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Multiple Scattering of Broadband Terahertz Pulses

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Abstract

Propagation of single-cycle terahertz (THz) pulses through a random medium leads to dramatic amplitude and phase variations of the electric field because of multiple scattering. We present the first set of experiments that investigate the propagation of THz pulses through scattering media. The scattering of short pulses is a relevant subject to many communities in science and engineering, because the properties of multiply scattered or diffuse waves provide insights into the characteristics of the random medium. For example, the depolarization of diffuse waves has been used to form images of objects embedded in inhomogeneous media.

Most of the previous scattering experiments have used narrowband optical radiation where measurements are limited to time averaged intensities or autocorrelation quantities, which contain no phase information of the pulses. In the experiments presented here, a THz time-domain spectrometer (THz-TDS) is used. A THz-TDS propagates single-cycle sub-picosecond pulses with bandwidths of over 1 THz into free space. The THz-TDS is a unique tool to study such phenomena, because it provides access to both the intensity and phase of those pulses through direct measurement of the
temporal electric field. Because of the broad bandwidth and linear phase of the pulses, it is possible to simultaneously study Rayleigh scattering and the short wavelength limit in a single measurement.

We study the diffusion of broadband single-cycle THz pulses by propagating the pulses through a highly scattering medium. Using the THz-TDS, time-domain measurements provide information on the statistics of both the amplitude and phase of the diffusive waves. We develop a theoretical description, suitable for broadband radiation, which accurately describes the experimental results. We measure the time evolution of the degree of polarization, and directly correlate it with the single-scattering regime in the time domain. Measurements of the evolution of the temporal phase of the radiation demonstrate that the average spectral content depends on the state of polarization. In the case of broadband radiation, this effect distinguishes photons that have been scattered only a few times from those that are propagating diffusively.
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Chapter 1

Introduction

1.1 Scattering of Waves

The propagation of electromagnetic waves through inhomogeneous media is a topic of considerable interest to a number of research communities. Inhomogeneities within the medium give rise to multiple scattering, which lead to a variety of interesting interference effects. We observe the effects of light scattering in everyday life. On a sunny day, the sky appears blue because of the wavelength dependent scattering of sunlight off of the molecules in the atmosphere. We are blinded when we shine our headlights into fog, because the light is backscattered strongly from the dense number of water particles in the air. The impressive display of the spectrum of colors in a rainbow is caused by the scattering of light from water droplets in the air.

Besides being a common phenomenon in nature, the issue of multiple scattering arises in many areas of research. In particular, imaging in the presence of scatterers is a topic of considerable interest. In the field of seismic imaging, for example, the quality of information that can be extracted is limited in part by the multiple scattering of acoustic waves as they propagate through different layers within the earth. Another example can be found in biomedical optics. For near infrared light, the human body does not absorb too heavily, but the light scatters strongly as it propagates. In these and in other examples, a good understanding of the statistical properties of the scattered radiation is necessary to retrieve quality images of objects buried within the random medium. Multiple scattering also leads to some interesting physics. In the limit of very strong
scattering, waves can become completely localized and cease to propagate out of the medium. The study of localized states of light has been a compelling research goal for many years.

Previous studies of electromagnetic wave scattering have relied on narrowband sources of microwaves [7-9] or visible light [10, 11] to probe random media. Measurements involving the visible spectrum are limited to time averaged intensity measurements or correlation quantities. Instruments do not have the speed necessary to measure the quickly oscillating electric field, which contains information on both the phase and the intensity of the radiation. Although, with microwaves, it is possible to directly measure the electric field, but the random media is typically placed in a waveguide geometry rather than a 3-dimensional sample as is customarily used with visible light [6]. Both microwave and visible light experiments involve narrowband sources, where the scattering parameters do not vary significantly over the bandwidth. Investigations of multiple scattering involving broadband radiation have been limited.

1.2 Terahertz Time-Domain Spectroscopy

T-rays, single cycle terahertz (THz) pulses, provide a unique source to study multiple scattering, because of their broad bandwidth, and the ability to directly measure their electric field. They lie in the region of the electromagnetic spectrum between microwaves and visible light, which is in the divide between photonics and electronics. T-rays have the best of both worlds in studying this subject. In addition, the longer wavelengths permit a higher degree of control over the properties of the random medium. Multiple scattering has not previously been studied with T-rays. A lack of efficient measurement techniques limited exploration in this spectral region. Recent advances in
semiconductor physics and ultrafast lasers have led to the development of a THz time-domain spectrometer (THz-TDS), which can generate broadband subpicosecond pulses of THz radiation and directly measure its electric field. This system provides a powerful tool to explore the broadband case of multiple scattering. The THz-TDS uses femtosecond optical pulses to generate THz radiation by gating a photoconductive switch. The radiation produced spans from 50 GHz to over 1 THz, which correspond to wavelengths of 0.3 mm to 3 mm.

One of the unique features about the THz regime of the electromagnetic spectrum is the spectral response of different materials. Many packaging materials such as plastic, styrofoam, and cardboard are virtually transparent to T-rays. Metals, such as aluminum, are very strong reflectors. Water absorbs very heavily in the THz regime. With submillimeter wavelengths, which are very long compared to visible wavelengths, permits easy manufacturing of samples on the order of the wavelength.

In this thesis, we describe the first measurements of multiply scattered THz pulses. This involves propagating THz pulses through collections of randomly distributed particles and measuring the scattered radiation. From the waveforms we collect, we measure statistics of the electric field. We compare the results to existing narrowband theories and extend those theories to address the broadband case. We also observe the influence to polarization from the scattering process and measure changes in the spectral content between the orthogonal polarizations.

1.3 Thesis Organization

In Chapter 2, we overview the details of the THz time-domain spectrometer and provide a background to its development. We include some potential applications of this
technology. Chapter 3 describes the basic concepts and theories of scattering. We discuss the scattering from a single particle as well as the propagation of waves through collections of scatterers. In Chapter 4, we describe our first measurements of diffusive T-rays. We measure the statistics of the electric field and extend the existing narrowband theory to address the broadband case. Chapter 5 discusses our investigation of the polarization dependence of diffuse T-rays. We observe a spectral shift between the orthogonal polarizations that can be used to distinguish between single scatter and diffuse photons. Finally, in Chapter 6, we discuss the results and suggest future directions of the research.
Chapter 2

Terahertz Time-Domain Spectroscopy

2.1 Historical Overview

Electromagnetic waves have been employed in a variety of ways to greatly improve the quality of life for many people. For example, radio waves have allowed millions of people to listen to their favorite talk radio personalities on their drive to work in the morning. The diets of college students primarily consist of food that can be heated with microwaves. Tourists use cameras to capture the visible light of a famous landmark and record a digital image of it. X-rays have saved many lives by aiding doctors in diagnosing patients' ailments. As one scans the electromagnetic spectrum (see Fig. 2.1) and views the enormous number of commercial products and applications available in each band of the spectrum, strikingly apparent is the lack of developments in the transition region between electronic and photonic frequencies. This radiation, which consists of frequencies on the order of $10^{12}$ Hertz or 1 terahertz (THz), exists in the far infrared directly between microwaves and visible light and is known as T-rays.

Lack of efficient sources and detectors have somewhat limited the exploration of the THz band. However, in the past two decades, novel techniques to generate and detect T-rays have opened new possibilities to develop devices to further exploit this region of the spectrum. The developments of ultrafast lasers and fast optical switches have provided a way to study this radiation and its interaction with matter. In the 1980’s, a series of experiments conducted by David Auston’s group in Bell Laboratories led to the development of a photoconductive switch, now known as the Auston switch, which
emitted electrical pulses that are subpicosecond in duration and propagated them into free space [12]. Prior to the development of these switches, electro-optic Cherenkov radiation was used to generate and coherently detect pulses [13], however, there were no methods demonstrated to efficiently extract the signals from the electro-optic materials until years later [14]. The use of Auston switches to transmit and measure THz radiation fueled the development of the THz time-domain spectrometer (THz-TDS) as a tool to explore the THz regime. Several improvements to the system were made through the use of paraboloidal mirrors and collimating lens, which resulted in an increase in power of the radiation and detection efficiency [15-17]. Since then, many other techniques have been
developed to investigate the THz gap including THz-TDS with electro-optic detection [14, 18, 19] and quantum cascade lasers [20].

With the development of the THz-TDS and advancements in generation and detection techniques, the THz gap has become more accessible and allowed researchers to focus on studying these waves. According to Mittleman, “researchers are able to concentrate more on what one can do with THz radiation, and less on simply how to produce it [21].” From the advent of THz-TDS, a powerful tool is now available to allow the research field to move beyond learning how to merely produce THz radiation into focusing on the search for potential applications. In 1995, Hu and Nuss demonstrated the feasibility of applying T-rays to imaging by developing the first imaging system that utilized the THz-TDS [1].

Presently, industry and researchers alike are looking to employ this technology to solve real world problems. The relevance of the THz-TDS has expanded beyond mere laboratory applications. In the year 2000, the first commercial THz-TDS became available, which was developed by Picometrix, Inc [22]. This signified an important evolution in the technology from that of a purely research tool to a commercialized product. This has resulted in T-rays being used to solve many important problems. On February 1, 2003, the United States witnessed a terrible tragedy when the space shuttle Columbia crashed, resulting in the deaths of all seven astronauts on board. NASA investigators believe that a defect in the foam on the shuttle tank caused a piece of the foam to break loose and strike the left wing, causing the shuttle to explode upon reentry. Returning to space depends on the ability to ensure that the foam is safe and does not contain any defects. In a comparison of several possible technologies to locate these
defects in the shuttle foam, T-rays out-performed all of the competing technologies by successfully locating more of the defects in a sample piece of foam. This emphasizes that the technology has progressed from a purely research instrument into a tool capable of changing the world.

In the future, the THz gap will benefit from improvements in generation and detection schemes, which will result in the region being filled with more powerful applications that solve real world problems. Recently, T-rays were named one of “10 emerging technologies that will change your world” [23]. Already, T-rays are being considered for applications in security, gas spectroscopy, imaging, and many others. The THz gap is rapidly closing.

2.2 Terahertz Time-Domain Spectrometer (THz-TDS)

The fiber-coupled THz time-domain spectrometer (THz-TDS) used in the experiments described in this dissertation is depicted in Fig. 2.2. The system, which was developed by Picometrix, Inc. [22], allows for the transmitter and receiver to be moved easily without concern for the alignment of the optical pulses. This results in a highly reconfigurable system, which can perform numerous types of experiment geometries and imaging modalities.

The THz-TDS, one of the more common methods for transmitting and detecting T-rays, relies on femtosecond pulse trains and photoconductive switches to generate and measure the electric field of the THz radiation in the time-domain. The THz transmitter and detector used in a THz-TDS are essentially photoconductive switches that are triggered by ultrafast laser pulses. A THz transmitter, pictured in Fig. 2.3, is composed of a GaAs semiconductor substrate with a gold planar antenna patterned onto the surface
Figure 2.2: The components of a fiber coupled THz time-domain spectrometer. The spectrometer uses femtosecond pulse trains and a scanning delay line to measure the electric field of the THz radiation in the time-domain.

through a lithography process. The GaAs is grown at low temperatures to reduce the carrier lifetime, which results in a faster photoconductive switch [24]. The dipole antenna is DC biased to create a static electric field across the antenna.

When an ultrafast laser pulse impinges on the surface of the GaAs semiconductor at the transmitter, electron-hole pairs are created from the absorption of photons. Carriers are accelerated in the direction of the static electric field, producing a transient photocurrent. The current quickly rises because of the short duration of the optical pulse, but the fall time is slightly longer due to the slower electron-hole recombination [24, 25]. Maxwell’s equations govern that the electromagnetic field produced is proportional to the derivative of the photocurrent. Therefore, the antenna will emit short bursts of
Figure 2.3: The THz transmitter module. The transmitter consists of a lithographed gold dipole antenna patterned on a GaAs substrate. The fs pulse causes a transient current to flow through the antenna structure, which generates THz radiation. A spherical lens is used to collect the generated THz radiation and to direct it into free space.

Subpicosecond radiation at THz frequencies. The radiation is then collected by a silicon substrate lens that is attached to the GaAs substrate as shown in the front view of Fig. 2.2. Silicon is the chosen material for the lens, because its index of refraction ($n_{Si} = 3.4$) does not differ too substantially from GaAs ($n_{GaAs} = 3.6$) nor does it absorb as much as GaAs [26]. Using a material like silicon that has a similar index of refraction as GaAs, reduces Fresnel reflections between the lens and the GaAs. The emitted radiation exhibits primarily a dipole pattern [27] superimposed on a weaker quadrupole angular pattern [28].

The receiver structure is very similar to the transmitter, the primary difference being that the DC voltage supply of the transmitter in Fig. 2.3 is replaced with a current meter in the receiver. When an ultrafast laser pulse arrives at the detector, the resistivity of the GaAs reduces for the duration of the laser pulse. This opens a time window when the THz electric field at the antenna lines can induce a current. A sample of the electric field is obtained by measuring the induced current, which is proportional to the
convolution of the electric field with the time window. The width of the time window limits the measurable bandwidth of the THz pulse, because for higher frequencies that will oscillate several times within the sampling window, the current will average out to zero.

The THz-TDS system illustrated in Fig. 2.2 consists of an ultrafast laser system, a control box, a transmitter, and a receiver. The laser system generates 100 fs pulses with a central wavelength of 800 nm at a repetition rate of 80 MHz. Within the laser system, the pulses travel through a pair of reflection gratings to compensate for the group velocity dispersion in the fibers. This is necessary because the glass in the fiber will cause certain wavelengths to travel faster than others, which will elongate the laser pulses and result in a loss of THz bandwidth. The pulses are then coupled into fiber and sent to the control box. The control box contains an optical delay line that controls the pulse arrival time.
between the transmitter and receiver. Inside the control box, the laser pulse beam propagates through a beamsplitter, which sends half of the laser light directly to the transmitter where THz radiation is then emitted. The other half of the light reflects off of a mirror on the optical delay line and then travels to the receiver where the electric field of the THz radiation is then measured.

Current technology is unable to directly sample multiple points of a single THz pulse, because electronics do not exist that are capable of sampling at the femtosecond rates necessary to resolve the pulse. In order to acquire a single sample of a THz waveform, averaged measurements are made over multiple THz pulses generated from the femtosecond laser pulse train. Each generated THz pulse is nearly identical, however, variations in the pulses are a source of jitter noise in the measurements. The optical delay line inside the control box increases the time delay between the pulse arriving at the transmitter and the pulse arriving at the receiver where the electric field is averaged. By incrementally increasing the delay, the THz-TDS collects discrete samples of the THz electric field and maps out a complete waveform over a window in time. The sampling period is limited by the temporal width of the femtosecond pulse.

Shown in Fig. 2.4(a) is a typical time-domain waveform measured with the THz-TDS over a 50 ps delay window. The duration of the pulse is less than 1 ps. The multiple reflections that arrive after the main pulse are due to both optical and electrical impedance mismatches in the system, such as the silicon lens-air interface. Fig. 2.4 (b) shows the spectrum of the THz pulse calculated by taking the Fast Fourier Transform of the sampled waveform [29]. The spectrum shows that signal energy extends as far out as 1.5 THz. Because the THz pulse consists of only one or two cycles, its spectrum has a
broad fractional bandwidth, where the central frequency is roughly half of the bandwidth. Note the several absorption lines in the frequency response of the THz pulse. Because water absorbs very strongly in THz regime from rotational transitions, vapor in the air will attenuate certain frequencies in the spectrum [30].

In the ideal situation, the measured waveform in Fig. 2.4(a) would exactly represent the THz radiation produced from the transmitter. Unfortunately, limitations in the detector’s bandwidth and sensitivity reshape the pulse. A slow detector will smear out the pulse and increase its duration, which results in a loss of bandwidth of the measured THz pulse. The speed of the detector is dependent on the carrier dynamics of the photoconductive material, but more importantly, the geometry of the antenna, because the detector’s bandwidth is inversely proportional to the length of its photoconductive gap [27]. There has been much work done to develop fast detectors by investigating alternate antenna geometries [31] and by using non-linear electro-optic materials [32-35], which has resulted in systems that cover over 120 THz in bandwidth.

2.3 Applications of Terahertz Time-Domain Spectroscopy

THz time-domain spectroscopy as well as other newly developed techniques for accessing this spectral range has opened the door to explore the far infrared region of the electromagnetic spectrum and search for potential applications. The responses of many common materials to THz radiation have been fully characterized using the THz-TDS and have been found to contain very unique properties. Water absorbs very heavily at approximately 1 THz. This allows for many possibilities in biomedical applications. Cardboard and plastics are virtually transparent to THz radiation, which suggests potential applications to package inspection and security. Transitions between rotational
quantum levels in polar gas molecules result in spectral signatures at THz frequencies, which can be used for detecting and sensing gases [36]. New applications of THz time-domain spectroscopy and uses of T-rays are continually being explored.

