictions of the actual reflected power and accurate THz images.

In this work, we designed and constructed an ancillary set of rotatable optical mounts into a THz imaging system to permit collection of scattered pulses from samples. The eventual goal of our studies is to use the same samples for study by our pulsed apparatus (NIST Gaithersburg) and a tunable single-frequency apparatus based on frequency multiplication (NIST Boulder). Direct comparison of the BRDF scattering maps from these results enables us to build confidence in determining scattering efficiencies across large angular spreads and to eventually incorporate these scattering measurements into advanced modeling codes.

## 2. METHODOLOGY

A home-built pulsed terahertz system (Figure 1) was modified to scan reflected/scattered terahertz pulses at varies angles while maintaining fixed emitter and detector positions in the optical system. A commercial Ti-Sapphire regenerative amplifier with output pulses at central wavelength of 800 nm, repetition rate of 1 KHz, pulse width of 50 fs and pulse energy of 1.8 mJ is used to generate and detect scattered terahertz waveforms. 90\% of the laser energy is used to induce photoconductive breakdown on a large area (37.5 × 37.5 mm\textsuperscript{2}, 350 \(\mu\text{m}\) thick), semi-insulated GaAs photoconductive antenna which is biased by a high voltage, pulsed transformer at 8000 V. Selectable horizontally or vertically polarized radiation (p or s-polarized to the sample surface, respectively) is generated and polarized along the direction of the bias field. The antenna is coated with clear nail polish (main ingredient: Acrylate co-polymer) to prevent air-borne dust from causing substrate breakdown. A delay generator producing triggering TTL pulses at every fifth laser pulse is used to modulate the transformer and a liquid crystal shutter at 200 Hz. 5\% of the laser output (front reflection from a wedged quartz splitter located before the liquid crystal shutter) is used to gate the scattered terahertz pulses using a 2 mm thick, ZnTe <110> crystal with electro-optic sampling\textsuperscript{14}. In the absence of a terahertz field, the polarization of the gating laser beam after the ZnTe crystal is rotated by a quarter-wave plate such that its output after a calcite polarizer is minimized. Under the influence of terahertz pulses, the instantaneous electric field of the terahertz pulse induces birefringence within the ZnTe crystal and causes changes in the gating beam polarization. The terahertz modulated gate beam intensity after the polarizer is detected by an unbiased silicon photodiode connected to a lock-in amplifier synchronized to the 200 Hz trigger signal. Careful shielding and grounding of the antenna was performed to reduce long-lasting (ca. >1\(\mu\text{s}\)), low frequency emission (ca. >15 GHz) which helps to reduce low frequency artifacts and noise. In this way, the relative peak power-to-noise ratio of the system for a gold mirror sample was measured to be about 65 dB. With this sensitivity, single time delay scans were adequate to observe weak (<40 dB below incident peak value) sample scattering and time-angle maps (typically 30 ps delay scans at up to sixty angles) were collected in about 40 minutes. In most cases, single time sweeps at each scattering angle were adequate to obtain time-dependent profiles of the scattered radiation and fast Fourier transforms (FFT) were applied to extract frequency maps. When signals were smaller, five or more time scans were averaged to obtain the resultant time-domain information before FFT. At this point, we have not performed detailed uncertainty analyses of all time and FFT power measurements.
Figure 1 Terahertz BRDF system schematic. Terahertz pulses are generated by photoconductive breakdown of a large area GaAs antenna by the incident laser beam. A small portion of this beam is split to detect the reflected/scattered THz field by electro-optic sampling with a ZnTe crystal. Measurement of scattering from the sample is performed by angularly scanning the output from the sample with the add-on shown in Figure 2.

To measure BRDF values from a sample under test, an additional mechanism is added to this pulsed terahertz system (Figure 2). The sample, attached and protruding by ~8 mm from a standard 50 mm mirror mount with a beveled Teflon mounting ring is placed on a rotational stage at an angle to the incident terahertz beam. To sample the reflected/scattered terahertz signal, a rotating arm holding a pair of periscope mirrors is rotated around the sample. The output from the periscope mirror pair is then redirected by a mirror that is rotating in the same direction at half the angular step of the periscope arm. To prevent undesired scattering from mechanical components in the add-on, two irises, one in front of the sample and the other 24 cm from the sample and located in-between the two periscope mirrors are placed in the terahertz beam path. The opening diameter of the input and output irises are 15 mm and 20 mm, respectively. The size of the incident beam on the sample is thus limited by the input iris to ~15 mm.

