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**The Propagation of Single-Cycle
Terahertz Pulses in Random Media**

by

Jeremiah Glen Pearce

**A THESIS SUBMITTED
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE**

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The Propagation of Single-Cycle Terahertz Pulses in Random Media

Jeremiah Glen Pearce

Abstract

We describe what are to our knowledge the first measurements of the propagation of coherent, single-cycle pulses of terahertz radiation in a scattering medium. We measure the propagation constants for pulses in a dense collection of spherical scatterers, and compare to the predictions of the quasi-crystalline approximation. Even though the fractional volume in our measurements exceeds the limit of validity of this model, we find that it still predicts certain features of the propagation with reasonable accuracy. By measuring the transmission as a function of the length L of the medium, we extract the scattering mean free path $\lambda_{sc}(\omega)$ over a broad bandwidth. We observe variations in λ_{sc} ranging over nearly two orders of magnitude, and covering the entire thin sample regime from $L/\lambda_{sc} \ll 1$ to $L/\lambda_{sc} \sim 10$.

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Chapter 1: Introduction

1.1 Wave Scattering

Why is the sky blue? Although this question is subject to much philosophical debate, a physical understanding can be found by investigating the propagation of waves through an inhomogeneous medium. The sky appears blue because of the wavelength-dependent scattering of visible light by molecules in the air (Kerker 1969). Light is an example of an electromagnetic wave; other examples of waves include acoustical waves and electrons. The study of the scattering of light and other types of waves has been a challenging research area for many years and extends over several different disciplines in science and medicine. In everyday life, the effects of scattered waves can be observed. The blinding glare from headlights being shone into fog is due to the scattering of light off of the water particles in the air. White paint, which consists of high-dielectric-constant particles embedded in a low-dielectric medium, appears white because of visible light of all wavelengths being scattered by the dielectric particles in the paint. Although the scattering of waves is a very common phenomenon in the world, it is very complicated and very difficult to model. This complexity leads to a wealth of unusual and interesting phenomena.

The propagation of waves through a homogeneous medium has been well modeled by Schrödinger's equation (for quantum mechanical waves) or the electromagnetic wave equation (for light and other EM waves) (Sheng 1995). The transport properties of light and other waves through homogeneous medium, such as the velocity, are well known and can be easily calculated. Even in inhomogeneous media, if

the inhomogeneity is spatially periodic, then the behavior can be understood with fairly simple models. However, in many realistic situations the medium is not homogeneous and there is a degree of disorder. Many of the most difficult, but interesting problems in characterizing waves arise when the system is extremely disordered. Anderson investigated the scattering of electrons in disordered crystalline solids (Anderson 1958). He demonstrated that the scattering of electrons increases suddenly as the disorder is increased; the electron waves localize and the material behaves as an insulator. Localization is a wave phenomenon, which could occur for other types of propagating waves. Although, localization has only been observed for microwaves, the observation of it for visible light is a goal for many researchers in the field (John 1984; Wiersma, Bartolini et al. 1997).

Despite the many years of research and collaborations from different disciplines of science, this is a topic of interest in which we have much to learn. The demand for higher quality images from the biomedical community, the exploration of the earth's crust, and the desire for an improvement of wireless communications motivates this research area. Using a recent technology known as terahertz time-domain spectroscopy, we can experimentally measure the different transport properties of electromagnetic waves in a strongly scattering medium. This technique has several advantages over predecessors in the field, because of the ability to measure the electric field directly and the ability to transmit and measure a broad spectrum of radiation, spanning more than one order of magnitude in wavelength. From these measurements, we hope to gain a better understanding of the phenomenon of wave scattering.

1.2 Applications

The propagation of waves in an inhomogeneous medium leads to multiple scattering events, and as a result can be extremely difficult to model. However, it is a very important problem in many areas of research. In the field of geophysical prospecting, for example, the information that can be extracted from seismic imaging is limited in part by the strong scattering of acoustic waves as they propagate in the earth (Dobrin and Savit 1988). Another example can be found in biomedical optics (O'Leary, Boas et al. 1996). For near infrared wavelengths of light, the human body does not absorb too heavily, but the light scatters strongly as it propagates. In these and other examples, a good understanding of the statistical properties of the scattered radiation can be used to form images of objects buried within the scattering medium. Finally, in the limit of very strong scattering, waves can become completely localized. The study of localized states of light has been a compelling research goal for many years (Legendijk 1994).