Within the past several years, researchers have realized that the THz-TDS is an effective tool to characterize and estimate the THz optical properties of many materials [37, 38]. Dorney et al. demonstrated that both the optical properties (absorption, index of refraction) and thickness could be extracted from the time-domain measured THz waveform [37]. The Fabry-Perot effect describes propagation through a slab of material, which produces multiple isolated pulses in the THz waveform. Using this model, they apply a gradient search algorithm that employs a total variation measure to simultaneously extract the thickness and complex index of refraction to a high degree of accuracy. This approach could be used for the identification of unknown materials.

Analyzing gases with T-rays is a very active area of research [36, 39, 40]. Many gases exhibit unique resonant frequencies from rotational transitions in the THz regime, which results in absorption lines in the spectrum. These are unique spectral fingerprints, which can identify or detect certain potentially harmful gases. The broad bandwidth and sensitive detection of T-rays permit the characterization of multiple absorption lines in a single measurement, which makes the THz-TDS a powerful tool for gas spectroscopy.

Using T-rays in biomedical applications is a very promising field [41]. Unlike X-rays, T-rays are non-ionizing radiation, which reduces the health risks associated with operating X-ray machines. Because water is a strong absorber of THz radiation, different types of tissues can be distinguished from their water concentration. However, this also limits the penetration depth of T-rays into the tissue to a few millimeters. Ex vivo studies
of human skin with T-rays have shown that basal cell carcinoma can be distinguished from healthy tissue by measuring the absorption of T-rays through the tissue [42]. Further studies have focused on in vivo measurements [43]. Other healthcare applications include using T-rays to detect decay in tooth enamel [44]. The THz-TDS is a safe, non-invasive tool that has the potential to significantly impact in the biomedical community.

Recent events such as the terrorist attacks on New York City and the anthrax letter scare have fueled interest in the area of security. T-rays have the potential to provide many solutions, because they can penetrate through most packaging materials. One potential application is airport screening. Because clothing is virtually transparent to THz radiation, T-ray scanners are capable of detecting concealed weapons [45]. Researchers have also used T-rays to probe illicit drugs [46], investigate explosives [47], and detect potential biological hazards [48]. Recently, a THz endoscope was developed, which can probe small spaces with T-rays [49]. T-rays are well suited for a wide range of security applications.

2.4 Imaging with T-rays

Hu and Nuss sparked interest in using THz radiation for imaging when they built the first T-ray imaging system in 1995 [1]. Their setup employed a rapid scanning optical delay line and a digital signal processor (DSP). It could acquire up to 20 waveforms per second. Their system raster scanned an object and measured the transmitted THz radiation at each pixel. Shown in Fig. 2.5 are the first reported T-ray images taken with their system. The left image is of a semiconductor integrated circuit encapsulated in plastic. The plastic casing is virtually invisible to the T-rays. However,
Figure 2.5: The first T-ray images. The image on the left is a semiconductor integrated circuit chip encapsulated in plastic [1]. The T-rays penetrate the plastic case, but do not transmit through the metal leads. The two images on the right are of the same leaf: when it was first cut and then 48 hours later. After 48 hours, the water evaporates, which increases the T-ray transmission.

the metal leads are clearly imaged, because they are opaque to THz radiation. The right two images are of the same leaf taken at two different times: when the leaf was first cut and 48 hours later. The leaf had a higher concentration of water and thus absorbed more T-rays when it was first cut as opposed to 48 hours later. Over the 48 hour period, the water evaporated and thus the stems of the leaf become more visible as the leaf dries out.

While the system of Hu and Nuss was limited to a transmission geometry, further work by others resulted in a reflection mode T-ray imaging system [2, 50, 51]. For many imaging applications using THz radiation, it is often advantageous to work in a reflection mode, especially in situations when only one side of the object is accessible to detectors
Figure 2.6: (Top) A conventional T-ray reflection image of 3.5 in. floppy disk. (Bottom) The time-of-flight image of the disk at the vertical dashed line (y = 15 mm) [2].

or the object is opaque. In addition, a reflection mode can use the pulse arrival times to probe depth information of an object.

Mittleman et al. demonstrated that an object’s index of refraction profile can be extracted using the arrival times of THz pulses from Fresnel reflections at different layers of a composite dielectric material [2, 51]. Mismatches in the refractive index between layers of a composite object result in reflections of the incident THz pulse. These reflected pulses arrive at different times and have different amplitudes. Therefore, the measured waveform consists of several isolated pulses, which contain time-of-flight
pulse arrival times and information about the different interfaces. Shown in Fig. 2.6 is a conventional T-ray reflection image of a 3.5 in. floppy disk computed using the total reflected power (top) and a time-of-flight image at a vertical slice ($y = 15$ mm) of the disk (bottom). Reflections from the front cover, back cover, diskette, and metal hub are clearly visible in the time-of-flight image. The bright spots correspond to a reflection from an interface that has a positive step in refractive index. The dark spots correspond to negative steps. This information is used to estimate the index of refraction of all the layers.

Tomography, which has been used extensively in the medical imaging community [52], now has been applied using T-rays. Tomography refers to retrieving an image of an object from transmission or reflection data collected at multiple viewing angles. Ferguson et al used T-rays in a transmission computed tomography (CT) modality that is analogous to an X-ray CT scan [53]. From measurements at several viewing angles, both absorption images and refractive index images could be obtained. Other T-ray tomography systems include synthetic aperture radar [54], time reversal imaging [55, 56], multistatic reflection imaging [57, 58], and synthetic phased-array techniques [59, 60]. These systems used reflection measurements obtained at multiple viewing angles to successfully capture high resolution THz images.

Recently, we presented another reflection imaging modality, T-ray wide aperture reflection tomography (T-ray WART) [61], which forms high-resolution images of an object’s cross-section. In T-ray WART, an object’s cross-section is illuminated with a T-ray transceiver at a set of different viewing angles, and the specular back-reflected waves are measured using a THz transceiver. This is analogous to computed ultrasonic
reflection tomography used in ultrasonic imaging community [62]. T-ray WART is the first computed tomography system for electromagnetic waves that works in reflection mode. The measured reflections correspond to parallel projections through the cross-section, and therefore, the filtered backprojection (FBP) tomographic reconstruction algorithm is applied to retrieve an edge map of the object’s cross-section.

As a test object, a 1 in. x 1 in. aluminum post with a complicated cross-section was used; a picture of this object is shown on the right side of Fig. 2.7. Because aluminum is highly reflective, a transmission geometry could not be used to image this object. Using the procedure outlined above, the image of the test object was reconstructed from the full 360° range of viewing angles. The reconstructed image, shown on the left side of Fig. 2.7, captures most of the details of the outside edges including the indentions around the border of the object, which are less than 1 mm across and less than 0.5 mm deep. However, because the THz radiation was unable to reach the inside corners or screw hole, the T-ray WART system was unable to retrieve any information about features that are obscured by the outside edges. It is possible to extend this technique into three dimensions by capturing images in successive elevations similar
to a traditional X-ray CT scan.

2.5 T-rays for Studying Scattering

The propagation of electromagnetic waves through an inhomogeneous media is a very complex process, because the waves undergo multiple scattering, which results in dramatic amplitude and phase variations of the electric field. The majority of previous studies of scattering have employed narrowband optical radiation where measurements are limited to time averaged intensity or autocorrelation quantities, which contains no information of the phase of the pulses. Purely optical measurements are insufficient for obtaining all the details on the effects of propagation through random media.

The THz-TDS is a unique tool to probe random media, because it provides access to both the intensity and phase of the radiation through direct measurement of the temporal electric field. This affords easy access to both the real and imaginary parts of the Fourier coefficients, providing a wealth of information that is not directly available at optical frequencies. Because of the broad bandwidth and linear phase of the pulses, it is possible to simultaneously study Rayleigh scattering and the short wavelength limit in a single measurement. Because of the scale invariance of Maxwell’s equations, T-ray measurements can provide insights for researchers studying scattering at optical wavelengths while using scatterers that are on order of THz wavelengths (~0.5 mm). In optical experiments, a common sample scattering medium is a suspension of latex spheres in water, for which characterization of the density and the size distribution is not straightforward. Sample preparation and characterization are not significant issues in the THz domain. T-rays can access a wealth of information concerning the complex
scattering process and probe many details of a random media that are unavailable with optical frequencies. The THz-TDS is a powerful tool to study such phenomena.
Chapter 3
Wave Scattering

3.1 Introduction

Propagation of waves through random media is a topic of considerable interest in many research communities because of the rich array of phenomena resulting from the scattering of electromagnetic waves [63, 64]. In particular, if the probability of scattering is high enough, light can localize [11]. With weaker scattering, photons will propagate diffusively [65, 66], which has generated a great deal of interest to the medical community who hopes to image the location of objects immersed in scattering media such as tissue [67]. The study of electromagnetic wave propagation through random or inhomogeneous media has been extensively studied using microwaves [5, 68] and visible light [10, 11, 69]. We have pioneered the use of THz radiation for investigating scattering [3, 70-74].

The phase, amplitude, and polarization of a wave that propagates through random media are dependent on a number of properties of the medium. First, the size of the scatterer relative to the wavelength and the shape affects the scattering anisotropy and probability. The scattered field from a single particle differs greatly from that of multiple particles. Whether or not the single scattering (Born) approximation is applicable or if multiple scattering needs to be considered is strongly dependent on the volume fraction of scatterers present. Furthermore, absorption in the scatterers will decrease the number of diffusely propagating photons. In this section, we will overview some theories of wave scattering and discuss the fundamental issues of light propagation through random
media. First, we will consider the scattering from a single particle and then the scattering from a collection of particles.

3.2 Single Scattering

3.2.1 Incident Wave on a Particle

When an electromagnetic wave encounters a particle, part of the incident radiation is scattered and some of it is absorbed by the particle itself. The scattered wave interferes with the incident wave, which causes disturbances in the background electric field. Describing the scattering and absorption of a particle present in a propagating wave is the subject of many textbooks [64, 75-77]. A summary of these descriptions is presented here.

Consider a linearly polarized monochromatic electromagnetic plane wave with amplitude of $E_0$ in units of V/m propagating through free space with electric permittivity $\varepsilon_0$, and a magnetic permeability $\mu_0$. The electric field at time $\tau$ and position $\mathbf{r}$ is given by

$$\hat{E}_{\text{inc}}(\mathbf{r}, \omega) = \hat{e}_x E_0 \exp(ik\hat{x} \cdot \mathbf{r} - i\omega\tau). \quad (3-1)$$

The wave vector $\hat{\mathbf{k}}$ points in the direction of propagation and has unit magnitude, and $k = \omega \sqrt{\mu_0 \varepsilon_0} = 2\pi / \lambda$ is the wave number, where $\omega$ is the frequency and $\lambda$ is the wavelength of the wave. The unit vector $\hat{\mathbf{e}}_x$ represents the direction of polarization.

As shown in Fig. 3.1, this wave encounters a scatterer with a spatially dependent dielectric constant given by

$$\varepsilon_s(\mathbf{r}) = \varepsilon_s'(\mathbf{r}) + i\varepsilon_s''(\mathbf{r}). \quad (3-2)$$
Figure 3.1: An incident plane wave $\mathbf{E}_i(\mathbf{r})$ is scattered by a single particle with a complex dielectric constant $\varepsilon_s(\mathbf{r})$ producing a spherical wave $\mathbf{E}_s(\mathbf{r})$ propagating in the direction $\hat{\mathbf{0}}$.

The scatterer’s dielectric constant $\varepsilon_s(\mathbf{r})$ is generally a complex quantity. The refractive index of the particle is related to the complex dielectric constant by $n_s(\mathbf{r}) = \sqrt{\varepsilon_s(\mathbf{r})}$, where $n_s(\mathbf{r})$ is also a complex quantity. The real part of the refractive index describes the speed in which a wave propagates through the material and the imaginary part denotes the absorption. After the incident wave encounters the particle, a portion of the radiation is absorbed and a portion is scattered. In the far field ($R > D^2/\lambda$ where $D$ is a dimension of the particle such as a diameter), the resulting scattered radiation comes in the form of a spherical wave. However, in the near field of the particle, the scattered field is very complicated because it consists of complicated interference effects from scattering off of different areas of the particle. At large distances, these individual interference effects are washed out and can be ignored. The scattered spherical wave, $\mathbf{E}_s(\mathbf{r}, \omega)$, is defined as
\[ \hat{E}_s(\hat{r}, \omega) = E_0 \hat{f}(\hat{0}, \hat{i})(\exp(ikR - i\omega \tau) / R), \] 

where \( \hat{f}(\hat{0}, \hat{i}) \) (units of m) is the scattering amplitude function, which represents the amplitude, phase, and polarization of the scattered electric field propagating in the direction of \( \hat{0} \) from a particle that is illuminated by an incident plane wave propagating in a direction \( \hat{i} \). The electric field at a distance \( R \) in the \( \hat{0} \) direction consists of contributions from the incident plane wave \( \hat{E}_i(\hat{r}, \omega) \) and the scattered spherical wave \( \hat{E}_s(\hat{r}, \omega) \).

### 3.2.2 Optical Cross-Sections

A scattering cross-section \( \sigma_s \) is defined by the total energy scattered in all directions that is equal to the amount of energy of the incident wave that falls on an area \( \sigma_s \). The scattering cross-section can be related to \( \hat{f}(\hat{0}, \hat{i}) \) by first considering the scattered power flux density vector \( \hat{I}_s(\hat{r}) = |\hat{E}_s(\hat{r}, \omega)|^2 / 2\eta_0 \hat{0} \) at a large distance \( R \) from the particle in the \( \hat{0} \) direction, where \( \eta_0 \) is the impedance of the medium. The scattered energy flux is the rate of energy \( \hat{I}_s(\hat{r}) \cdot d\hat{A} \) that passes through a fixed area \( d\hat{A} \). By integrating over the entire area in which radiation passes, the total scattered energy flux is given by

\[ \Phi_s = \int_S \hat{I}_s(\hat{r}) \cdot d\hat{A} = \int_{4\pi} |\hat{E}_s(\hat{r}, \omega)|^2 / 2\eta_0 R^2 d\sigma = \int_{4\pi} |\hat{f}(\hat{0}, \hat{i})|^2 / 2\eta_0 d\sigma. \] 

The total scattered energy flux of a spherical wave is the rate of energy that passes through an area of the sphere \( R^2 d\sigma \), integrated over \( 4\pi \) of solid angle \( d\sigma \). The energy
flux through an area $\sigma_s$ of the incident wave is computed by similar means and is given by

$$\Phi_{inc} = \sigma_s |E_0|^2 / 2\eta_0. \quad (3-5)$$

Note that plane waves have uniform intensity, and therefore, its energy flux scales linearly with the size of the area that the radiation propagates through. From the definition of the scattering cross-section, the incident flux $\Phi_{inc}$ is set equal to the scattered flux $\Phi_s$, which gives a scattering cross-section of

$$\sigma_s = \int \hat{\mathbf{f}}(\hat{\mathbf{r}}, \hat{\mathbf{i}})^2 d\omega. \quad (3-6)$$

Scattering is not the only mechanism that removes energy from the incident wave. Absorption in the particle is also an energy sink. In a similar manner, an absorption cross-section $\sigma_a$ is defined by the total energy absorbed that is equal to the energy of the incident wave that falls on an area $\sigma_a$. A general expression for the absorption cross-section is derived from Maxwell’s equations, but the derivation is beyond the scope of this dissertation. The absorption cross-section is expressed as

$$\sigma_a = \int k \varepsilon_0 \varepsilon''(\mathbf{r}) \left| \mathbf{\hat{E}}(\mathbf{r}, \omega) \right|^2 dV', \quad (3-7)$$

where $\mathbf{\hat{E}}(\mathbf{r}, \omega)$ is the field inside the particle, which is typically unknown for arbitrary shaped particles, and the integration is over the volume of the particle.

The total energy removed from the original wave by the particle that equals to the energy incident on the area $\sigma$, defines the total or extinction cross-section of the particle. By the conservation of energy, the total, scattering, and absorption cross-sections are related by
\[ \sigma_i = \sigma_s + \sigma_a. \]  

These three cross-sections have dimensions of area and are generally functions of the particle size, shape, and orientation, as well as the polarization of the incident wave. In the special case when the particle is non-absorbing, \( \sigma_i = \sigma_s. \)

The forward scattering theorem, also called the optical theorem, relates the total cross-section to the amplitude of the scattered wave in the forward direction. H. C. van de Hulst first formulated the expression of the forward scattering theorem in the domain of classical optics [78]. This theorem states that the power removed from an incident plane wave by scattering and absorption of a particle is proportional to the imaginary part of the scattering amplitude function in the forward direction. This is expressed by

\[ \sigma_i = (4\pi / k) \text{Im}[\hat{f}(\hat{i}, \hat{i})] \cdot \hat{e}_i, \]

where \( \hat{e}_i \) is a unit vector in the direction of polarization of the incident wave.