To eliminate the instrumental variation, the BRDF is defined as division of the reflected radiance $L_r$ by a sample surface over the total incident irradiance $E_i$ with the power independent unit of 1/sr. Mathematically, it is expressed as

$$f_r(\theta_i, \phi_i; \theta_r, \phi_r) = \frac{dL_r(\theta_i, \phi_i; \theta_r, \phi_r)}{dE(\theta_i, \phi_i)}$$

where $E_i = d\Phi / dA$ and the incident irradiance is defined as the incident flux divided by the illuminated area $A = \pi(7.5 \text{ mm})^2 / \cos30^\circ$ in W/m$^2$. The reflected radiance $L_r = d^2\Phi_r / d\cos\theta_r d\omega$ in W/m$^2$sr is defined as the output flux divided by the reflecting sample area projected to the output angle and the solid angle extended from the sample surface to the limiting size in the beam path of the detection optics. $\theta_i, \phi_i$ and $\theta_r, \phi_r$ are the incident and detection angles with respect to the sample front surface. Hence, a perfectly reflecting mirror would yield a BRDF value of an inverse solid.
angle at the specular reflection peak. In our case, the output solid angle is limited by the output iris placed between the two periscope mirrors (Figure 2). Given the 20 mm diameter output iris and the distance from the sample front surface to this iris, 178 mm, the solid angle is calculated as $\pi(10 \text{ mm})^2/(240 \text{ mm})^2 \approx 5.4 \times 10^{-3} \text{ sr}$.

![Diagram of terahertz system](image)

Figure 2 BRDF add-on to the pulsed terahertz system. (a) Schematic diagram of the add-on. Reflection and scattering of the incident THz beam is sampled by a periscope mirror pair. The resulting THz scatter is redirected by the top mirror to be measured by the ZnTe electro-optic gating laser pulses. (b) A photograph of the BRDF add-on.

In Figure 3, we show the experimentally determined system signature or instrument background of our BRDF setup. The terahertz beam is incident at the sample at 30° to the surface normal and is assumed to be a plane wave with a Gaussian beam waist of 7.5 mm. The measurement is conducted using a flat gold mirror with the specular reflection spectrum used as the incident flux $\Phi_i$ and reflection/scattering spectrum at all angles used as the reflected flux $\Phi_r$. As the pulsed terahertz system is a time-domain signal, the angularly dependent time domain signals $E(\theta, t)$ are converted to the frequency domain $E(\theta, f)$ by a Fast Fourier Transform (FFT) algorithm. Note that the pulse terahertz system directly measures the field variation of the incident terahertz pulses, in order to obtain the incident and output flux $\Phi(\theta, f)$, one has to take the square of the obtained spectral amplitude $\Phi(\theta, f) \propto |E(\theta, f)|^2$. Hence, by substituting the ratio of the square of the reflected and incident spectra into Eq. 1, we are able to obtain the reflection/scattering spectrum map in BRDF units. For most of the detectable spectrum > 0.2 THz, the distribution is relatively constant at full width half maximum (FWHM) of < 4°, indicating that the incident terahertz beam is well collimated. The signals begin to attenuate heavily beyond ±5° as only pure specular reflection occurred from the gold mirror. Below 0.2 THz the incident beam begins to broaden, which is likely due to two reasons: 1. The wavelength of this frequency range approaches the size of the input iris, and hence the plane wave assumption is no longer valid. 2. The low frequency fluctuations of the pumping laser intensity, which is caused by fluctuation in the chiller water cycle, airflow in the laboratory, diode pumping current etc. The overall effect is a slowly varying fluctuation in the multiple minute regimes, which could be reduced by scanning the measurements faster and taking multiple scans under the same conditions.
Lastly, the presence of vertical bands at 1.2 THz and 1.4 THz is due to water vapor absorption lines present in this frequency range.

Figure 3. (a) Angular dependent time domain map of the terahertz reflection from a flat gold mirror. The terahertz is incident at an angle of 30° to the surface and the time delay is 0 ps. Signal ringing after 0 ps is due to water absorption in the terahertz beam path. (b) The corresponding BRDF map for the time window of -15ps to ~10ps. The frequency resolution of the system is currently limited to 80 GHz due to the existence of an internal reflection peak from the GaAs generator at 12 ps.

3. RESULTS

A paper index card of 0.2 mm thickness is used as a low-scattering sample. Comparison of the specular reflection peak amplitude of the front surface (Figure 2(a)) to that of the gold mirror shows that the peak reflectivity is roughly 1/7 = 0.14%. Using Fresnel reflection theory, assuming a negligible imaginary part of the refractive index and scattering loss, the real part of refractive index is estimated to be ~1.4. The much weaker and broader back surface reflection peak at 2.2 ps, corresponds to the portion of the terahertz signal that is transmitted from the front surface and subsequently reflected by the back surface of the index card. The broadening of this reflected pulse in comparison to the first front surface reflection peak and the lack of features at off-specular angles suggests that most of this broadening is due to the intrinsic dispersion and absorption of the composite materials of the card. The optical path difference between this secondary reflection peak and the reflection peak at the front surface is calculated as $2nd/\cos\theta_1 = 2 \times 1.4 \times 0.2 \text{ mm}/\cos (30°) = 0.65 \text{ mm}$ which corresponds to a 0.46 THz modulation in the spectral domain as seen in Figure 2(b). Here nd is the single pass optical path length defined as the multiplication of sample refractive index and sample thickness.