In the biomedical community, there has been an increase in demand for noninvasive exploratory procedures. For example, recent literature suggests using backscattered light to create images, in order to detect skin cancer (Gurjar, Backman et al. 2001). In a recent paper (Jacques, Roman et al. 2000), a basic polarized light camera system is described, shown in Fig. 1.1. An incoherent light source illuminates the skin with white light at a small angle. A polarizing filter linearly polarizes the white light. A glass optical flat is pressed against the skin to enforce a flat skin surface in order to reduce the glare from reflecting photons. Two images are captured by the CCD camera, one consisting of parallel polarized light and one consisting of perpendicular polarized light. Each image has an undesired "noise" component from the multiple scattered light.

Figure 1.1: Basic polarized light camera system (Jacques, Roman et al. 2000), The CCD captures two images, one for each polarization, by using the polarization filter to select either perpendicular or parallel light.

The polarization of this light is completely randomized and is distributed the same in both images. Therefore, the “noise” component is added to both images. In order to correct for this, the perpendicular-polarized image is subtracted from the parallel-polarized image. This technique allows for the imaging of the superficial layers of the skin, which is where cancer commonly arises.

An issue that is not addressed in the paper is the information contained in the randomly polarized light. It is possible that the randomly polarized light could be used to create images below the superficial layer of skin. Much work has been done investigating diffuse photon density waves, which are waves created by light that has been scattered multiple times (Li, Durduran et al. 1997). Using these diffuse photon density waves, objects that have been embedded in a low absorbing strongly scattering media have been imaged successfully. Much work is being done to understand the complexity of diffuse light (Ripoll and Nieto-Vesperinas 1999; Zaccanti, Alianellis et al. 1999). For instance, there are limitations on the effectiveness of diffusion theory in the

Figure 1.2: The 94 GHz rain scattering experimental setup (Gloaguen and Lavergnat 1996).

vicinity of the light source (Graaff and Ten Bosch 2000). There are many research areas and applications in the biomedical community that would benefit from a better understanding of the transport properties of scattered light.

At longer wavelengths, meteorological effects such as rain, snow, and hail can have a significant impact on the propagation of waves with frequencies above 1 GHz (Ishimaru 1978). A topic in research for workers in the field of antenna theory is how to compensate for the scattering from different types of weather for specific applications. One of the primary issues with rain is that the scattering properties for a wave at a given frequency are dependent upon the size and shape of the water droplets. The particle size and shape and therefore the scattering cross-section change as a function of precipitation rate. This is a problem in wireless communications where there is a demand to transfer information through the air over a high quality channel.

As an example of research in this area, one group investigated the effects of the scattering of 94 GHz waves in rain (Gloaguen and Lavergnat 1996). The objective was

Figure 1.3: Diagram of Ground-Penetrating Radar being used in the reflection-profiling mode on soil over bedrock (Davis and Annan 1989).

to determine the significance of the effects of multiple scattering by randomly distributed raindrops. This was accomplished by installing a receiver and a transmitter 312 m apart from each other as shown in Fig. 1.2. Temperature and humidity sensors were used to measure the temperature, rainfall rate, and rainfall size distributions. The resulting measurements of the receiver power were compared to theory. This work demonstrated the significance of multiple scattering of the propagating wave. There are many issues that arise in the field of antenna theory from the scattering of particles of the atmosphere and there is much work continuing being done on understanding the propagation of electromagnetic waves in scattering media.