### 3.2.3 Rayleigh Scattering

For particles with arbitrary shapes and sizes, solving for the scattering amplitude function and three cross-sections requires a formal solution of Maxwell's equations with appropriate boundary conditions. However, for particles that are either much smaller or much larger than the wavelength, good approximate solutions exist. In the case that the particle is much smaller than the wavelength, a simplification is made in the formalism by assuming that the particle is placed in a homogeneous field. This type of scattering is known as Rayleigh scattering. For larger particles, a ray optics approach is used, which follows a bundle of narrow rays as they reflect, refract, and diffract through the particle.
Figure 3.2: Rayleigh scattering. When the particle is small compared to the wavelength, it radiates similar to an oscillating dipole. The scattered field $\mathbf{E}_s(\mathbf{r}, \omega)$ at an angle $\theta$ from the direction of the dipole, propagates in the direction of the radius vector $\mathbf{r}$.

Rayleigh scattering simplifies the problem by noting that the electric field of the incident plane wave does not vary much within the particle when it is small compared to the wavelength and therefore is assumed to be constant. Assuming isotropic polarizability, the applied field induces a dipole moment

$$\mathbf{p} = \frac{\alpha \mathbf{E}_i(\mathbf{r}, \omega)}{4\pi\varepsilon_0},$$

where the dipole moment $\mathbf{p}$ points in the direction as $\mathbf{E}_i(\mathbf{r}, \omega)$ with units of $\text{C/m}^2$ and $\alpha$ is the polarizability of the particle in units of $\text{m}^3$. The electric field causes the charges in the dielectric particle to oscillate at a frequency of $\omega$. A scattered field is produced when the oscillating dipole radiates and is given by

$$\mathbf{E}_s(\mathbf{r}, \omega) = \mathbf{e}_z \frac{k^2 p \sin \theta}{4\pi\varepsilon_0 r} e^{i(kr - i\omega t)}.$$

The unit vector $\mathbf{e}_z$ points in the direction perpendicular to the radius vector, $\theta$ is the angle between the polarization and the propagation direction of the scattered wave, and $p$ is the magnitude of the polarization. By integrating the power flux of the incident and
scattered waves and using the definition of the scattering cross-section given in equation (3-6), $\sigma_s$ is found to be

$$\sigma_s = \frac{8}{3} \pi k^4 |\alpha|^2. \hspace{1cm} (3-12)$$

Rayleigh scattering predicts that the strength of scattering is inversely proportional to the fourth power of the wavelength. Lord Rayleigh used this result to explain why the sky is blue. As light propagates down from the overhead sun into the earth’s atmosphere, small molecules in the air cause the light to scatter out of the incident direction. Because blue wavelengths are shorter than red, the blue portion of the spectrum scatters more and thus will appear blue to an observer. On the other hand, when the sun is setting, the light propagates in the direction tangential to the earth’s surface and the molecules in the air scatter out much of the blue light from the line of sight direction, which then results in a red sky.

A more precise derivation of the scattering cross-section is made within the limits of Rayleigh scattering for the special situation of spheres. Lorentz showed that the polarizability of a sphere with radius $a$ and volume $V$ is

$$\alpha = \frac{3(n_s^2 - 1)}{4\pi (n_s^2 + 2)} V, \hspace{1cm} (3-13)$$

where $n_s$ is the refractive index of the sphere. Substituting this expression of the polarizability into equation (3-12) yields a scattering cross-section of

$$\sigma_s = \frac{8\pi k^4 a^6}{3} \frac{n_s^2 - 1}{n_s^2 + 2}^2. \hspace{1cm} (3-14)$$
3.2.4 Short Wavelength Scattering

Rayleigh scattering accurately describes scattering in the long wavelength limit by considering the field within the particle to be constant; however, in the short wavelength limit, the situation is much different. When the wavelength of the incident light is very short compared to the size of the particle, the light can be considered as a bundle of independent rays. Rays incident upon the particle are either reflected from the surface or refracted into the particle. The refracted light will emerge after propagating through the particle and possibly after multiple internal reflections. Any light that does not emerge is absorbed by the particle. The reflection, refraction, and absorption of light from the particle remove an area of the incident beam that is equal to the geometrical cross-section of the particle $\sigma_g$. In addition, these mechanisms block a portion of the wavefront, which results in diffraction. The diffraction from the hole in the wavefront produces a secondary wave that interferes with the incident wave. From an application of Huygen’s principle, this secondary wave is completely out of phase and therefore destructively interferes with the incident wave, which removes an additional cross-section $\sigma_g$ of energy. The total energy extinguished corresponds to a total cross-section of

$$\sigma_i = 2\sigma_g.$$  \hspace{1cm} (3-15)

This seeming contradiction, that a large particle removes twice the amount of the energy it intercepts, is known as the extinction paradox.

3.2.5 Mie Scattering

For more general wavelengths and particles, solving for the scattered field and cross-sections involve numerical calculations such as finite element methods and $T$-matrix methods [79]; however, in the case for spherical particles, exact solutions for the
scattered field exist and are described by Mie theory [80]. The Mie derivation involves solving Maxwell’s equations with boundary conditions and can be found in a number of textbooks [76, 77].

Here, the results of Mie’s solution for the scattering of a homogeneous sphere are presented. The frame of reference is chosen such that \( \hat{z} \) is the propagation direction, \( \hat{x} \) is the polarization direction of the incident wave, and \( \hat{y} \) is orthogonal to \( \hat{z} \) and \( \hat{x} \). With these coordinates, the linearly polarized plane wave defined in equation (3-1) becomes

\[
\hat{E}_{inc}(\hat{r}, \omega) = \hat{x}_m E_0 \exp(ikz - i\omega \tau).
\]

The scattered field is related to the incident field by

\[
E_{s,\perp}(\hat{r}, \omega) = E_0 S_1(\theta)(i \exp(ikR - i\omega \tau) / kR) \sin \varphi,
\]

\[
E_{s,\parallel}(\hat{r}, \omega) = E_0 S_2(\theta)(i \exp(ikR - i\omega \tau) / kR) \cos \varphi,
\]

where \( \theta \) is the angle between the incident wave and the scattered wave. The subscripts \( \parallel \) and \( \perp \) denote the polarizations that are parallel and perpendicular to the plane of scattering defined by the incident and scattered wave. The angle \( \varphi \) is between the polarization direction of the incident wave \( \hat{x} \) and the plane of scattering. The complex amplitude functions \( S_1(\theta) \) and \( S_2(\theta) \) relates the field of the incident wave to the scattered field in the parallel and perpendicular polarizations. Spheres have the special property that they do not rotate the polarization, which is generally not true for arbitrarily shaped particles. These amplitude functions are related to the scattering amplitude function defined in equation (3-3) as follows:

\[
\hat{f}(\hat{0}, \hat{i}) = \frac{i}{k} S_1(\theta) \sin \varphi \cdot \hat{e}_\perp + \frac{i}{k} S_2(\theta) \sin \varphi \cdot \hat{e}_\parallel,
\]

(3-18)
where $\hat{e}_\parallel$ and $\hat{e}_\perp$ are vectors pointing in the directions parallel and perpendicular to the scattering plane respectively and perpendicular to the propagation direction of the scattered wave.

Mie’s expressions for the scattering amplitude function are given by

$$S_1(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[ a_n \pi_n(\cos \theta) + b_n \tau_n(\cos \theta) \right],$$

(3-19)

$$S_2(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[ b_n \pi_n(\cos \theta) + a_n \tau_n(\cos \theta) \right],$$

where

$$\pi_n(\cos \theta) = \frac{1}{\sin \theta} P_n^i(\cos \theta),$$

(3-20)

$$\tau_n(\cos \theta) = \frac{d}{d\theta} P_n^i(\cos \theta).$$

and $P_n^i(\cos \theta)$ is a Legendre polynomial. The complex coefficients $a_n$ and $b_n$ are

$$a_n = \frac{\psi_n''(n,ka)\psi_n(ka) - n_n \psi_n(n,ka)\psi_n'(ka)}{\psi_n'(n,ka)\zeta_n(ka) - n_n \psi_n(n,ka)\zeta_n'(ka)},$$

(3-21)

$$b_n = \frac{n_n \psi_n'(n,ka)\psi_n(ka) - \psi_n(n,ka)\psi_n'(ka)}{n_n \psi_n'(n,ka)\zeta_n(ka) - \psi_n(n,ka)\zeta_n'(ka)}.$$

The functions $\psi_n(x)$ and $\zeta_n(x)$ are the Riccati-Bessel functions and are related to the spherical Bessel functions by

$$\psi_n(x) = (\pi x / 2)^{1/2} J_{n+1/2}(x),$$

(3-22)

$$\zeta_n(x) = (\pi x / 2)^{1/2} H^{(2)}_{n+1/2}(x),$$
where $J_{n+1/2}(x)$ is the Bessel function of the first kind and $H_{n+1/2}^{(2)}(x)$ is the Bessel function of the second kind.

At $\theta = 0$ both amplitude functions, $S_1(\theta)$ and $S_2(\theta)$ have the same value as a consequence of the symmetry of the sphere:

$$S(0) = \frac{1}{2} \sum_{n=1}^{\infty} (2n+1)(a_n + b_n), \quad (3-23)$$

where $\pi_n(1) = \tau_n(1) = \frac{1}{4} n(n+1)$. Using the equations above, the total, scattering, and absorption cross-sections can be calculated. The forward scattering theorem relates the scattering amplitude functions in (3-19) to the total cross-section by

$$\sigma_t = \frac{4\pi}{k^2} \operatorname{Re}\{S(\theta = 0)\} = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \operatorname{Re}\{(a_n + b_n)\}. \quad (3-24)$$

Deriving an expression for the scattering cross-section as defined in (3-6) requires an integral over $\theta$ that contains a doubly infinite series. Fortunately, most of the terms cancel out and results in this expression for the scattering cross-section:

$$\sigma_s = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1)(|a_n|^2 + |b_n|^2). \quad (3-25)$$

Finally, from the relationship between the three cross-sections in (3-8), the absorption cross-section follows from subtraction:

$$\sigma_a = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \operatorname{Re}\{(a_n + b_n)\} - \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1)(|a_n|^2 + |b_n|^2). \quad (3-26)$$

Fig. 3.3(a) shows the Mie solution of the total cross-section normalized by the geometrical cross-section plotted over the spectral range of 50 GHz to 10 THz for a non-absorbing sphere. The sphere has a radius of 0.397 mm and an index of refraction $n_e$ of 1.4333. This is representative of the type of scatterers used in the experiments that are
described in subsequent chapters. The Mie solution is very complex and is comprised of many resonances. The strongest scattering occurs when the wavelength is on the order of the particle size. For a sphere with a diameter of 0.794 mm, this corresponds to frequencies in the neighborhood of 0.4-0.7 THz. The Mie solution in Fig. 3.3(a) spans the frequency range between Rayleigh scattering and the short wavelength limit. The Rayleigh approximation calculated from (3-14) appears to agree well with the Mie solution at frequencies up to 0.25 THz. At higher frequencies, the Mie solution converges toward 2, which is explained by the extinction paradox.

Fig. 3.3 (b) shows the scattering amplitude function of the sphere at 1 THz when the incident wave is polarized in the direction parallel with the scattering plane. The majority of the radiation is scattered in the forward direction. When the size of the particle is comparable to the wavelength, forward scattering dominates.

In 1998, Cheville et al used THz time-resolved experiments to investigate the scattering from dielectric spheres and compare it to Mie theory [81]. The frequency domain solution for the scattered field calculated by Mie theory does not provide insights to the physical mechanisms that contribute to the multiple resonances of the solution. They illuminated a dielectric sphere with single-cycle THz pulses and measured the backscattered radiation. Using the arrival times of the pulses, they isolated the mechanisms responsible for glory scattering: specular, surface wave, and back axial reflections. They directly compared the isolated surface wave to Mie theory with excellent agreement.
Figure 3.3: The Mie solution for the (a) total cross-section as a function of frequency normalized by the geometrical cross-section and the scattering amplitude function as a function of scattering angle at (b) 1 THz and at (c) 0.1 THz. The non-absorbing sphere has a 0.397 mm radius and an index of refraction 1.4333. The dashed lines in (a) represent the approximate Rayleigh and short wavelength limit solutions.

3.3 Overview of Multiple Scattering

Several theories exist, both exact and approximate solutions, for the scattering of a single particle. However, the presence of many particles results in multiple scattering and further complicates the solution for the scattered field. The scattering of electromagnetic waves can lead to a variety of interesting physics [63, 64] including photon localization [11] and diffusion [65, 66]. Multiple theories exist that describe the
propagation of an electromagnetic wave through an inhomogenous material, depending on the shape, size, distribution, and density of the particles [64, 75, 82].

Consider an incoming wave that propagates through a slab of a homogeneous material containing randomly distributed dielectric particles as shown in Fig. 3.4. The index of refraction of the particles differs from that of the background material. The $x$ and $y$ dimensions are considered infinite, and the $z$ dimension has a finite thickness $L$. The slab thickness dramatically influences the wave propagation. By increasing $L$, three regimes of scattering are encountered: the ballistic regime, the diffusion regime, and the absorption regime [4].

Fig. 3.5 shows a heuristic of the transmission coefficient as a function of the slab thickness along with the three scattering regimes. In the first regime, the transmission coefficient initially decays exponentially as a result of photons being scattered out of the
direction of the incident light. The decay rate is the scattering mean free path $\lambda_{sc}$, which is the average distance between scattering events. This is denoted as the ballistic regime. After further increasing the slab thickness, the diffusion regime is encountered, which has a decay rate of $1/(L + L_0)$ that is much slower than an exponential decay. As photons scatter multiple times, a few of them will reenter the direction of the incident wave, thereby decreasing the decay rate of the transmission coefficient. The last region is the absorption regime, where the effects of absorption over long path lengths dominate resulting in an exponential decay of the transmission coefficient. These three regimes characterize the transport of light propagating through an inhomogeneous slab.
In the case of spherical scatterers, ballistic and diffuse photons not only differ in their transmission characteristics, but also in their degree of polarization. Ballistic photons scatter only in the forward direction. Recall that the scattering off of a single sphere does not change the polarization of the incident wave. Rotating the polarization requires more than one non-forward scattering event. Therefore, ballistic photons maintain their polarization. Diffuse photons have encountered many scattering events,
which randomizes their polarization and thus are unpolarized. Between those two extremes exists a third type of photons, the snake [83], which have encountered more than one scattering event, but still recalls some memory of its initial polarization. Fig. 3.6 illustrates these three photons and their propagation paths.

At this point, only disordered materials have been considered. This begs the question how does a wave propagate in a highly ordered material? Photonic crystals are such structures, which are periodic dielectric materials along one, two, or three dimensions [84]. By controlling the periodicity and materials, they can be tuned to prevent certain frequencies from propagating, which results in bandgaps in the transmitted light. Recently, work has been done to study these structures at THz frequencies. As an example, Jian et al. constructed a 2-D crystal out of a thin slab of silicon with a hexagonal array of small circular holes patterned onto it [85]. They then placed defects within the crystal by filling several holes with silicon powder. This resulted in enhanced transmission within the bandgap region. Ordered materials are of much interest to the THz community, because of the demand for components that can manipulate THz radiation.

3.4 Ballistic Light

3.4.1 Complex Scattering Index of Refraction

Ballistic photons maintain their polarization, and their intensity decays exponentially as path length increases. They also provide information about the properties of the scattering medium. Consider the outgoing wave \( \hat{E}_r(\mathbf{r}, \omega) \) resulting from the incoming wave \( \hat{E}_i(\mathbf{r}, \omega) \) after encountering a slab of a non-absorbing inhomogeneous
material as pictured in Fig. 3.4. It is assumed that $L$ lies within the ballistic scattering regime. The outgoing wave $\hat{E}_s(\hat{r},\omega)$ propagates in the incident direction and is related to the incoming wave by

$$\hat{E}_s(\hat{r},\omega) = \hat{E}_i(\hat{r},\omega) \exp(-j\omega n_{sc} L/c),$$

(3-27)

where $n_{sc} = n_r - jn_i$ is the effective complex scattering index of refraction of the inhomogeneous slab. The imaginary part of the scattering refractive index $n_i$ describes the attenuation of the incident wave due to scattering and absorption in the medium.