Commercially available, 60-grit aluminum oxide sandpaper was also used as a sample with high scattering loss. If the aluminum oxide front surface is perfectly smooth, then using Fresnel reflection theory and the refractive index value of 2.1 from the literature, the front surface reflectivity should be ~30%, which is much higher than the measured signal strength of ~10%. This rough estimation and appearance of scattering lines emanating outward after the front surface reflection (see Figure 5(a)) indicated the occurrence of scattering events inside the sample. The now strongly attenuated and broadened back-surface reflection peaks at ~5 ps delay, indicated that the terahertz signal is severely attenuated by the scattering loss and out-of-phase interference of the terahertz pulse from different tangential locations on the sandpaper. The corresponding spectral domain map in BRDF units (Figure 5(b)) shows severe attenuation at a frequency range beyond 0.7 THz, which is consistent with an estimated average particle size of 269 μm.
As an example of periodic sample, we measured scattering from a corduroy cloth with lobe spacing of 3.18 mm (Figure 6). A comparison between the peak amplitude of the front reflection between the corduroy and the gold mirror gave a reflectivity value of ~0.05/7=0.7%, which corresponds to a refractive index of <1.1. As with the sandpaper, we expected that this value significantly underestimated the actual refractive index of the composit materials in the cloth sample due to a low filling factor and significant diffraction of the reflected terahertz signal by the periodic undulating surface.

Unlike using a narrow-band continuous wave (CW) source, the terahertz pulses of our system are almost single cycle (ca. 1 ps FWHM) and broadband (spanning ca. 0.1 to 2 THz). Hence, little or no constructive or destructive interference can occur as there is minimal time overlap between pulses reflected from the different grating lobes. The periodic grating structure, however, introduces a periodic spacing between each scattered pulse from different locations at the surface. This geometry results in pseudo CW pulse trains at each angle that are as wide in time as the number of grating lobes.
under illumination (Figure 4(a)). Although there is no temporal overlap between scattered terahertz pulses, the BRDF map exhibits strong diffraction-like behavior as predicted by the grating diffraction equation

\[
\sin \theta_m = \sin \theta_i + \frac{m \lambda}{d},
\]

where \( m \) is the diffraction order, \( \lambda \) is the wavelength of the incident electromagnetic wave, \( d \) is the grating spacing, \( \theta_i \) and \( \theta_m \) are the angles of the incident beam and \( m \)-order diffracted beam with respect to the normal of the grating surface. For this particular corduroy cloth, only the \( m = 0, \pm 1 \) orders are clearly seen, however the diffraction efficiency is fairly strong at roughly one order less than the specular reflection peak at low frequency (<0.2 THz) and equivalent in magnitude at high frequency (0.3–0.6 THz).

![Figure 6](Image)

Figure 6. (a) Angular dependent time domain reflection/scattering from a corduroy cloth. Around 7-8 reflected pulses are clearly seen at large off specular angles (> 50° & < 10°) which corresponds well with the illuminated area on the corduroy cloth surface. (b) The BRDF map for the same sample. Black lines indicate the calculated diffraction angle for diffraction orders of \( m = 0, \pm 1, \pm 2 \). Vertical traces at a frequency of 0.55 THz is due to water vapor absorption in the laboratory environment.

4. CONCLUSIONS

We successfully developed a time-domain terahertz method to measure reflection and scattering from low reflectivity samples. In the current implementation, the sensitivity of the system is calculated to be near 65dB. In principle, one may also readily convert the current system to measure transmission characteristics and polarization effects from samples by rearranging routing mirrors. One advantage of the time domain method presented here is that spurious signals due to reflection, diffraction or refraction from components in the optical system are easy to isolate and eliminate. These artifacts are difficult to identify using a conventional continuous wave BRDF approach.

The frequency resolution of the system in its current incarnation is primarily limited by the internal reflection in the GaAs generator. For samples that do not exhibit strong diffraction or scattering or have sharp dielectric properties, this problem is not a limiting factor. However, as we can see from the measurement from sandpaper and corduroy cloth, the diffraction of the main terahertz pulse and the first generator’s reflected pulse overlap at large off-specular angles, and hence introduce errors in the BRDF measurement. This problem will be addressed in the future by using a thick, wedged GaAs wafer as the generator. To reduce the intrinsic phonon absorption in the generator, we plan to utilize a reflection scheme where the generated terahertz beam is counter-propagating to the incident pump beam used for our scattering measurements.
As we directly measure the electric field distribution, in principle we obtain both the spectral amplitude and phase information from a sample. Up until now, we only present the reflected amplitude information which is equivalent to conventional BRDF measurement. The phase component, however, could provide additional information such as occurrence of individual scattering events. Currently the accuracy of the phase information is limited by the delay shift caused by the centering error among the three rotational stages. This error could be further corrected by a proper alignment procedure or by reducing the positioning flexibility using a simpler scanning mechanism such as a fiber coupled photoconductive detector.

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