Yet another example can be found in the field of geophysical prospecting. Here, one of the primary objectives is to be able to create subsurface images. A current technique is to use Ground-Penetrating Radar (GPR) (Davis and Annan 1989), shown in Fig. 1.3, to form images of the earth. The radar transmits a short pulse of high frequency (10-1000 MHz) electromagnetic energy into the ground. The pulse returns at different times depending on the different electrical properties of the geological strata. Different materials cause the pulses to travel at different velocities. The water content of the soil is

the most important factor in determining the electrical properties of the geological materials. The pulses received at the surface are used to calculate the different dielectric constants of the materials encountered by the propagated pulse. From those constants, a high-resolution image of the soil can be formed. Another technique used for seismic exploration to determine the composition of the earth involves propagating acoustic waves into earth and then measuring the reflections at the surface (Clay 1990). Scattering can adversely effect the accuracy in which the composition of the earth can be determined (Fokkema and van den Berg 1993).

In geophysical prospecting, scattered waves can sometimes create inaccuracies in parameter estimation, but it also can be used to locate objects embedded in the ground. For example, a group recently used the generalized Radon transform to perform GPR imaging (Wang and Oristaglio 2000). Whereas the Radon transform uses a set of line integrals over a plane in space to project a two dimensional object onto one dimension, the generalized Radon transform performs a set of line integrals over a general surface in space. The researches buried two plastic pipes and two metal pipes underground and took a set of GPR waveforms to measure the waveforms that were scattered from the pipes. They constructed the images by inverting the generalized Radon transform. Their results were very good for the plastic pipes, which scatter weakly, but the images of the metal pipes were limited due to the higher conductivity and the stronger scattering. There is still much work to be done in this area in order to create higher quality images. A better understanding of wave propagation in an homogeneous medium such as the ground is needed to further the research of this area in geophysics.

It is apparent that there are many different disciplines of science and medicine, which would benefit from more knowledge of wave scattering. The purpose of this thesis is to experimentally investigate the transport properties of light in an inhomogeneous medium. This may be regarded as a model for the general topic of wave propagation in complex media. The results should therefore inform many of these other fields. We employ a newly developed technique known as terahertz time-domain spectroscopy, which provides us with a number of distinct advantages in the study of scattering phenomena.

1.3 Terahertz Time-Domain Spectroscopy

Using terahertz time-domain spectroscopy, it is possible that a better model for wave scattering could be developed. The recently developed system for transmitting and detecting terahertz pulses gives several advantages over other imaging systems in that it retains the phase information of the pulse and provides extremely high temporal resolution (Mittleman, Jacobsen et al. 1996; Mittleman, Gupta et al. 1999). Also, common materials exist with a wide range of properties in the terahertz regime, so model samples are easily fabricated. In this thesis, we study the propagation of few-cycle wave packets in scattering medium by measuring terahertz light pulses propagating through a scattering medium generated by a terahertz time-domain spectrometer.

A terahertz time-domain spectrometer has the ability to generate broadband terahertz pulses ranging from 100 GHz to 1.5 THz., which corresponds to wavelengths from 0.2 mm to 3 mm. Pictured in Fig. 1.4 is the spectrum of a generated pulse. One of the unique features about the terahertz regime of the electromagnetic spectrum is the spectral response of different materials. Materials such as plastic, Styrofoam, and

Figure 1.4: The frequency bandwidth of a single THz pulse. The water absorption lines are marked with arrows.

cardboard are virtually transparent to terahertz radiation. Metals such as aluminum are very strong reflectors. Water absorbs very heavily in the terahertz regime; the absorption lines appear in Fig. 1.4 (van Exter, Fattinger et al. 1989). At larger wavelengths compared to visible light, models can easily be produced with scatterers that have sizes ranging from much smaller to much larger than the wavelength.

We have the ability to construct models of the experiments done in other disciplines using the terahertz time-domain spectrometer in order to reproduce their results with several advantages. For example, the group using the Ground-Penetrating Radar in order to construct images of the earth cannot practically dig up the entire earth to test their theories. However, we can easily create a similar experiment where all the

parameters of the model are well known and test the theories of the geophysicists. With the unique properties that materials exhibit in the terahertz regime, many experiments can be conceived in order to observe the transport properties of scattered waves.