A scattering mean free path $\lambda_{sc}$ is defined as the average distance between scattering events. This length corresponds to the distance that the incident wave has to travel to obtain a decay of intensity to $1/e$ of its initial value and is related to $n_i$ by

$$\lambda_{sc} = \frac{c}{2 n_i \omega L}.$$  

(3-28)

When the scatterers fill a small volume fraction, the scattering mean free path $\lambda_{sc}$ is also related to the scattering cross-section by

$$\lambda_{sc} = \frac{1}{n_0 \sigma_s},$$  

(3-29)

where $n_0$ is the number density of scatterers and $\sigma_s$ is the scattering cross-section as defined by equation (3-6). If the wavelength is on the order of the size of the scatterer, forward scattering predominates. The critical parameter is no longer the mean free path $\lambda_{sc}$, but the transport mean free path $\lambda_t = \lambda_{sc} / (1 - \langle \cos \theta \rangle)$, where $\langle \cos \theta \rangle$ is the average cosine of the angle of scattering and is given by

$$\langle \cos \theta \rangle = \frac{1}{k^2 \sigma_s / 4\pi} \int \int \hat{f}(\hat{0}, \hat{i}) \left| (\hat{0} \cdot \hat{i}) \right| d\omega,$$

(3-30)
where the integration is over $4\pi$ of solid angle. The transport mean free path is the distance traveled to lose memory of the forward direction. For spherical particles, Mie theory can be applied to calculate $\lambda_{sc}$ from $\sigma_s$ as in equation (3-29); however, this is only applicable for situations where the volume fraction is on the order of 1% or less.

The real part of the scattering index of refraction $n_r$ is equivalent to the effective index of refraction of the inhomogeneous slab $n_{avg}$, which is the fraction of $c$ that light travels through the medium. There are several methods to estimate $n_{avg}$, one of which is volume weighted averaging given by

$$n_{avg} = \text{Re}\{\sqrt{\varepsilon_s f + \varepsilon(1-f)}\}$$  \hspace{1cm} (3-31)

where $\varepsilon_s$ is the permittivity of the sphere, $\varepsilon$ is the background permittivity, and $f = n_0 V$ is the volume fraction of the spheres occupying the volume $V$ of the slab. Another formula to estimate the effective index of refraction is called the Maxwell-Garnett mixing formula [86], which predicts that the effective permittivity is

$$\varepsilon_{eff} = \varepsilon \frac{1 + 2 fy}{1 - fy}$$  \hspace{1cm} (3-32)

where

$$y = \frac{\varepsilon_s - \varepsilon}{\varepsilon_s + 2\varepsilon}.$$  \hspace{1cm} (3-33)

The average index refraction is simply $n_{avg} = \text{Re}\{\sqrt{\varepsilon_{eff}}\}$. This equation is valid at small volume fractions and assumes that the particle is much smaller than the wavelength.

3.4.2 Quasi-Crystalline Approximation (QCA)

Numerous theoretical models have been developed to compute these effective propagation parameters for waves in a dense collection of scatterers at a greater level of
accuracy than these simple mixing equations. These models are generally based on the multiple scattering equations, a system of formulas relating the field incident on a particular scatterer to the fields arriving from all other scatterers. Tsang et al provides a simple explanation of the multiple scattering equations [82]. Consider a situation where there are \( N \) randomly distributed scatterers contained in a volume \( V \) similar to Fig. 3.4. An incident electric field \( \hat{E}_{\text{inc}} \) is applied to the volume. The resulting macroscopic electric field \( \hat{E} \) encompasses contributions from each of the scattered fields from the particles \( \hat{E}_{ij} \) and the incident field \( \hat{E}_{\text{inc}} \) given by:

\[
\hat{E} = \hat{E}_{\text{inc}} + \sum_{j=1}^{N} \hat{E}_{ij}.
\]  

(3-34)

For a single particle \( j \), there is an exciting field \( \hat{E}^E_j \) that induces a field of \( \hat{E}_{ij} \) in the absence of all other particles. \( \hat{E}^E_j \) and \( \hat{E}_{ij} \) are related to each other by a transition operator \( T_j \), which is assumed to be known and is a function of the shape, size, and location of the particles,

\[
\hat{E}_{ij} = T_j \hat{E}^E_j.
\]  

(3-35)

The exciting field \( \hat{E}^E_j \) contains contributions from all of the particles. \( \hat{E}^E_j \) can also be expressed in terms of \( \hat{E} \) and \( \hat{E}_{ij} \) by

\[
\hat{E}^E_j = \hat{E} - \hat{E}_{ij}.
\]  

(3-36)

Combining equations (3-34), (3-35), and (3-36), the exciting electric field of a single particle is expressed as
\[ \hat{E}^E_j = \hat{E}_{\text{inc}} + \sum_{l=1 \atop l \neq j}^{N} T_l \hat{E}^E_l. \] (3-37)

The exciting electric field can now be calculated, because equation (3-37) consists of \( N \) equations with \( N \) unknowns. The scattered fields can also be calculated by equation (3-35). A more rigorous derivation of this system of equations can be derived using Green’s function or T-Matrix formalism.

An assumption was made that the locations and orientations of all the scatterers (and therefore \( T_j \) ) was known. However, this is rarely the case. The propagation constants can be calculated by averaging the equations of (3-37) over the entire number of different particle configurations. When the multiple scattering equations are averaged over all configurations of the scatterers, a hierarchy of solutions results in expressing the average Green’s function in terms of the conditional averaged Green’s function with the location of one particle fixed. The conditional average of the Green’s function with one particle location fixed is then related to the conditional averaged Green’s function with the locations of two particles fixed. Generally, the conditional average of the Green’s function with \( n \) particles fixed is related to that with \( n+1 \) particles fixed. These expressions can be truncated to only take into consideration one or two particle correlations. For higher volume fractions, the positions of the particles become more and more correlated; this requires higher order solutions that consist of more particle correlations and are much more difficult to solve.

The effective field approximation (EFA) or Foldy’s approximation truncates this hierarchy at the lowest order to obtain a simple solution for the propagation constants [87]. This is equivalent to neglecting all correlations among the scatterer locations. The
EFA is usually only valid for small particle densities ~ 1%. For higher volume fractions, below ~40%, the quasi-crystalline approximation (QCA) is a more appropriate formalism. It consists of a second-order truncation of the hierarchy, so two-particle correlations are included. It permits the computation of the effective propagation constant of the wave, given the volume fraction, the complex dielectric of the spheres, the size parameter, and the two-particle distribution function [88]. For spherical scatterers, the Percus-Yevick pair distribution function provides an adequate description of the positional correlation [89]. The higher density regime (above 40%) has not been thoroughly investigated yet. A solution would have to include higher orders of scattering and a 3 or more particle distribution function, which would severely increase the complexity. Currently, the only theoretical methods for studying high densities of scatterers are numerical, such as Monte Carlo methods. In fact, there is also a paucity of experimental work at fractional volumes over 40%.

3.4.3 Terahertz Measurements of Ballistic Photons

In a previous work, we experimentally tested the limits of validity of the quasi-crystalline approximation by probing random media with single-cycle THz pulses [73, 74]. We constructed a model random medium using commercially available polytetrafluoroethylene (PTFE, teflon) spheres, with a diameter of 0.794 mm. Teflon is an excellent material for these studies since its absorption coefficient is quite low at THz frequencies. Also, the refractive index of teflon, $n_{\text{PTFE}} = 1.4330$, is nearly independent of frequency throughout the spectral range of the measurements [90]. The spheres were poured in a series of teflon sample cells with increasing path lengths ranging from 1.19 mm to 20.64 mm. The spheres filled a volume fraction of $\phi = 0.56 \pm 0.04$, which is much
Figure 3.7: The open squares show the measured reduced scattering coefficient as a function of wavelength. The error bars show typical uncertainties due to variability in repeated measurements. The solid curves are the predictions of the (red) quasi-crystalline approximation and (green) effective field approximation.

greater than the 40% limit of the QCA. The electric field of the transmitted pulse was measured for each path length.

Using equations (3-27) and (3-28), the reduced scattering coefficient $1/\lambda_r$ was extracted from the measurements. Fig. 3.7 shows a comparison of the experimental and theoretical reduced scattering coefficient as a function of wavelength using the quasi-crystalline approximation. These data show a variation in $1/\lambda_r$ by roughly a factor of 40 in magnitude. Typical error bars are shown, indicating the uncertainty associated with repeated measurements. The solid line shows the predictions of the QCA, calculated using parameters corresponding to those of the experiment. Clearly, the theory
Figure 3.8: The open circles show the measured group velocity, obtained by differentiating the measured effective refractive index. The QCA prediction (solid curve) matches the data quite well.

underestimates the strength of the scattering, a result of neglecting higher-order correlations. Even so, the correspondence between the resonant features in the two curves is quite good.

Because these data were acquired with a broadband spectrometer, it was also possible to determine the effective group velocity. The group velocity of the wave depends not only on the effective index but also on its derivative, according to

\[ v_G = c \left( n_{\text{eff}} + \omega \frac{dn_{\text{eff}}}{d\omega} \right)^{-1}. \]  

(3-38)

This is the velocity with which a wave packet moves through the medium. The group velocity was determined experimentally by numerically differentiating the measured effective index. This is shown in Fig. 3.8, along with the QCA result (solid line). The agreement is much improved in comparison with Fig. 3.7, demonstrating that the QCA
can be used to accurately compute group velocities even when the volume fraction greatly exceeds 40%.

These measurements involved volume fractions exceeding 50%, which is beyond the limits of validity assumed for the QCA. In fact, the very high density regime (above 40%) has not been thoroughly investigated, so the validity of this second-order theory is not well established in this regime. These results demonstrated that the QCA provides some indication of the scattering coefficient, and in the case of the group velocity, the agreement with experiment was very good. This work showed that, while not perfect, the QCA is still useful even far beyond its expected limits of the validity. These results indicated the diminishing importance of higher-order correlations, at least in the case of moderate dielectric contrast studied in this work.

3.5 Diffuse Light

3.5.1 Photon Time-of-Flight Distribution

When the slab thickness $L$ in Fig. 3.4 is much larger than the transport mean free path $\lambda_r$, the photons will propagate diffusively. In this case, the photons lose memory of the incident direction and their polarization is completely randomized. Because diffuse photons travel through long paths lengths, they contain information about the properties of the random medium. This has generated interest in the medical community to use diffuse waves for imaging [91, 92] and spectroscopy [93, 94] of tissue, because near infrared light does not absorb through the skin, but scatters significantly.

Patterson et al. developed a model to describe the transport of photons through a slab of tissue [95]. However, this model is valid for any random medium in which $L \gg \lambda_r$. The assumption made in this model is that light propagation in a diffusive
random medium obeys the diffusion equation

\[ \frac{1}{c} \frac{\partial}{\partial t} \phi(\hat{r},t) - D \nabla^2 \phi(\hat{r},t) + \mu_a \phi(\hat{r},t) = S(\hat{r},t) \]  

(3-39)

where \( \phi(\hat{r},t) \) is the diffuse photon fluence rate, which is proportional to the intensity averaged over all the configurations of scatterers. The factor on the second term is the diffusion coefficient \( D \):

\[ D = 3[\mu_a + (1 - g)\mu_s]^{-1}, \]  

(3-40)

where \( \mu_a \) is the absorption coefficient, \( \mu_s = 1/\lambda_{sc} \) is the scattering coefficient, and \( g = \langle \cos \theta \rangle \) is the average cosine defined in equation (3-30). The term \( S(\hat{r},t) \) on the left hand side of (3-39) is the photon source.

In an infinite medium with a short input pulse \( S(\hat{r},t) = \delta(0,0) \), the solution to equation (3-41) is

\[ \phi(\hat{r},t) = c(4\pi Dct)^{-3/2} \exp\left( -\frac{r^2}{4Dct} - \mu_a ct \right). \]  

(3-41)

They apply this solution for an infinite medium to solve for the slab by making two assumptions: all the incident photons are scattered after propagating a distance

\[ z_0 = [(1 - g)\mu_s]^{-1} \]  

(3-42)

into the medium, and the diffuse fluence rate at the boundaries is 0. This boundary condition is met by adding a series of positive and negative photon sources at

\[ z = 2sL \pm z_0, \]  

(3-43)

where \( s = \{ \ldots, -1, 0, 1, \ldots \} \) and \( z = 0 \) at the incident face of the slab. By retaining two pairs of photon sources in their solution and integrating the fluence rate over the outgoing plane of the slab, they calculate the transmittance to be
Figure 3.9: The photon time-of-flight distribution as calculated by equation (3-44). The parameters are as follows: \( g = 0.70, \mu_s = 0.275 \text{ mm}^{-1}, \mu_a = 0.0 \text{ mm}^{-1}, L = 40.0 \text{ mm} \).

\[
T(t) = (4\pi Dc)^{-1/2} t^{-3/2} \exp(-\mu_a ct) \\
\times \left\{ (L - z_0) \exp \left[ -\frac{(L - z_0)^2}{4Dct} \right] - (L + z_0) \exp \left[ -\frac{(L + z_0)^2}{4Dct} \right] \right\} \\
+ (3L - z_0) \exp \left[ -\frac{(3L - z_0)^2}{4Dct} \right] - (3L + z_0) \exp \left[ -\frac{(3L + z_0)^2}{4Dct} \right].
\]  

Equation (3-44) describes the photon time-of-flight distribution of a short pulse that propagates through a semi-infinite slab of inhomogeneous material.

A typical solution of equation (3-44) is shown in Fig. 3.9. It displays the theoretical prediction for the photon time-of-flight distribution using the model. The chosen parameters (\( g = 0.70, \mu_s = 0.275 \text{ mm}^{-1}, \mu_a = 0.0 \text{ mm}^{-1}, L = 40.0 \text{ mm} \)) are representative of the samples used in the experiments performed in this thesis. The curve
has a fast rise time at early times as a result of photons scattering in the forward direction. At later times, there is a much slower decay due to contributions from longer path lengths. The random medium temporally spreads a short light pulse as it propagates. The time-of-flight distribution contains information about the properties of the scatterers, such as the diffusion coefficient \( D \) and anisotropy \( g \), which can be used to characterize various media.

### 3.5.2 Statistic of the Scattered Field

Diffuse photons have traversed a number of different paths with varying lengths before exiting the medium. Multiple scattering events within the inhomogeneous medium randomize the direction and phase of the incident wave. Therefore, it is appropriate to study the statistical characteristics of transmitted waves through random media. For narrowband or monochromatic light, the random phasor model accurately represents the statistics of the emitted electric field [96].

As shown in Fig. 3.10, the outgoing wave \( E_o(\hat{r}, \omega) \) from an inhomogeneous slab is the sum of a large number \( N \) of random phasors from contributions of waves that propagate through different paths of the medium:

\[
E_o(\hat{r}, \omega) = \exp(i \omega t) \frac{1}{\sqrt{N}} \sum_{k}^{N} e_k \exp(i \phi_k), \tag{3-45}
\]

where \( e_k \) and \( \phi_k \) are the respective amplitude and phase of a contribution from one path. In this analysis, the polarization is ignored, but can easily be incorporated by analyzing each polarization individually. For an ensemble of random configuration of scatterers, it is assumed that the random variables \( e_k \) have identical probability distributions and the phases \( \phi_k \) are uniformly distributed over \( (-\pi, \pi) \). For slabs much larger than the
Figure 3.10: Different paths photons travel through a random configuration of scatterers.

wavelength, these assumptions are valid, because the randomization does not favor any particular path. Dropping the $\exp(i\omega t)$ term, the real and imaginary parts are defined as

\[
    r = \text{Re}\{E_o(\hat{r})\} = \frac{1}{\sqrt{N}} \sum_k^N \bar{e}_k \cos(\phi_k) \\
    i = \text{Im}\{E_o(\hat{r})\} = \frac{1}{\sqrt{N}} \sum_k^N \bar{e}_k \sin(\phi_k).
\]  

(3-46)

Because $r$ and $i$ are the sums of many independent random variables, the central limit theorem predicts that they are Gaussian distributed.