In this thesis, we describe the first such measurement. This involves propagating THz pulses through slabs of randomly distributed scatterers. From the waveforms we collect, we can measure many of the parameters associated with a scattering medium, such as the mean free path and the effective index of refraction. We compare the results to existing theories and other experimental results.

1.4 Thesis Overview

In chapter 2, the different concepts and theories on wave scattering will be introduced as well as an extensive literature review of previous experiments. The terahertz time-domain spectrometer will be discussed in chapter 3. Details on how terahertz pulses are generated, some of the previous applications with the tool, and some of the advantages over other imaging tools will be shown. In chapter 4, the wave scattering experiment will be described. Finally, in chapter 5, conclusions will be presented and suggestions for future work will be made.

Chapter 2: Background

2.1 Single Scattering Case

The propagation of waves through a multiple scattering medium is a very complicated phenomenon to model. It is important that we first understand the scattering of a wave off of a single particle. Ishimaru presented a simple formalism for the scattering of wave by a single particle (Ishimaru 1978). Consider a linearly polarized electromagnetic plane wave (Hecht 1987) propagating through vacuum ($n_{med} = 1$) with the electric field given by:

$$\hat{E}_i(\hat{r}) = \hat{e}_i \exp(ik\hat{i} \cdot \hat{r}). \quad (2-1)$$

The wave vector $\hat{i}$ points in the direction of propagation and has unit magnitude, k is the wave number which equals $2\pi / \lambda$, where λ is the wavelength of the wave, $\hat{e}_i$ is a unit vector in the direction of polarization, and $\hat{r}$ is the position in space.

As shown in Fig. 2.1, this wave encounters a particle with a dielectric constant given by:

$$\epsilon_r(\hat{r}) = \epsilon'_r(\hat{r}) + i\epsilon''_r(\hat{r}). \quad (2-2)$$

$\epsilon_r(\hat{r})$ is the complex dielectric constant as a function of position. The complex refractive index of the particle is related to the complex dielectric constant by $n_r(\hat{r}) = \sqrt{\epsilon_r(\hat{r})}$. 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. The incident wave encounters a dielectric particle, the particle scatters the wave, and a scattered electric field $\hat{E}_s(\hat{r})$ is

Figure 2.1: A plane wave $\hat{E}_i(\hat{r})$ encounters a particle with a complex dielectric constant $\epsilon_r(\hat{r})$ produces a scattered field $\hat{E}_s(\hat{r})$ in the direction $\hat{\theta}$.

generated, propagating in a direction $\hat{\theta}$. The electric field at a distance R along the vector $\hat{\theta}$ contains contributions from the incident field $\hat{E}_i(\hat{r})$ and the scattered field $\hat{E}_s(\hat{r})$. In the near field of the particle, the scattered field is very complicated because it has interference effects from different areas of the particle. At a distance $R > D^2/\lambda$, where D is a dimension of the particle such as a diameter, these interference effects can be ignored and $\hat{E}_s(\hat{r})$ is considered a spherical wave given by:

$$\hat{E}_s(\hat{r}) = \hat{f}(\hat{\theta}, \hat{i})(\exp(ikR)/R). \quad (2-3)$$

$\hat{f}(\hat{\theta}, \hat{i})$ is called the scattering amplitude and contains all the amplitude, phase, and polarization information of the scattered electric field propagating in a direction of $\hat{\theta}$, illuminated by an incident plane wave propagating in a direction $\hat{i}$.

At this point we have enough information to define the cross-sections related to scattering. There are three cross-sections of interests: the scattering cross-section σ_s , the absorption cross-section σ_a , the total cross-section σ_t . They are related to each other by:

$$\sigma_t = \sigma_s + \sigma_a. \quad (2-4)$$

Figure 2.2: The Mie Theory solution for the scattering cross-section of a sphere with a radius of 0.397 mm and a dielectric constant of 1.433. Note the complexity of the cross section consisting of multiple resonances.

The scattering cross-section is related to the total scattering probability and is given by:

$$\sigma_s = \int_{4\pi} |\hat{f}(\hat{\mathbf{o}}, \hat{\mathbf{i}})|^2 d\omega. \quad (2-5)$$

The absorption cross-section is the probability for energy to be deposited in the scatterer, and is therefore related to ϵ_r'' according to:

$$\sigma_a = \int_V k\epsilon_r''(\hat{r}) |\hat{E}(\hat{r})|^2 dV', \quad (2-6)$$

where the integral is over the volume V of the particle. The total cross-section is comprised of the total power loss by scattering and the total power loss by absorption.

The forward scattering theorem relates the total cross-section σ_t to the imaginary part of the scattering amplitude $\hat{f}(\hat{\mathbf{o}}, \hat{\mathbf{i}})$ in the forward direction by:

$$\sigma_i = (4\pi/k) \text{Im}[\hat{f}(\hat{i}, \hat{i})] \cdot \hat{e}_i. \quad (2-7)$$

It appears that solving for these cross-sections, which are directly related to the scattering probabilities, is fairly trivial. This is certainly not the case. The scattering amplitude $\hat{f}(\hat{0}, \hat{i})$ function is very complicated to determine. It is dependent on the electric field inside the dielectric particle, which is typically not a known function. These equations give us a basis to begin to understand the scattering of a propagating wave.

There are certain situations where the scattering amplitude function can be determined. If it is known that the dielectric particle is a homogeneous sphere an exact solution to the scattering amplitude function exists and is provided by Mie Theory (Kerker 1969). The solution, albeit a rigorous one, allows for the total, scattering, and absorption cross-sections to be calculated given the radius of the sphere, the dielectric constant of the sphere, the dielectric constant of the medium, and the spectral range of the incident light. Experiments have demonstrated that the Mie solution accurately describes the scattering from a sphere (Mark 1990). In Fig. 2.2, the scattering cross-section of a sphere (in air) with radius 0.397 mm and an index of refraction 1.4333 is shown for frequencies below 2 THz. The Mie solution is very complex and is comprised of many resonances. Solutions exist for particles of various shapes; however, their solutions are even more complicated than for the sphere. Often, when one wants to estimate parameters for the scattering of a non-spherical particle or a group of non-spherical particles, Monte Carlo methods are used (Mishchenko, Hovenier et al. 2000). Monte Carlo methods use random walks of photons through an inhomogeneous medium to obtain many realizations and calculate averages of various parameters, such as photon

Figure 2.3: A slab of scatterers embedded into a homogeneous medium. The scatterers have a refractive index different than the surrounding medium. Light propagates through a finite distance L of the material and then exits. The x and y dimensions of the slab are considered to be infinite.

travel time and attenuation. Other approaches include the T-matrix formalism, which is also numerically cumbersome.

The Mie solution gives us insight to the complexity of wave scattering. Understanding the scattering by a single dielectric particle is a very interesting and challenging problem. Analytical solutions exist for the scattering amplitude functions of homogeneous spheres from Mie Theory. The total, scattering, and absorption cross-sections describes the power loss from the incident wave and are the primary parameters of interest when discussing single scattering. The scattering by a single particle provides a basis to understanding the propagation of multiply scattered waves.

2.2 Multiple Scattering Case

Mie theory gives an exact solution for the scattering by a single homogenous sphere, however it does not take into account multiple scattered light. Consider a case, as shown in Fig. 2.3, where many such spheres are embedded into a slab of a homogeneous

Figure 2.4: The transmission coefficient of light propagating through a slab of scatterers of arbitrary finite length L (Mark 1990).

material with a contrast in index of refraction to that of the background material. The x and y dimensions can be considered infinite, and the z dimension has an arbitrary finite length L . Using the results from Mie theory and taking into account particle correlations, a formalism for the multiple scattering case is derived.