The means of $r$ and $i$ are calculated by

\[
    \bar{r} = \frac{1}{\sqrt{N}} \sum_k^N \bar{e}_k \cos(\phi_k) \\
    \bar{i} = \frac{1}{\sqrt{N}} \sum_k^N \bar{e}_k \sin(\phi_k).
\]  

(3-47)

Because $\phi_k$ is uniformly distributed over $(-\pi, \pi)$, $\cos(\bar{\phi}_k) = \sin(\bar{\phi}_k) = 0$ and thus $\bar{r} = \bar{i} = 0$. Also, the variances are computed by
\[
\overline{r^2} = \frac{1}{N} \sum_{k}^{N} \sum_{n}^{N} \overline{\epsilon_k \epsilon_n \cos(\phi_k) \cos(\phi_n)} \\
\overline{i^2} = \frac{1}{N} \sum_{k}^{N} \sum_{n}^{N} \overline{\epsilon_k \epsilon_n \sin(\phi_k) \sin(\phi_n)}.
\]

Again, because of the uniform distribution of the phases and the independency of the amplitudes and phases, the variances of the real and imaginary parts of the field is
\[
\sigma^2 = \overline{r^2} = \overline{i^2} = \frac{\langle I(\hat{r}) \rangle}{2},
\]
where \( \langle I(\hat{r}) \rangle = \langle E^*(\hat{r})E(\hat{r}) \rangle \) is the configurationally averaged intensity. The correlation between \( r \) and \( i \) is
\[
\overline{ri} = \frac{1}{N} \sum_{k}^{N} \sum_{n}^{N} \overline{\epsilon_k \epsilon_n \cos(\phi_k) \sin(\phi_n)} = 0.
\]

The real and imaginary parts are therefore uncorrelated. From the central limit theorem, the probability density function (pdf) of \( r \) and \( i \) is
\[
P(r, i) = \frac{1}{2\pi\sigma^2} \exp \left[ -\frac{r^2 + i^2}{2\sigma^2} \right].
\]

Furthermore, it can be shown that the intensity \( I = r^2 + i^2 \) obeys a Rayleigh distribution [96]:
\[
P(I) = \begin{cases} 
\frac{I}{\sigma^2} \exp \left( -\frac{I^2}{2\sigma^2} \right) & I > 0 \\
0 & \text{otherwise.}
\end{cases}
\]

Chabanov and Genack measured the field distributions of the real and imaginary parts of narrowband microwaves propagating through a collection of polystyrene spheres in a waveguide [6]. They investigated the distributions in the crossover regime from ballistic to diffusive wave propagation. In purely diffuse waves, the distribution is zero-
mean jointly Gaussian. However, in the ballistic regime, the distribution is no longer zero-mean because forward propagating waves begin to dominate in the phasor sum. This results in a breakdown of the independency assumption of the amplitudes $e_k$. In their work, they observed as the medium thickness approached the ballistic regime, the distribution became more elliptical and the mean deviated further from zero.

In a later work, Genack et al studied the statistics of the phase derivative, which is proportional to the delay time [5, 8]. They derived an expression for the distribution of the phase derivative $\phi' \equiv d\phi / d\omega$

$$P\left(\phi' \equiv \langle \phi' \rangle \right) = \frac{Q}{2 \left[ Q + (\phi' - 1)^2 \right]^{3/2}},$$  \hspace{1cm} (3-53)

where $Q$ is a parameter related to the absorption length and the sample geometry. This expression was another result of the random phasor model. They compared this distribution to experimental measurements of the phase derivative and obtained very good agreement.

Although these theories and experiments studied extensively the statistics of monochromatic or narrowband fields in random media, the treatment of the statistics of broadband fields remains largely unaddressed. Recently, we investigated the statistics of single-cycle THz pulses propagating in a collection of scatterers [72]. We discovered that in fact the distribution for broadband radiation is non-Gaussian. This led to the development to a new theory that accurately represents the broadband case. The broadband distribution for the real and imaginary parts was found to be a superposition of zero-mean Gaussian distributions with frequency-dependent variances. This work is further described in Chapter 4.
3.5.3 Angular Dependence

We previously demonstrated the application of diffuse waves to characterize individual scattering events using THz time-domain spectroscopy [3]. We directly measured the diffusing electric field in a random medium, with high temporal resolution. This measurement permitted us to obtain information about the locations of individual scattering events experienced by portions of the diffusing field. This information is typically not experimentally accessible because the measured field generally consists of a superposition of many random phasors, corresponding to waves that have traversed many different possible paths [96]. However, by computing the temporal correlations between waves measured at different spatial locations, we were able to highlight the portions that originate from a single scattering event. This represented a completely new method for characterizing a random medium, since the concept of locating individual scattering events within a diffusing wave has not previously been considered. Because it has recently become possible to measure the amplitude and phase of ultrashort optical pulses [97], our results could in principle be extended to biologically relevant wavelengths in the near infrared. Thus, these measurements open up new possibilities for imaging in biological media.

To simulate a random medium, we used a large number of teflon spheres, with diameter $0.794 \pm 0.025$ mm. The spheres were poured into a cylindrical Teflon cell with internal radius of 2.3 cm. The volume fraction of the spheres in the sample cell was $0.56 \pm 0.04$. The THz transmitter was fixed, but the fiber-coupled detector was mounted at the end of a rail, which pivoted around the central axis of the cylindrical sample cell, as shown in Fig. 3.11. For each fixed configuration of the random medium, we measured
Figure 3.11: Schematic of the angular dependence scattering experimental setup [3], showing the fiber-coupled THz detector in two different positions (labeled $\theta_1$ and $\theta_2$). A spherical wave produced by a hypothetical scattering event, centered to the right of the axes of both of the displayed detector locations, is also shown. This partial spherical wave produces a correlated signal at the two detectors, with a temporal shift resulting from the geometrical path length difference. The detector at $\theta_2$ receives the signal earlier.

waveforms of the electric field at angles of $\theta = 65^\circ$, $66^\circ$, $75^\circ$ and $\theta = 105^\circ$, $106^\circ$, $115^\circ$, corresponding to forward and backward scattering, respectively.

A surprising result was found by computing the correlation function between portions of the measured time-domain waveforms, rather than using the entire waveform. By choosing a temporal window, one can highlight correlations that occur at particular time delays. For example, the later parts of a particular pair of waveforms show smaller correlations than the earlier parts, since these late-arriving parts have scattered more times [98]. In addition, partial waves may arrive at each detector location with a different delay (see Fig. 3.11); so correlated signals may appear at nonzero values of the correlation offset. To formalize the computational procedure, we defined a correlation function with a variable-delay window:
Figure 3.12: Correlation function $C_{\theta\theta}(\tau, T)$ plotted versus the correlation offset $\tau$ and the delay of the temporal window [3]. These data represent an ensemble average of all pairs of measured fields with the indicated angular separation for one particular realization of the random configuration of scatterers. In this example, an extended oscillatory correlation is observed at a window delay of $T \sim 730$ ps. This correlation persists even for large angular separations. The dashed lines indicated cuts through these data sets used to locate individual scattering events.

$$C_{\theta\theta}(\tau, T) = \frac{1}{C_0} \left( \int_{-\infty}^{\infty} [E_{\theta_1}(t) \cdot W_T(t)][E_{\theta_2}(t + \tau) \cdot W_T(t + \tau)] dt \right)$$  (3-54)

where $\tau$ is the correlation offset. $W_T(t)$ is a window function defined to be unity in a symmetric window about $t = T$ and zero otherwise. The window width was fixed at 50 ps, approximately equal to the inverse coherence bandwidth for the experimental geometry [64]. As usual, the angular brackets indicate an ensemble average over all waveform
pairs with the specified value of $\delta \theta$. $C_0$ is a normalization factor which ensures that $C_{\delta \theta=0}(\tau = 0, T) = 1$.

A typical result for $C_{\delta \theta}(\tau, T)$ is shown in Fig. 3.12, for three different values of $\delta \theta$. Clearly, the correlation at zero offset ($\tau = 0$) decreases with increasing window delay $T$, as a result of the increasing average number of scattering events. Also as expected, the correlation between pairs of waveforms with $\delta \theta = 8^\circ$ is nearly zero, since this is more than twice the angular speckle decay width. However, a strong oscillatory signal, indicating a correlation extending over several cycles of the field, is observed at a window delay of $T \sim 730$ ps. This extended correlation is observed at this value of $T$ for many angular separations, even those that are larger than the angular width of a typical speckle spot. This surprising feature arises when a partial wave from one particular scattering event gives rise to synchronized (though not simultaneous) signals at all detector locations. In the example shown here, only one scattering event is observed. However, for other configurations of the random medium, we observed numerous oscillatory signatures, occurring at various values of $T$.

From $C_{\delta \theta}(\tau, T)$, it was possible to determine the location of the particular scattering event that gives rise to the observed correlation. We used the evolution of the phase of the correlation to determine the direction from the detector to the scattering event. This phase evolves in a systematic way with increasing $\delta \theta$. In these data, the oscillations shifted to a larger negative correlation offset with increasing $\delta \theta$. This indicated that, for waveforms with larger angular separation, a larger (negative) temporal offset was required to cause the oscillations in one waveform to coincide with those in the other. This offset was a result of the geometrical path-length difference arising from
the tilt of the wave front. This increasing negative shift with increasing $\delta \theta$ means that the correlated portion of the wave front arrived earlier at detectors with larger values of $\theta$, and therefore the corresponding scattering event took place on the side of the detector axis closer to the sample input facet (as illustrated in Fig. 3.11). Conversely, an increasing positive shift with increasing $\delta \theta$ would be observed if the scattering event took place on the opposite side of the detector axis from the input facet. Thus, the evolution of the correlation phase is a direct indication of the direction of the scattering event relative to the detector axis.

3.5.4 Polarization Dependence

Diffuse photons not only differ in their statistics as compared to ballistic photons, but also in their polarization. Ballistic photons completely maintain their initial polarization. However, after several scattering events when there is diffusive propagation, light depolarizes and loses memory of its initial polarization. The influence to polarization from light propagation through random media is a considerable interest to the biomedical community, because polarization information can be used to image objects buried in random media such as tissue [99].

The four instantaneous Stokes parameters completely describe the polarization of the optical field at a particular spatial location [100]. Consider two scalar fields $E_1(t)$ and $E_2(t)$ that oscillate along two orthogonal directions $\hat{e}_1$ and $\hat{e}_2$, which are mutually orthogonal to the propagation direction $\hat{\imath}$. $E_1(t)$ and $E_2(t)$ are the complex analytic representations of the signals, meaning that there is zero spectral content at negative frequencies. The instantaneous Stokes parameters are defined as

$$S_0(t) = |E_1(t)|^2 + |E_2(t)|^2 \quad (3-55)$$
Table 3.1 Different types of polarizations and corresponding Stokes parameters

<table>
<thead>
<tr>
<th>Polarization</th>
<th>Stokes Parameters</th>
<th>Symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>Linear in ( \hat{e}_1 ) direction</td>
<td>( {1,1,0,0} )</td>
<td>← →</td>
</tr>
<tr>
<td>Linear in ( \hat{e}_2 ) direction</td>
<td>( {1,1,0,0} )</td>
<td>↑</td>
</tr>
<tr>
<td>Linear (+45^\circ)</td>
<td>( {1,0,1,0} )</td>
<td>∴</td>
</tr>
<tr>
<td>Linear (-45^\circ)</td>
<td>( {1,0,-1,0} )</td>
<td>_</td>
</tr>
<tr>
<td>Right-handed circular</td>
<td>( {1,0,0,1} )</td>
<td>⊙</td>
</tr>
<tr>
<td>Left-handed circular</td>
<td>( {1,0,0,-1} )</td>
<td>⊙</td>
</tr>
<tr>
<td>Unpolarized</td>
<td>( {1,0,0,0} )</td>
<td>*</td>
</tr>
</tbody>
</table>

\[
S_1(t) = |E_1(t)|^2 - |E_2(t)|^2 \\
S_2(t) = E_1(t)E_2^\ast(t) + E_1^\ast(t)E_2(t) \\
S_3(t) = i(-E_1(t)E_2^\ast(t) + E_1^\ast(t)E_2(t)).
\]  

(3-56)  
(3-57)  
(3-58)

In this formalism, we have defined the instantaneous Stokes parameters \( \{S_0, S_1, S_2, S_3\} \).

For optical signals, it is impossible to measure \( E_1(t) \) and \( E_2(t) \) or \( \{S_0, S_1, S_2, S_3\} \) at short timescales, therefore, time averaged quantities are used. However, with THz signals, \( E_1(t) \) and \( E_2(t) \) are both accessible and so we will use the instantaneous quantities.

The Stokes parameters determine whether the polarization is circular, linear, elliptical, or unpolarized. Relations within the parameters describe the degree of polarization and the geometry of the elliptical polarization if present. Table 3.1 shows several types of polarizations with their corresponding Stokes parameters. For unpolarized light, the fields oscillate with equal intensity independently in all directions.
Linearly polarized light only oscillates in the \( \hat{e}_1, \hat{e}_2 \), or a linear combination of the two, but does not oscillate in the orthogonal direction. Circularly polarized light rotates its polarization as it propagates.

The length scale in which the polarization is randomized in an isotropic random medium is generally comparable or larger than the transport mean free path [101]. This indicates that it requires more than one scattering event to rotate the polarization. Ballistic, snake, and diffuse photons are characterized by their polarizations as illustrated in Fig. 3.6. Ballistic photons are unscattered and completely maintain their initial polarization, whereas diffuse photons scatter many times and are unpolarized. Snake photons scatter more than once, but still maintain some memory of their initial polarization. The degree of polarization \( P(t) \) is defined as

\[
P(t) = \sqrt{\frac{\langle S_1(t) \rangle^2 + \langle S_2(t) \rangle^2 + \langle S_3(t) \rangle^2}{\langle S_0(t) \rangle}},
\]

(3-59)

where the angular brackets denote ensemble averaging. When the fields \( E_1(t) \) and \( E_2(t) \) are mutually incoherent, the degree of polarization is [102]

\[
P(t) = \frac{\langle S_1(t) \rangle}{\langle S_0(t) \rangle}
\]

(3-60)

or in terms of intensity

\[
P(t) = \frac{\langle I_1(t) \rangle - \langle I_2(t) \rangle}{\langle I_1(t) \rangle + \langle I_2(t) \rangle}.
\]

(3-61)

Several groups have demonstrated how polarization information can be used to image objects embedded in scattering medium by methods based on polarization contrast or timegating [83, 103-106]. In polarization contrast imaging [103], linearly polarized
light propagates into a turbid medium and separate images are taken of the co-polarized and cross-polarized light. The turbid medium randomizes the polarization of the photons that diffuse, however, the photons that reflect off the buried object will maintain some of their initial polarization. The diffuse photons contribute equally to the co-polarized and cross-polarized images, but the reflected photons will contribute mainly to the co-polarized image. By subtracting the cross-polarized image from the co-polarized, an image of the buried object is obtained. Polarization time gating takes advantage of snake photons, which propagate through the random medium, arrive at the detector earlier than diffuse photons [99]. Linearly polarized light is transmitted through a sample and images are taken of the co-polarized and cross-polarized light. The degree of polarization is computed as in equation (3-61) to discriminate between the early arriving snake photons and the later diffuse photons. The number of snake photons that arrive can be used to distinguish different types of tissues.

Besides for the use of imaging, the polarization dependence of light propagating through random media is an active topic of research. Bicout et al defined a characteristic length scale, which is the distance of propagation through a random media that the photons depolarize and showed that this length is typically greater than the transport mean free path [101]. Several groups have demonstrated that the degree of polarization could be used to time-gate the ballistic and snake photons [107, 108]. Recently, we observed this effect with THz radiation propagating through random media [70]. In addition, we showed that the spectral content depends on the state of polarization. In the case of broadband radiation, this effect can be used to distinguish photons that have
scattered a few times from those that propagate diffusively. This work is described in more detail in Chapter 5.

3.6 Coherent Backscattering and Localization

The study of wave propagation in a very strongly scattering medium is an active field of research with the goal of observing the localization of light. Anderson localization describes the scattering of electrons in a material with a large number of defects. If the scattering is strong enough, the diffusion regime breaks down and electrons no longer transport. The material will then behave like an insulator. Electrons and photons behave very similarly since they both exhibit characteristics of waves, and it is believed that similar effects can occur for light. In Anderson localization of photons, light gets trapped in closed loops of scattering paths and no longer propagates away from the source. This can be explained by the following. Light emitted from a source that propagates within a closed loop can return to the origin by two opposite paths as shown is Fig. 3.13. Because these path distances are equal, waves will interfere coherently at the source. This increases the probability of the wave returning to the source and therefore decreases the probability of propagating away from the source. This results in localized states.

A characteristic of localization is that the diffusion regime is expected to transition from a $L^1$ to an exponential decay of intensity as a function of slab thickness [11]. As shown in Fig. 3.5, absorption also appears as an exponential decay; therefore, it is a challenge to distinguish the effects of absorption and localization. The Ioffe-Regel criterion determines that localization will occur for $k\lambda_{sc} \leq 1$ [109], where $k$ is the
magnitude of the wave vector and $\lambda_{sc}$ is the mean free path. The Ioffe-Regel criteria then becomes $2\pi\lambda_{sc}/\lambda \leq 1$, which indicates that the wave is scattered before it can perform even one oscillation of the electric field.