There are two primary variables of importance when constructing a scattering media, the length of the medium L and the volume fraction f . First, consider a case where the volume fraction of the particles is fixed and the length of the medium varies. When considering the propagation of light through a slab of a scattering material, there are three regimes of scattering that are encountered as L increases: the ballistic scattering regime, the diffusion regime, and the absorption regime (Mark 1990). Fig. 2.4 shows a heuristic of the transmission coefficient as a function of arbitrary slab thickness L . The transmission coefficient initially decays exponentially with a decay rate of λ_{sc} , the scattering mean free path, which is the average distance between scattering events. A

physical explanation to as why the transmission rate decays exponentially is due to photons being scattered once and then propagating in a direction which is not parallel to the incident light. This is known as the ballistic scattering regime. The second region in Fig. 2.4 is known as the diffusion regime, which has a decay rate of $1/(L + L_0)$ that is much slower than an exponential decay. The decrease in decay rate of the transmission coefficient can be explained as photons scattering multiple times, but some eventually being redirected in the same direction as the incident light. The last region is the absorption regime where the effect of absorption of the spheres leads to an exponential decay of the transmission coefficient, due to the long path length for the light in the medium. These three regimes describe the transmission of light being scattered by an inhomogeneous slab.

Consider an incoming wave $\hat{E}_i(\hat{r})$ encountering a slab of a non-absorbing inhomogeneous material as pictured in Fig. 2.3. Assume L is within the ballistic scattering regime and assume that the medium has a low volume fraction of particles. A scattered wave $\hat{E}_s(\hat{r})$ propagating in the same direction as the incident wave can be related to the incident wave by:

$$\hat{E}_s(\hat{r}) = \hat{E}_i(\hat{r}) \exp(-j\omega n_{sc} L / c) \quad (2-8)$$

where ω is the frequency of the wave, c is the speed of the wave in vacuum, L is the length of the slab, and n_{sc} is the effective complex scattering index of refraction of the inhomogeneous slab.

$$n_{sc} = n_r - jn_i \quad (2-9)$$

From the imaginary part of the scattering index of refraction n_i , the scattering mean free path λ_{sc} can be obtained by:

$$\lambda_{sc} = \frac{c}{2n_i\omega L}. \quad (2-10)$$

The scattering mean free path λ_{sc} describes the average distance between scattering events, and is related to the scattering cross-section by:

$$\lambda_{sc} = \frac{1}{n_0\sigma_{sc}} \quad (2-11)$$

where n_0 is the number density of scatterers and σ_{sc} is the scattering cross-section as defined by equation (2-5) (Mark 1990). 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 λ_{sc} , but the transport mean free path $\lambda_r = \lambda_{sc} / (1 - \langle \cos \theta \rangle)$, where $\langle \cos \theta \rangle$ is the average cosine of the angle of scattering (Ishimaru 1978). Due to the anisotropic scattering, it takes several scattering events to completely lose memory of the original direction. The distance travelled to lose this directionality is the transport mean free path. Mie Theory can be used to calculate σ_{sc} ; however, this is only applicable for situations where the volume fraction is low $< 1\%$.

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} . There are several methods to theoretically calculate n_{avg} , one of which is volume weighted averaging given by:

$$n_{avg} = \text{Re}\{\sqrt{\epsilon_s f + \epsilon(1-f)}\} \quad (2-12)$$

where ϵ_s is the permittivity of the sphere, ϵ is the background permittivity, and $f = n_0 v_0$ is the volume fraction of the spheres occupying the volume of the slab. Another formula to estimate the effective index of refraction is called the Maxwell-Garnett mixing formula (Maxwell-Garnett 1904). It begins by considering dipoles being induced within the spheres by an exciting electric field. When dipoles are induced, the spheres become polarized and produce an electric field in the opposite direction of the exciting field. The macroscopic field can be related to the exciting field by

$$\hat{E}^e = \hat{E} + \hat{P} / 3\epsilon \quad (2-13)$$

where $\hat{E}^e$ is the exciting field, $\hat{E}$ is the macroscopic field, and $\hat{P} = n_0 \hat{p}$ is the polarization per unit volume. $\hat{p} = \alpha \hat{E}^e$ is the induced dipole moment where $\alpha = v_0 3(\epsilon_s - \epsilon) \epsilon / (\epsilon_s + 2\epsilon)$ is the polarizability of a sphere and v_0 is the volume of one sphere. The polarization $\hat{P}$ can be related to the macroscopic electric field $\hat{E}$ by:

$$\hat{P} = \frac{n_0 \alpha \hat{E}}{1 - (n_0 \alpha / 3\epsilon)}. \quad (2-14)$$