One research group observed $k\lambda_{sc} = 1.5$, which is the smallest value measured for near infrared light [11]. In their experiment, they used gallium arsenide (GaAs) to create samples consisting of a semi-conductor powder. They were able to observe a breakdown in the diffusion regime, where the transmission coefficient transitioned into an exponential decay. Another group followed up their work using germanium (Ge) powder, which has a higher index of refraction than GaAs. They reported a value for the localization parameter of $k\lambda_{sc} = 4.5$, which is much higher than the GaAs powder [110].
Figure 3.14: Coherent backscattering and weak localization effect. The light scattered multiple times but returns to the source. The waves add constructively in the opposite direction of the emitted light, doubling the intensity of the backscattered light.

This is surprising result, because it would be expected that the higher index Ge, would result into a smaller localization parameter. Obviously, the subject still has many unanswered questions. Localization has also been observed in the microwave regime using a sample consisting of a mixture of teflon and metallic spheres [9, 111].

The precursor to Anderson localization is known as the coherent backscattering effect or weak localization [10, 69, 112]. Light transmitted into a strongly scattering medium will scatter several times and form a loop inside the medium; therefore, the light that exits in the backward direction could have traveled either direction along the closed path. Thus, the backscattered light experiences constructive interference [113]. Experiments have demonstrated that the intensity of the backscattered light is nearly twice the surrounding background intensity [112], because of this constructive interference. An example of the coherent backscattering effect is shown in Fig. 3.14.
Figure 3.15: A heuristic of the intensity of backscattered light as a function of angle. The backscattering cone has twice the intensity as the incoherent background due to constructive interference of the backscattered light. The width of the cone is inversely proportional to $k\lambda_{sc}$.

Light is propagated into a scattering medium and the backscattered radiation is measured as a function of angle; a heuristic plot of this is shown in Fig. 3.15. The intensity of the light escaping the medium in the backward direction is related to the angle of exit. As the angle of exit increases, there is a steep decline in the intensity. The width of the backscattering cone is approximately $0.7/k\lambda_{sc}$ [114]. The coherent backscattering effect demonstrates the propagation of waves in a closed path, which is an early signature of the onset of localization.

Propagation of electromagnetic waves through random media gives rise to a number of interesting phenomena, both coherent and incoherent effects. Multiple scattering influences the phase, amplitude, and polarization of light. The majority of previous studies have examined these quantities with narrowband sources. Accurate descriptions for these quantities are only available for special cases. However, there are still many more questions to be answered concerning the properties of diffuse photons.
The random phasor model describes the probability distribution of the electric field for narrowband light, but how do the statistics of diffuse waves change when a broadband source is employed? Besides the degree of polarization, what other markers provide indicators for the onset of diffusion? T-rays provide a wealth of information for answering these questions and probing the complex process of scattering, because of their broad bandwidth and accessibility of both the intensity and phase.
Chapter 4

Statistics of Multiply Scattered Broadband Terahertz Pulses

4.1 Introduction

The propagation of classical waves in the presence of random scattering is a topic of considerable interest in many research communities. In particular, scattering of electromagnetic waves can lead to a rich array of phenomena [63, 64]. In a random medium, the propagating field can be described as a superposition of unscattered and scattered waves. Diffusive propagation occurs after the incident wave travels a distance much larger that the Boltzmann transport mean free path \( l_n \) [64]. In this case, the incident light beam is completely randomized, and only multiply scattered photons are transported through the medium. This diffusive wave is of much interest because it can be used for locating and imaging objects buried in the random medium [91, 92, 115]. In addition, the statistics of the diffusive wave can be used to extract information on the nature of the random medium [116], and are a key indicator of the onset of localization [110]. Much of the research on diffusive optical waves has concentrated on the case of monochromatic or narrow-band waves [7, 8, 68, 115, 117-120]. Several authors have used short optical pulses as a means for separating the diffusive portion of the wave from the ballistic light [110, 121-124]. Others have used low-coherence interferometry to extract relative phase information [125-129]. Short acoustic pulse propagation is also extensively studied in the context of seismic tomography [98]. However, the treatment of the statistics of broadband fields in random media remains largely unaddressed.

Here, we report measurements of the electric field of multiply scattered
broadband optical pulses. We compute the statistics of these random fields, and demonstrate the connections to the case of monochromatic radiation. These measurements employ THz time-domain spectroscopy (THz-TDS), in a configuration quite similar to the one described previously [50, 73, 74]. Using this technique, it is possible to generate pulses with a fractional bandwidth in excess of 100% (50 GHz – 1 THz). Furthermore, the coherent measurement of the electric field permits extraction of both the amplitude and phase of the field, with a temporal resolution better than one optical cycle, without the use of interferometric techniques. As a result, one directly observes, among other things, the distribution of photon transit times (i.e., the path-length distribution function). This quantity usually must be extracted from time-integrated measurements using spatial intensity correlations [120]. Finally, we emphasize that these measurements have been performed in a three-dimensional sample, rather than in the waveguide geometry customarily employed in microwave measurements [6].

4.2 Experimental Setup

We construct a model random medium using commercially available polytetrafluoroethylene (PTFE, teflon) spheres, with a diameter of 0.794 ±0.025 mm. Teflon is an excellent material for these measurements, because of its low absorption: $\alpha < 0.1$ cm$^{-1}$ for frequencies below 500 GHz, and $\alpha \approx 0.5$ cm$^{-1}$ at 1 THz [90]. Also, the refractive index of teflon, $n = 1.4330$, is nearly independent of frequency throughout the spectral range of the measurements. The spheres are poured into a cubic teflon cell with dimensions of (4 x 4 x 4 cm$^3$). The volume fraction of the spheres in the sample cell is measured to be $0.56 \pm 0.04$.

In a previous experiment, we measured the mean free path $\lambda_{sc}(\omega)$ of a similarly
Figure 4.1: The scattering mean free path $\lambda_{sc}(\omega)$ versus free-space wavelength, on a log-scale. Within the bandwidth of the THz pulse, the mean free path ranges from ~1 to ~70 mm.

constructed random media [71]. We measured the ballistic pulse (shown in Fig. 4.4) through a series of sample cells with increasing path length that contained these teflon spheres. By measuring the transmission decay as a function of path length, we were able to extract $\lambda_{sc}(\omega)$. Shown in Fig. 4.1 is the measured scattering mean free path versus wavelength, on a log scale. In these samples, $\lambda_{sc}(\omega)$ ranges from ~1 to ~70 mm within the bandwidth of the THz pulse. As a result, certain wavelengths within the THz pulse scatter many times and propagate diffusively, while others scarcely scatter.
Figure 4.2: A schematic of the diffusion experiment. The detector is situated at 90° to the input beam direction, in order to avoid measuring ballistic THz photons.

Because our primary interest is in studying diffusively propagating photons in this experiment, we must ensure that only multiply scattered photons are measured and no ballistic radiation is detected. The experimental setup is illustrated in Fig. 4.2. Single-cycle THz pulses are generated using the Picometrix T-Ray2000™. This fiber based system allows for reconfigurable transmitter and receiver locations. The radiation is then focused into the random media using a lens (focal length = 6 cm), and the emerging radiation is measured at an angle of 90° with respect to the incident beam direction. This
setup ensures only diffusive radiation is measured and that no ballistic radiation reaches the detector. The generated THz radiation is primarily horizontally polarized, and the detector measures only the horizontal component.

The temporal electric field of the THz radiation is measured via ultrafast photoconductive sampling [21]. The peak amplitude of the scattered radiation is roughly a factor of 500 smaller than the incident pulse. This requires that we modulate the beam with a mechanical chopper and use a lock-in amplifier to obtain appreciable signal-to-noise. Each waveform is scanned over a delay window of 400 ps at a rate of 0.50 ps/s and averaged 12 times, which took almost 3 hours to acquire.
Figure 4.4: Electric field measurements of the incident and ballistic (scaled by a factor of 2) radiation. The waveforms have been offset for clarity sake. The ballistic and incident waveforms have much larger amplitudes and contain fewer oscillations than the diffusive waveform shown in Fig. 4.3.

Fig. 4.3 shows several measurements of diffusive THz waveforms. Between measurements, the spheres are stirred. The time axis has an arbitrary start delay that is dependent on the distance between the transmitter and receiver. Each waveform corresponds to a realization of a unique configuration of the random medium. These waveforms have been spectrally filtered at both low and high frequencies to improve the signal-to-noise ratio, which is about 10:1 at the spectral peak. Filtering is necessary, because the raw waveforms contain a low frequency drift that is due to an effect within the T-ray system and is not a result from the random medium. For reference, the signal-
to-noise ratio for a measurement of the incident single-cycle pulse (shown in Fig. 4.4) exceeds 20,000 after equivalent spectral filtering. We measure waveforms for 22 different sample configurations. The result of the multiple scattering is a randomization of the phase, which produces many oscillations in the scattered field that persist for hundreds of picoseconds. In contrast, the incident pulse consists of one oscillation and has a duration of less than one picosecond. Multiple scattering in a diffusing medium will cause photons to travel a variety of paths of different lengths through the medium and therefore will arrive at the detector at different times. A consequence of this is a short pulse will temporally spread after propagating through a diffusing medium.

We compare the measurements of the diffusive, ballistic, and incident radiation. The ballistic radiation is measured by placing the receiver on the opposite side of the sample medium and detecting the ballistic photons. As shown in Fig. 4.4, the ballistic waveform is a short pulse, containing only a few oscillations, similar to the incident waveform. This is in stark contrast to the many cycles and long extent of the diffusive radiation. Also, the ballistic pulse is longer in duration than the incident pulse. This can be explained as follows. Higher frequencies, in general, have shorter mean free paths (see Fig. 4.1) and thus have a higher likelihood of scattering. The ballistic waveform consists of those photons that remain unscattered. Therefore, the random media acts as a low pass filter to the incident pulse, removing the higher frequencies [74], which results in the spreading of the pulse. However, the many oscillations that are present in the diffusive waveform are absent in the ballistic waveform, because the ballistic radiation maintains memory of its original phase whereas the phase of the diffusive radiation is completely randomized.
Figure 4.5: The autocorrelation functions of the incident (black), ballistic (red), and diffusive light (blue). The width of the autocorrelation is inversely proportional to the bandwidth. The incident pulse and multiply scattered waveforms have the same spectral content, whereas the ballistic light loses the higher frequencies.

The diffusive waveforms not only differ from the ballistic waveforms in their temporal extent, but also in their spectral content. Fig. 4.5 shows the autocorrelation functions of the incident, ballistic, and diffusive radiation. The Fourier transform of the autocorrelation function is the power spectrum of the signal. Therefore, the temporal width of the autocorrelation function is inversely proportional to the bandwidth of the signal. The autocorrelation functions of the incident and diffusive waves have very similar widths, which suggests that they contain similar spectral bandwidths. The random medium scatters photons at all frequencies, therefore the diffusive radiation contains the same spectral content as the incident pulse. In contrast, the autocorrelation function of the ballistic waveform has a much broader width than the diffusive photons,
indicating a narrower spectral bandwidth. This is because the higher frequencies are filtered out by the random medium.

Owing to our measurements in the time-domain, we can calculate the time-of-flight distribution of the photons transmitted through the random medium by configurationally averaging the intensity of the waveforms:

$$\langle I(t) \rangle = \langle E^*(t)E(t) \rangle$$

(4-1)

where $E(t)$ is the analytical representation of the transmitted electric field and the angular brackets denote ensemble averaging. Fig. 4.6 shows the calculated photon time-of-flight distribution of the transmitted radiation.

The diffusion equation describes the transport of a short light pulse through a strongly scattering medium [95]. Here, the transmission equation derived from the diffusion equation is restated from equation (3-44) for convenience:

$$T(t) = (4\pi Dc)^{-1/2} t^{-3/2} \exp(-\mu_a c t)$$

$$\times \left\{ (L - z_0) \exp\left[ -\frac{(L - z_0)^2}{4Dt} \right] - (L + z_0) \exp\left[ -\frac{(L + z_0)^2}{4Dt} \right] \right\}$$

$$+ (3L - z_0) \exp\left[ -\frac{(3L - z_0)^2}{4Dt} \right] - (3L + z_0) \exp\left[ -\frac{(3L + z_0)^2}{4Dt} \right] \right\},$$

(4-2)

where $D = 3[\mu_a + (1-g)\mu_s]^{-1}$ is the diffusion coefficient, $c$ is the speed of light within the medium, $t$ is delay time, $L$ is the propagation length, and $z_0 = [(1-g)\mu_s]^{-1}$ is the distance traveled before the first scattering event. The diffusion coefficient is a function of the absorption coefficient $\mu_a$, the scattering coefficient $\mu_s = 1/\lambda_{sc}$, and the average cosine $g = \langle \cos \theta \rangle$.

We fit the transmittance equation in (4-2) to the measured photon time-of-flight
distribution using a least squares fitting procedure. The speed of light was assumed to be 0.299/1.25 mm/ps. The factor of 1.25 is the estimated effective index of refraction of the medium [74]. Although, teflon does have some absorption, it was set to 0.0 mm\(^{-1}\), because we found that it does not greatly influence the diffusion curve. The propagation length is 28.3 mm, which is the distance from the center of the incident face to the center of the exit face. Using these fixed values, we fit \( \mu_s \) and \( g \) from (4-2) to the measured photon time-of-flight distribution and they were found to be 0.62 and to 0.223 mm\(^{-1}\) respectively. The result is shown in Fig. 4.6. Qualitatively, the measured distribution behaves similarly to the transmittance equation; an initial fast upshot followed by a slow
decay. However, at later times, the measured distribution decays much slower than the transmittance equation. There are several reasons for the discrepancy between theory and experiment. First, both $\mu_s$ and $g$ depend on frequency, where as the diffusion model assumes that they are constant. Second, the diffusion model requires that $L \gg \lambda_e$. Although this is true at higher frequencies, the lower frequencies have a mean free path that exceeds the propagation length. Finally, the model assumes that the medium is an infinite slab, whereas our experimental geometry is a cube of scatterers. This illustrates the need to further develop theories of the broadband case.

4.3 Statistics of the Scattered Field

We examine the statistics of the electric field. By taking the Fourier transform of these waveforms, we can extract both the real $r = \text{Re}[E(\omega)]$ and the imaginary $i = \text{Im}[E(\omega)]$ parts of the scattered electric field $E(\omega)$. From these measurements, we are able to obtain the probability distribution of the real and imaginary parts of the transmitted electric field, $P(r)$ and $P(i)$. Fig. 4.7 shows the joint probability distributions of the real and imaginary parts calculated from the diffusive waveforms. The distribution is approximately zero-mean, which implies that the waveforms do not contain any ballistic photons [6]. The distribution is circularly symmetric, indicating that the real and imaginary parts are uncorrelated.

We can formulate the probability distribution functions of the real and imaginary parts. Assuming that the complex electric field component at a given frequency is the sum of a large number of random phasors, the central limit theorem predicts that the scattered field should obey Gaussian statistics [6, 96]. Assuming that the phase is
Figure 4.7: The joint probability distribution function of the real and imaginary parts of the electric field. The distribution is zero-mean, because the waves are purely diffusive and no ballistic photons are present. Because the real and imaginary parts are uncorrelated, the distribution is circularly symmetric.

uniformly distributed, the joint probability distribution of the real and imaginary parts at a given frequency $\omega$ can be considered zero-mean, jointly Gaussian variables, and therefore

$$P(r, i | \omega) = \frac{1}{2\pi \sigma_\omega(\omega)^2} \exp\left[ -\frac{r^2 + i^2}{2\sigma_\omega(\omega)^2} \right]$$  \hspace{1cm} (4-3)

where the variance $\sigma_\omega(\omega)^2 = \langle I(\omega) \rangle / 2$. $\langle I(\omega) \rangle$ is the spectral intensity of the diffusive light averaged over all configurations of the medium, and it is dependent on the input pulse and the scattering parameters of the random media. To determine the joint
Figure 4.8: The probability distribution of the normalized real (triangles) and imaginary (open squares) parts of the complex scattered electric field, $P(r/\sigma)$ and $P(i/\sigma)$, plotted on a log scale. The dashed line shows the Gaussian distribution, which is the result expected for monochromatic radiation [6]. The solid curve is the prediction of equation (4-5), using an experimentally determined estimate for the mean spectral intensity.

distribution of the real and imaginary parts within a finite frequency range $\Delta \omega = \omega_2 - \omega_1$, we integrate (4-3) over $\omega$ and normalize by the bandwidth $\Delta \omega$,

$$ P(r,i) = \frac{1}{\pi \Delta \omega} \int_{\omega_1}^{\omega_2} \frac{1}{\langle I(\omega) \rangle} \exp \left[ - \frac{r^2 + i^2}{\langle I(\omega) \rangle} \right] d\omega $$  \hspace{1cm} (4-4)

This expression may be interpreted as the superposition of a large number of zero-mean Gaussian distributions (one for each spectral component), each with a unique variance proportional to $\langle I(\omega) \rangle$. The marginals $P(r)$ and $P(i)$ are equivalent to each other and
Figure 4.9: The configurationally averaged intensity of the transmitted radiation. The amplitude depends on the intensity of the incident light, as well as, the parameters of the random media such as the mean free path and the average cosine.

can be computed as

\[ P(a) = \frac{1}{\Delta \omega} \int_{a_1}^{a_2} \frac{1}{\sqrt{\pi (\langle I(\omega) \rangle)}} \exp \left[ -\frac{a^2}{\langle I(\omega) \rangle} \right] d\omega \]  

(4-5)

with \( a = \{ r, i \} \), and with variance

\[ \sigma^2 = \frac{1}{2\Delta \omega} \int_{a_1}^{a_2} \langle I(\omega) \rangle d\omega. \]  

(4-6)

The variance of \( P(a) \) is proportional to the integrated average intensity of the diffusive light. This is analogous to diffusive monochromatic waves where the variance is proportional to the average intensity [6]. We extract the complex parts of \( E(\omega) \) over the
50 GHz – 500 GHz spectral range, where there is appreciable signal in the measured waveforms. The probability distributions of the normalized real and imaginary parts $P(r/\sigma)$ and $P(i/\sigma)$ are shown in Fig. 4.8. The real and imaginary parts are zero-mean and have nearly identical distributions as predicted by (4-5). As expected, the Gaussian distribution expected for the case of monochromatic illumination [6] (dashed line) does not accurately fit the data. In order to compare to the predicted result (equation (4-5)), we extract an estimate of the average intensity $\langle I(\omega) \rangle$ (shown in Fig. 4.9) by averaging the frequency-dependent intensity spectrum over the 22 measured waveforms. By substituting $\langle I(\omega) \rangle$ for the average intensity in (4-5), we can numerically calculate $P(a/\sigma)$. The result (solid line) is in excellent agreement with the experimental data.

In order to predict $P(a)$ without using the data, it is necessary to predict the power spectrum. This is not straightforward. The shape of the probability distribution $P(a)$ is greatly influenced by the power spectrum of the transmitted radiation. Shown in Fig. 4.9 is the configurationally averaged intensity $\langle I(\omega) \rangle$ from 50 GHz – 500 GHz. The transmitted intensity is dependent on several parameters: the mean free path, the system geometry, and the power spectrum of the incident beam. However, we do not have an adequate model available to describe our experiment. The diffusion model only describes the transmission of the higher frequencies, because the lower frequencies have too large of a mean free path and the geometry is too small to assume that the photons propagate diffusively. There has been some recent work, which involved applying Monte Carlo methods to numerically predict the transmission of THz pulses through a slab of scatterers [130]. Obviously, more work needs to be done to model THz propagation
through strongly scattering media.

The random phasor sum model also predicts the statistics of the intensity. For monochromatic waves, the probability distribution of the intensity is predicted to obey a Rayleigh distribution [96]:

\[
P(I \mid \omega) = \begin{cases} 
\frac{I}{\sigma(\omega)^2} \exp\left(-\frac{I^2}{2\sigma(\omega)^2}\right) & I > 0 \\
0 & \text{otherwise},
\end{cases} \tag{4-7}
\]

where \(\sigma(\omega)^2\) is the variance of the intensity \(I(\omega)\). As with the real and imaginary parts in (4-4), we can integrate over the spectrum to compute the intensity distribution for broadband radiation:
Figure 4.11: The probability distribution of the normalized spectral phase derivative, plotted on a log scale. The solid line is the probability distribution given in (4), equivalent to the monochromatic case [5], with $Q = 0.234$.

$$P(I) = \begin{cases} 
\frac{1}{\Delta \omega} \int_{\omega_1}^{\omega_2} \exp\left(-\frac{I^2}{2\sigma(\omega)^2}\right) d\omega & I > 0 \\
0 & \text{otherwise.}
\end{cases} \tag{4-8}$$

Fig. 4.10 shows the measured intensity distribution and the prediction computed by (4-8). The variances were estimated from data. The prediction agrees quite well with the estimated probability distribution function of the intensity for over seven standard deviations ($I > 7\sigma$).

The statistics of the phase derivative $d\phi / d\omega \equiv \phi'$ are also of great importance. In the case of narrow-band wave packets, the ensemble average of this quantity is inversely proportional to the transport velocity, so it can be interpreted as a time delay for
photons in the medium. For broadband waves, its connection to the concept of a delay time is questionable, because of the randomization of the spectral phase. Nevertheless, it is instructive to investigate the statistics of $\phi'$, because of its relevance in the study of higher-order correlations [5]. For narrow-band wave packets, the probability distribution for the normalized phase derivative has been derived within the Gaussian approximation as [5, 8]

$$P\left(\frac{\hat{\phi}'}{\langle \phi' \rangle} = \frac{\phi'}{\langle \phi' \rangle}\right) = \frac{1}{2} \frac{Q}{[Q + (\phi' - 1)^2]^{3/2}},$$  \hspace{1cm} (4-9)

where $\hat{\phi}'$ is the phase derivative normalized to its ensemble-averaged mean, and where $Q$ is a parameter related to the absorption length and the sample geometry [131]. For broadband waves, $P(\hat{\phi}')$ can be derived by integrating (4-9) over frequency with an appropriate weighting function, as in equations (4-4) and (4-5) above. However, because in our measurements the absorption length is approximately constant over the entire bandwidth of the radiation, $Q$ should not vary much over frequency. Since this is the only parameter, the distribution of the phase derivative for broadband waves should also be given by (4-9). Fig. 4.11 shows the probability distribution for $\hat{\phi}'$, extracted from the Fourier transforms of the measured waveforms. The solid curve is the predicted result (equation (4-9)), with $Q = 0.234$. As anticipated, the theoretical expression derived for the monochromatic case can also accurately predict the statistics of the broadband wave packet.

4.4 Summary

We find that the probability distributions of the amplitude and phase derivative of
diffusive broadband radiation is simply the integration over the frequency band of their respective narrowband distributions with a frequency dependent variance. The probability distributions of the real and imaginary parts of the field are a superposition of jointly Gaussian distributions. Their zero-means verify that we indeed are measuring diffusively propagating photons. The intensity distribution follows a superposition of Rayleigh distributions. However, the statistics of the phase derivative exactly match that of the narrowband case, because they are independent of frequency.

In order to predict the broadband statistics, we had to estimate the frequency dependence variance from the measurements itself. Future work would involve developing a model to predict these variances from the physical properties of the medium: shape, volume fraction, index of refraction, etc. Although Monte Carlo methods can make accurate predictions, they are computationally intensive and are not concerned with the physical properties of the medium as much as with the mean free path. A more satisfying approach would be to employ finite element methods to develop a better model for our data.

In conclusion, we report the first use of THz time-domain spectroscopy in the study of diffusive waves. The direct measurement of the multiply scattered electric field allows for the computation of statistics for both amplitude and phase. We have extended the theoretical framework, developed for monochromatic waves, to the broadband case and found excellent agreement with our measured results. Using these time-resolved measurement techniques, it should also be possible to extract information on the nature of specific scattering events within the random medium.
Chapter 5
Polarization Dependence of Multiply Scattered Broadband Terahertz Pulses

5.1 Introduction

The study of the polarization of multiply scattered light waves has been a topic of considerable interest recently. When considering the propagation of light in a random medium, a natural length scale is $\lambda_{sc}$, the transport mean free path, which is the mean propagation distance required to randomize the direction [64]. In the study of depolarization, Bicout et al. [101] introduced a second characteristic length, $\xi$, which is the propagation distance for randomization of the polarization. In an isotropic random medium, this depolarization length is generally comparable to or somewhat larger than $\lambda_{sc}$, indicating that more than one scattering event is required to randomize the polarization [101, 132-134]. As a result, it is not surprising that time-resolved studies have observed a delay in the emergence of cross-polarized radiation from a random medium illuminated with linearly polarized light, due to the larger average number of scattering events experienced by this component [83, 99, 107, 135, 136]. This result has inspired new imaging schemes based on polarization gating or polarization contrast [83, 103-106, 137]. Such ideas rely on the notion of ‘snake’ or ‘quasi-straightforward-propagating’ photons [138], which have scattered a few times but retain some degree of spatial coherence and polarization memory. If one can distinguish ballistic and snake photons from diffusive photons using the degree of polarization, then useful image information can be extracted.
We point out that the majority of these studies have employed narrowband radiation, for which the spectral dependence of the properties of the medium (e.g., the mean free path) can be neglected. Here, we explore the opposite situation, that of extremely broadband light. In this case, we show that the spectral content of the radiation emerging from the random medium is also related to the number of scattering events. For broadband waves, this is an alternate measure of the loss of spatial coherence of the wave. This result should have important implications for broadband polarization contrast imaging [83, 103-106, 137].

For these measurements, we use terahertz (THz) time-domain spectroscopy, in a configuration similar to that described in Chapter 4 [71-74]. This technique permits generation of single-cycle pulses of radiation, with spectral content spanning more than one order of magnitude in frequency. The radiation can be detected coherently, so that both the field amplitude and temporal phase are measured with sub-cycle time resolution. As a result, quantities such as the photon time-of-flight distribution [88, 126] are directly observed. In addition, as described below, we can experimentally determine the precise limits of the single-scattering regime, in the time domain.

5.2 Experimental Setup

The experimental setup is illustrated in Fig. 5.1. Single-cycle THz pulses are focused at the edge of the sample scattering medium, and the emerging radiation is measured at an angle of 90° to the incident pulse direction. The random medium consists of a dense collection of teflon spheres (refractive index $n = 1.4330$ [74, 90]) with a diameter of $0.794 \pm 0.025$ mm. The spheres are poured into a cubic Teflon cell with dimensions of $(4 \text{ cm})^3$ at a measured volume fraction of $0.56 \pm 0.04$. In these samples,
the mean free path of the radiation varies dramatically within the bandwidth of the THz pulse, by a factor of \approx 70 \ (see \ Fig. \ 4.1) \ [74].

The THz radiation is generated and measured with the Picometrix® T-Ray 2000™, which uses fiber-coupled photoconductive antennas to allow for reconfigurable transmitter and receiver locations without disturbing the alignment of the femtosecond laser pulse on the antenna. The temporal duration of the emitted THz radiation is less
than one picosecond, and the spectral range spans from 50 GHz to 1 THz. The generated radiation is almost a pure horizontal linear polarization, with a small vertical component [139] that is filtered out by a wire grid polarizer placed before the sample. The detector is placed behind a second polarizer to ensure that only one polarization is measured. The detector and second polarizer are rotated to either 0° or 90°, to measure the electric field $E_{\perp}(t)$ of each polarization separately. We designed a special casing for the detector in order to rotate it on its axis. Great care is taken to ensure that the position of the detector does not change during this rotation, so that the temporal delay calibration is maintained. Each waveform is scanned over a delay window of 600 ps at a rate of 0.50 ps/s and averaged 12 times, which took over 4 hours to acquire. We have measured $E_{\perp}(t)$ and $E_{\parallel}(t)$ for 33 different realizations of the disorder. The waveforms are very similar to those in Fig. 4.3 in the previous chapter with the waveforms of the horizontal polarization having larger amplitudes than the vertical polarization. The waveforms contain many oscillations and persist for hundreds of picoseconds as a result of the randomization of the phase from the scattering process. In contrast, the incident pulse consists of one oscillation and has a duration of less than one picosecond.

Since the receiver is fiber-coupled, the rotation has a negligible effect on the alignment of the laser pulse on the antenna. However, bends and rotations in the fiber influence the polarization of the laser pulse. The amplitude of the emitted THz radiation is dependent upon the polarization of the laser pulse that is incident on the antenna. To characterize this dependence, we set the transmitter at 45° and measure the horizontal and vertical polarization of the emitted radiation separating by rotating the receiver and the filter to 0° and 90°. The emitted radiation should be equal at each angle if the rotation
Figure 5.2: The three types of photons that exit at 90 degrees after scattering from a collection of spheres: single scatter, snake, and diffuse. At 90 degrees, there are no ballistic photons present, but single scatter photons maintain their polarization, because more than one scattering event is required to rotate polarization. Diffuse photons have a completely randomized polarization. Snake photons span the intermediate regime between being fully polarized and unpolarized.

procedure does not disturb the polarization in the optical fiber. We observed that the amplitude of the measured THz radiation at 90° is roughly 80% of the amplitude at 0°. This measurement serves as a calibration of the sensitivity of the detector in the two different orientations. In order to correct for this, we scale the perpendicular waveforms accordingly.
In Section 3.3, we discussed the three types of photons that propagate through a slab of scatterers: ballistic, snake, and diffuse. These photons exit the medium in the same direction of the incident wave. Each type of photon differs in its polarization. Ballistic photons are unscattered and completely maintain their initial polarization, whereas diffuse photons scatter many times and are completely unpolarized. Snake photons span the intermediate regime from completely polarized to unpolarized. In our experimental geometry, these definitions are modified, because the photons are measured at 90° as illustrated in Fig. 5.2. Ballistic photons are no longer present, because every photon must scatter at least once. Instead, there are single scatter photons, which for the case of isolated spherical particles, completely maintain their polarization, because Mie theory tells us that spherical particles require more than one scattering event to rotate polarization. However, for the situation of close-packed spheres, this is only approximately true. Snake photons are now defined as those photons that scatter more than once, but still maintain some polarization memory. Diffuse photons have random polarization.

5.3 Photon Time-of-Flight and Stokes Parameters

The photon time-of-flight (TOF) distributions of the parallel \( \langle I_\parallel(t) \rangle \) and perpendicular \( \langle I_\perp(t) \rangle \) polarizations can be computed by averaging the intensity of the scattered radiation over all configurations of the random medium. Fig. 5.3 shows the TOF distributions for both polarizations as well as the sum \( \langle I_{\text{Total}}(t) \rangle \). Our measurements were limited to time delays between 400-1000 ps, because of the finite length of the optical delay line in the THz time-domain spectrometer. The TOF distribution should decay to 0 at delay times beyond the limits of our measurement window. The cross-
Figure 5.3: The measured time of flight distributions of the parallel polarization \(\langle I_{\parallel}(t)\rangle\), perpendicular polarization \(\langle I_{\perp}(t)\rangle\), and the sum \(\langle I_{\text{total}}(t)\rangle\). Note that the time axis here and throughout this paper has an arbitrary \(t = 0\), which is determined by the absolute transmitter-to-receiver distance and is the same for all of our measurements.

Polarized radiation exits the medium approximately 30 ps later than the co-polarized radiation. The co-polarized radiation consists of photons that have scattered one or more times, whereas the cross-polarized radiation requires more than one scattering event to occur in order to rotate the polarization. The 30 ps delay results from the larger average number of scattering events experienced by the perpendicular radiation, and is consistent with previous reports in which it was found that \(\xi > \lambda_{sc}\) \([83, 99, 107, 135, 136]\).

The TOF distributions describe the arrival times of the different polarizations, but do not provide any information of their relationship to each other. However, the instantaneous Stokes parameters characterize the degree and type of polarization of an
Figure 5.4: The instantaneous stokes parameters normalized by the total intensity $\langle S_0(t) \rangle$: $\langle S_1(t) \rangle$, $\langle S_2(t) \rangle$, $\langle S_3(t) \rangle$. Between 500-600 ps, the radiation is horizontally polarized. After 600 ps, it is unpolarized.

An electromagnetic wave. The instantaneous Stokes parameters are defined in relation to the polarization of the incident radiation as

\begin{align}
S_0(t) &= |E_{||}(t)|^2 + |E_{\perp}(t)|^2 \\
S_1(t) &= |E_{||}(t)|^2 - |E_{\perp}(t)|^2 \\
S_2(t) &= E_{||}(t)E_{||}^*(t) + E_{\perp}(t)E_{\perp}^*(t) \\
S_3(t) &= i(-E_{||}(t)E_{\perp}^*(t) + E_{||}^*(t)E_{\perp}(t)),
\end{align}

(5-1) (5-2) (5-3) (5-4)
where $E_1(t)$ and $E_2(t)$ represent the complex electric field of the polarizations that are respectively parallel and perpendicular to the incident pulse. In our experiment, we measure the real part of the electric field, but the complex signal can be obtained via the Hilbert transform [100]. Fig. 5.4 shows the normalized Stokes parameters computed from (5-1) – (5-4) and averaged over the different configurations of spheres. We smooth the Stokes parameters with a moving average window to reduce some of the measurement noise. At early times, $\langle S_1(t) \rangle / \langle S_0(t) \rangle$ decays from a maximum value of 1, while the other two parameters remain at the noise level. These are the Stokes values that are characteristic of horizontal linearly polarized light. Since $\langle S_2(t) \rangle = \langle S_3(t) \rangle = 0$, there
is no circularly polarized radiation present. However, at later times (after 600 ps), the
Stokes parameters are not appreciably different from zero, which means that the radiation
is unpolarized.