Using the fact that the displacement is $\hat{D} = \epsilon \hat{E} + \hat{P}$ and that $\hat{D}$ can be related to the macroscopic electric field by $\hat{D} = \epsilon_{eff} \hat{E}$, the effective permittivity can be calculated as:

$$\epsilon_{eff} = \epsilon \left[\frac{1 + (2n_0 \alpha / 3\epsilon)}{1 - (n_0 \alpha / 3\epsilon)} \right]. \quad (2-15)$$

From equation (2-15), the effective permittivity can be simplified to:

$$\epsilon_{eff} = \epsilon \frac{1 + 2fy}{1 - fy} \quad (2-16)$$

where

$$y = \frac{\epsilon_s - \epsilon}{\epsilon_s + 2\epsilon}. \quad (2-17)$$

The average index refraction is simply $n_{avg} = \text{Re}\{\sqrt{\epsilon_{eff}}\}$. Equation (2-16) is known as the Maxwell-Garnett mixing formula. From this simple model of scattering, many useful parameters can be calculated. However, its applicability is also limited to small values of f and also $\lambda \gg$ size of the particle.

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 the simple mixing rates. These models are generally based on the multiple scattering equations, a system of equations relating the field incident on a particular scatterer to the fields arriving from all other scatterers. Tsang, Kong, and Shin provide a simple explanation of the multiple scattering equations (Tsang, Kong et al. 1985). Consider a situation where there are N randomly distributed scatterers contained in a volume V similar to Fig. 2.3. An incident electric field $\hat{E}_{inc}$ is applied to the volume. The resulting macroscopic electric field $\hat{E}$ encompasses contributions from each of the scattered fields from the particles $\hat{E}_{sj}$ and the incident field $\hat{E}_{inc}$ given by:

$$\hat{E} = \hat{E}_{inc} + \sum_{j=1}^N \hat{E}_{sj}. \quad (2-18)$$

For a single particle j , there is an exciting field $\hat{E}_j^E$ that is the exciting field of $\hat{E}_{sj}$ in the absence of all other particles. $\hat{E}_j^E$ and $\hat{E}_{sj}$ are related to each other by a transition operator T_j , which is assumed to be known and is a function of the shape and size of the particles and the wave vector.

$$\hat{E}_{sj} = T_j E_j^E \quad (2-19)$$

The exciting field $\hat{E}_j^E$ contains contributions from all of the particles. $\hat{E}_j^E$ can also be expressed in terms of $\hat{E}$ and $\hat{E}_{sj}$ by:

$$\hat{E}_j^E = \hat{E} - \hat{E}_{sj}. \quad (2-20)$$

Combining equations (2-18), (2-19), and (2-20), an expression for the exciting electric field of a single particle can be given by:

$$\hat{E}_j^E = \hat{E}_{inc} + \sum_{\substack{l=1 \\ l \neq j}}^N T_l \hat{E}_l^E. \quad (2-21)$$

The exciting electric field can now be calculated, because equation (2-21) consists of N equations with N unknowns. The scattered fields can also be calculated by equation (2-19). A more rigorous derivation of this system of equations can be shown using Green's function or T-Matrix formalism (Tsang, Kong et al. 1985).

An assumption above is that the configuration of the scatterers (and therefore T_j) was known. However, this is rarely the case. In order to calculate the propagation constants, the equations of (2-21) need to be averaged over all of the 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. The averaged Green's function is expressed 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 simplest solution consists of truncating this hierarchy at lowest order. This is known as the effective field approximation (EFA), also known as Foldy's approximation, and is equivalent to neglecting all correlations among the locations of scatterers (Foldy 1945). The EFA is usually only valid for small particle densities $\sim 1\%$. For higher volume fractions, below $\sim 40\%$, 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 (Tsang, Kong et al. 1985). 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 (West, Gibbs et al. 1994). For spherical scatterers, the Percus-Yevick pair distribution function provides an adequate description of the positional correlation (Tsang, Kong et al. 1982). 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 this high density regime are numerical, such as Monte Carlo methods. In fact, there is also a paucity of experimental work at fractional volumes over 40%.