We further explore the relationship between the two polarizations by examining
the intensity correlations between them. However, rather than using the entire
waveforms, we choose a temporal window to highlight correlations that occur at
particular time-delays, similarly to the method used by Jian et al. [3]. We compute the
intensity correlation function with a variable-delay window as defined by

$$C_\gamma(\tau, T) = \frac{1}{C_0} \left( \int_{-\infty}^{\infty} \left[ I_\parallel(t) \cdot W_\tau(t) \right] \left[ I_\perp(t+\tau) \cdot W_\tau(t+\tau) \right] dt \right)$$  \hspace{1cm} (5-5)$$

where $\tau$ is the correlation offset. $W_\tau(t)$ is a window function defined to be unity in a 100
ps symmetric window about $t = T$ and zero otherwise. The means of the intensities in
each window are subtracted out before the computing the correlation. The angular
brackets indicate an ensemble average over all polarization pairs. $C_0$ is a normalization
factor that ensures that $C_{I_\parallel=I_\perp}(\tau = 0, T) = 1$.

The result of this computation is shown in Fig. 5.5. The two polarizations are
weakly correlated with a maximum value that is less than 30%. Nevertheless, there is
appreciable correlation at window delay times of 500-600 ps and offsets of 0-50 ps. The
window of 500-600 ps corresponds to the time that the radiation has a measureable linear
polarization component as shown in Fig. 5.4. Outside this range, when the radiation is
unpolarized, the correlation is nearly 0. The correlation function is not symmetric about
the 0 offset and is larger in amplitude at positive offsets. Thus, the intensity of the
perpendicular polarization is more strongly correlated to that of the parallel polarization
Figure 5.6: The degree of polarization $P(t)$ (open circles) and the time-windowed intensity correlation function $C_\parallel$ (solid line). $C_\parallel$ is computed by windowing $I_\parallel(t)$ and $I_\perp(t)$ in the time domain using a 100 ps square window and computing the correlation as a function of the window position, as described in reference [3]. The vertical dash lines indicate the range of times during which single scattered photons arrive, as determined from Fig. 5.6.

when the co-polarized radiation is delayed. This corresponds to the result in Fig. 5.3, when we observed that the cross-polarized radiation exits at a later time than the co-polarized radiation.

We may also compute the time-resolved degree of polarization as shown in Fig. 5.6. As the measured fields $E_\parallel(t)$ and $E_\perp(t)$ are mutually incoherent, this quantity is equal to the normalized Stokes parameter $\langle S_1(t) \rangle / \langle S_0(t) \rangle$ [102]:

$$P(t) = \frac{\langle S_1(t) \rangle}{\langle S_0(t) \rangle} = \frac{\langle I_\parallel(t) \rangle - \langle I_\perp(t) \rangle}{\langle I_\parallel(t) \rangle + \langle I_\perp(t) \rangle}.$$  \hspace{1cm} (5-6)
Equation (5-6) is also known as the polarization contrast, which is used for polarization-difference imaging [103]. For early times, before the arrival of any photons, this quantity is undefined. After ~510 ps, the polarization begins to decay, until it essentially vanishes after ~600 ps. The later times are characteristic of diffuse photons, which have completely random polarizations. Fig. 5.6 also compares the averaged intensity cross-correlation \( C_\tau = \langle I_i(t)I_\perp(t) \rangle \), computed using a sliding time window with \( \tau = 0 \), on the same time axis, to the degree of polarization. The cross-correlation also shows an enhancement in roughly the same time window. We find that later delay times have different degrees of polarization and correlation as compared to that of early times.

5.4 Determining Single-Scatter Arrival Times

Several authors have noted that the decay of \( P(t) \) is exponential in the number of scattering events according to \( P(t) \sim \exp(-n/\langle n \rangle) \) where \( \langle n \rangle \) is defined as the average number of scattering events [101, 134, 135]. In the broadband case, that analysis is complicated by the fact that the mean free path varies strongly within the bandwidth of the radiation. However, because of the high time resolution of our measurements, we are able to estimate the mean number of scattering using a novel experimental procedure. Without changing any of the other parameters of the measurement, we place a long aluminum rod vertically into the random medium, as illustrated in Fig. 5.7. Because aluminum is such a strong reflector, the intensity of radiation scattered only from the rod is much larger than the scattered radiation measured without the rod. Thus, the signal measured with the rod in place is dominated by the radiation scattered directly from this metal object. The signal from photons scattered both from the rod and also from any of
Figure 5.7: Top: A schematic illustrating the method used to determine the arrival times of single scattered photons. A metal rod is placed at several different locations in the medium, giving rise to a very strong single-scattering signature. Bottom: Waveforms (b) and (c) correspond to the locations of the metal rod that resulted in the earliest (PS) and the latest (PL) arrival times. Waveform (a) is measured without the rod present.

the teflon spheres is much weaker than the very strong single-scattering signal from the rod alone. Thus, by moving the rod to many different locations in the path of the incident radiation, it is possible to determine the maximum and minimum transit times for single-
scattered photons. This procedure essentially permits us to calibrate the arbitrary delay axis in terms of the earliest and latest arriving single-scattered photons.

Fig. 5.7 illustrates the two rod locations that corresponded to the longest and shortest photon paths, $P_L$ and $P_S$ respectively. The measured waveforms corresponding to those limits exhibit clear signatures of the single scattering of radiation from the rod and are therefore unambiguous markers of the limits of the temporal window corresponding to single scattering. The dashed vertical lines in Fig. 5.7 show that these limits correspond well to the temporal window in which $P(t)$ is non-zero, and also to the window during which the correlation $C_r(\tau)$ is enhanced. This result suggests that only two scattering events are needed to randomize the polarization, so $\xi / \lambda_{sc} < 2$. We note that most earlier methods for determining this depolarization rate have relied on a computation of the scattering cross-section using Mie theory [83, 101, 133-135], which is difficult to apply in the case of irregularly shaped or densely packed scatterers [79].

5.5 Spectral Shifts Between Polarizations

For broadband waves, the evolution of the spectral content of the radiation may be another useful quantity for distinguishing the ballistic from the diffusive regime. To extract this spectral evolution, we perform a Hilbert transformation on the measured waveforms to obtain a complex representation of $E(t)$:

$$E(t) = a(t)e^{-j\phi(t)},$$

(5-7)

where $a(t)$ is the real, positive amplitude function and $\phi(t)$ is the temporal phase function. This representation is known as the analytic signal. It has the property that the
negative frequencies have zero amplitude. The angle of this complex quantity is the temporal phase \( \phi(t) \), which is related to the instantaneous frequency by

\[
\nu_{\text{inst}}(t) = \frac{1}{2\pi} \frac{d\phi}{dt}.
\] (5-8)

We compute the average phase function for both the co-polarized and cross-polarized waveforms, \( \langle \phi_\parallel(t) \rangle \) and \( \langle \phi_\perp(t) \rangle \), by configurationally averaging the phase functions of all the individual waveforms. The results are shown in Fig. 5.8.

These data show several interesting features. First, the phase functions are both slightly non-linear with a positive chirp, indicating that, on average, the later portions of the waveforms have a higher average frequency. This can be qualitatively understood
Figure 5.9: The difference between the two phase functions. The difference increases roughly linearly after 650 ps. The slope of this line indicates the difference in the average instantaneous frequency, $\Delta v_{\text{inst}}$, between the two orthogonal polarization components.

from the frequency-dependence of the mean free path [74]; since the higher frequency components scatter much more readily, it is expected that these would have longer average paths in the medium, and therefore emerge later. Second, $\langle \phi_\perp(t) \rangle$ is essentially equal to $\langle \phi_\parallel(t) \rangle$ up to roughly $t = 650$ ps, after which it increases more rapidly. The larger slope indicates a higher average frequency for the perpendicular component. This is also consistent with the fact that the higher frequency components have shorter mean free paths. Since the perpendicular component must have experienced (on average) more scattering events, it is reasonable to expect that it is comprised of those photons that are more likely to scatter. Figure 5.9 shows the difference $\langle \phi_\perp(t) \rangle - \langle \phi_\parallel(t) \rangle$, which is nearly zero up to 650 ps and then shows a roughly linear increase. The slope of this line gives
the average frequency difference \(\sim 45\) GHz between the two perpendicular field components at these later times, when the propagation is completely diffusive. Such spectral shifts are not unknown in multiple scattering [140], but this is the first demonstration to our knowledge of a polarization-dependent frequency shift.

We discuss the window of time during which \(\langle \phi_\parallel(t) \rangle = \langle \phi_\perp(t) \rangle\). Prior to 650 ps, both polarizations contain the same average spectral content, which suggests that the mean number of scattering events is equivalent. It is important to note that this regime extends about 50 ps beyond the single scattering regime illustrated in Figs. 5.7 and 5.9. Thus, there is a time window during which no single-scattered photons emerge from the medium (and polarization memory is lost), but at the same time the frequency-dependence of the mean free path has not yet given rise to any substantial spectral shifts of the type described above. Unfortunately, signal-to-noise limits the characterization of this intermediate regime; however, it may provide a means to distinguish between photons that have scattered once from those that have scattered a few times (e.g., snake photons). Future work would involve further exploring this transition regime from single scatter to diffuse photon arrivals.

5.6 Summary

We find that there exists a spectral shift between the two polarizations at later times when the photons are propagating diffusively. Although this shift is small (\(\sim 45\) GHz), it is still measurable. This description is only applicable to broadband radiation, where an instantaneous frequency can be clearly defined. We also observe that intensity correlations exist between the polarizations of single scatter and snake photons, which are absent for diffuse photons. For the broadband case, it seems more reasonable to
characterize the single scatter and snake photons using spectral content and intensity correlations rather than polarization contrast. Further exploration of the spectral differences between the polarizations of diffusive photons could lead to novel approaches to imaging objects embedded in random media.

Limitations of signal-to-noise and bandwidth prevented us from investigating the intermediate regime between the single-scatter and diffuse photon arrival times. However, this regime has interesting applications to imaging objects in clutter, because single-scatter photons contain information only about the object of interest, whereas both the object and the surrounding clutter influence snake photons. Distinguishing the single-scatter photons from the snake photons will improve image quality. More work needs to be done characterizing the transition region between arrivals of the single-scatter and diffuse photons.

In conclusion, we have observed that, in the diffusive regime, the average spectral content of the two orthogonal polarization components are distinct. This divergence occurs after the regime of single-scattering, where both the degree of polarization and the intensity correlation have vanished, and is therefore a feature of diffusive broadband radiation. It arises from the frequency-dependence of the scattering parameters such as the mean free path, which vary strongly within the bandwidth of the incident radiation. We note that this frequency shift goes hand in hand with the delayed emergence of the perpendicular component of the radiation, which gives rise to the variation of $P(t)$ – both are a consequence of the larger number of scattering events experienced, on average, by the perpendicular component.
Chapter 6
Discussion and Future Work

6.1 Summary of Results

We present the first set of experiments that investigate the multiple scattering of THz radiation. The topic of electromagnetic wave propagation through scattering media has previously been studied with microwaves, infrared light, and visible light. Until recently, a lack of efficient generation techniques limited the exploration of scattering in the far infrared regime. The THz time-domain spectrometer provides a very powerful tool to investigate wave scattering. With its broad spectral coverage and ability to measure both the phase and intensity of the radiation, each sample measurement yielded a plethora of information. The majority of previous scattering studies employed narrowband sources, where the parameters of the medium, such as the mean free path, are constant throughout the bandwidth of the source. We investigated scattering in the extreme situation of very broadband radiation, which had remained largely unaddressed.

We acquired the first measurements of diffuse T-rays and studied the statistics of the electric field. Narrowband theory predicted that the real and imaginary parts of the electric field follow a Gaussian distribution. However, our experiment revealed that the distribution for broadband radiation differs substantially from Gaussian statistics. By extending the narrowband theory to the broadband case, we demonstrated that the distribution was in fact a superposition of Gaussian distributions with zero means and frequency dependent variances. Furthermore, we extended the distributions for the intensity and phase derivative to incorporate broadband radiation.
Many researchers have applied the diffusion equation to describe the time-of-flight (TOF) distribution of a short pulse propagating through a random medium. However, we found that our measurements of the TOF distribution deviated from the diffusion equation description, because our experiment geometry and bandwidth violated the assumptions made with the diffusion model. Most importantly, the diffusion model assumes that the mean free path is constant over the bandwidth of the source, which is clearly not the case in our experiment. This illustrates the need to further model the broadband case of scattering.

Our measurements of the polarization yielded some surprising results. At late delay times, when the photons are propagating diffusively, we observed a spectral shift between the co-polarized and cross-polarized radiation. This corresponded to late-arriving photons, after the degree of polarization and intensity correlations decayed to zero. At early times, when there was a substantial degree of polarization and the photons had only scattered a few times, there was no spectral shift present. This suggested that the spectral shift between the polarizations provides another signature of diffuse photons, which is a distinct feature of broadband radiation. This signature may be an alternative marker to polarization contrast or polarization gating for imaging applications.

In this thesis, we have investigated the subject of multiple scattering in random media using THz time-domain spectroscopy. This new technology enabled us to examine the scattered electric field directly, as opposed to current optical measurements, which are limited to time-averaged intensity quantities. Also, our measurements were performed in a 3-dimensional sample, rather than a waveguide geometry customarily employed with microwaves. These experiments demonstrated that the THz time-domain
spectrometer is well suited for probing random media and has opened the door to further our understanding of scattering.

6.2 Future Research Directions

These experiments were the first steps toward understanding THz pulse propagation through random media, but there are still many questions remaining and new research directions to explore. In this work, we have made some new observations regarding wave propagation through random media and have attempted to provide some possible explanations to our results. However, for many of our observations, we lack a complete theoretical model. For example, we observed that the diffusion model failed to predict diffusive photon transport for broadband radiation. The majority of theoretical models that describe wave propagation through random media are tailored for narrowband radiation and do not adequately address the broadband case. Our data demands a better theory in order to fully model it. With the advent of powerful processors and efficient algorithms, finite element methods (FEM) and finite difference time-domain (FDTD) techniques have been applied to physics problems with great success. An interesting followup to our work would be to apply these techniques to model THz scattering experiments. This would allow us to interpret our results at a greater precision and understand the mechanisms that are responsible for our observations. An accurate model would predict the frequency dependent intensities, which control the statistics of the diffuse radiation. Also, it would be interesting to characterize the measured spectral shift between the polarizations as this phenomenon is observable only with broadband radiation.
The microwave and seismic communities have considerable interest in imaging through clutter. Scattering of seismic waves from inhomogeneities within the rock or scattering of microwaves from trees severely degrade image quality. Imaging with THz radiation is a topic of considerable interest, because of its short wavelengths and unique material responses. However, to our knowledge, imaging with THz radiation through clutter has never been investigated. Such an investigation may provide insights to the microwave and seismic communities as well as improve on current THz imaging techniques.

The photon analogy to Anderson localization of electrons has been observed with microwaves [9] and visible light [11], but not at THz frequencies. A precursor to Anderson localization is the coherent backscattering, which is the constructive interference in the backward direction as a result of multiple scattering. There have been several experiments that have measured this effect [69, 112]. This would be interesting to study with THz radiation, because unlike visible lights, both intensity and phase are accessible. Also, due to the broadband nature of the radiation, certain portions of the spectral range would scatter more than others, which would allow for the mean free path dependence of the effect to be characterized.

Future research would investigate the coherent backscattering effect with THz radiation by constructing random media consisting of higher index scatterers, which have larger scattering cross-sections, and measuring the backscattered radiation. Figure 6.1 shows a schematic of the proposed coherent backscattering experiment. The random medium could consist of a high index powder such as silicon ($n = 3.418$). The transmitter would propagate THz pulses into the random medium. The beam splitter
Figure 6.1: The proposed backscattering experiment. THz pulses are propagated into a strongly scattering medium. A beam splitter directs the backscattered radiation to the detector. The detector is placed on a rotating cantilever arm to measure the angular profile of the radiation.

would direct the radiation toward the detector, which would be placed on a cantilever arm in order to measure the scattered radiation at multiple angles. From the electric field measurements, it would be possible to study the correlations between the fields at different backscatter angles. This experiment would provide insights into the influence of the coherent interference of multiple scattering to the electric field and potentially the observation of photon localization.

There are many more possibilities for future research of THz scattering. We have attempted the initial steps to explore this subject. The THz time-domain spectrometer has proven to be a powerful investigational tool, which is able to aid both experimental and theoretical work. Because of its broad capabilities, there is great potential for future significant contributions by this tool to the field of scattering.
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