2.3 Strong Scattering 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 many defects. The

Figure 2.5: Anderson localization in disordered systems. Point A is a light source in the medium. A random light path can be traversed in two directions. Two waves that traverse in opposite directions will acquire the same phase and add constructively.

scattering strength can reach a point where the diffusion regime breaks down and electrons no longer transport. The material behaves like an insulator. Electrons and photons behave very similarly since they are both waves and it is believed that similar effects occur for light. A characteristic of localization is that the diffusion regime is expected to transition from a L^{-1} to an exponential decay, as a function of slab thickness (Wiersma, Bartolini et al. 1997). From Figure 2.4, it is observed that absorption also appears as an exponential decay; therefore it is difficult to distinguish the effects of absorption and localization. The Ioffe-Regel criterion determines that localization will occur for $k\lambda_{sc} \leq 1$ (Mott 1974), where k is the magnitude of the wave vector and λ_{sc} is the mean free path. The wave vector is related to the wavelength by $k = 2\pi/\lambda$, where λ is the wavelength. The Ioffe-Regel criteria then becomes $2\pi\lambda_{sc}/\lambda \leq 1$, which implies that the wave is scattered before it can even perform one oscillation of the electric field and is scattered after it travels a fraction of the wavelength.

One research group observed $kl = 1.5$, which is the smallest value measured for near infrared light (Wiersma, Bartolini et al. 1997). 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. There was a question on whether their results were due solely to scattering or if their results are the effects of absorption introduced from the grinding process (Scheffold, Lenke et al. 1999). They responded by providing a strong argument against the significance of the absorption and concluded that their results are indeed the localization of light (Wiersma, Rivas et al. 1999). Another group followed up their work using germanium (Ge) powder, which has a higher index of refraction than GaAs. They report a value for the localization parameter of $kl = 4.5$, which is much higher than the GaAs powder (Rivas, Sprik et al. 2001). This is an interesting result, because it would be expected that the higher index Ge, would result into a smaller localization parameter. Evidently, the subject is still a matter of debate. Localization has also been observed in the microwave regime using a sample consisting of a mixture of teflon and metallic spheres (Garcia and Genack 1991; Genack and Garcia 1991).

The precursor to Anderson localization is known as the coherent backscattering effect or weak localization (Albada and Lagendijk 1985; Wolf and Maret 1985; Mark, Albada et al. 1988). In a disordered medium, where there are a number of scattering events, waves take many random paths. If there are enough scattering events and the scatterers are close enough to each other, the light could return to a point of origin forming a loop as shown in Figure 2.5. In a loop, two paths of propagation of opposite

Figure 2.6: Coherent backscattering of 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.

direction occur with equal probability because both paths encounter the same scatterers. Both directions acquire the same phase and therefore add with constructive interference. 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 via either direction along the closed path. Thus, backwards-scattered light experiences constructive interference. Experiments have demonstrated that the intensity of the backscattered waves is nearly twice the surrounding background intensity (Albada and Lagendijk 1985), because of this constructive interference. A model of the coherent backscattered effect is shown in Figure 2.6. Light is propagated through a scattering medium and is measured in the backward direction as a function of angle; a heuristic plot of this is shown in Figure 2.7. 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 proportional to

Figure 2.7: A heuristic of the intensity of backscattered light as a function of angle. The backscattering cone has twice the intensity as the background due to constructive interference of the backscattered light.

λ/λ_{sc} . The coherent backscattering effect demonstrates the propagation of waves in a closed path, which is an early signature of the onset of localization.

We have seen that single scattering of a particle can be described using Mie theory, but multiple scattering involves a much more complex solution. Mie theory provides an analytical solution for a wave scattered by a sphere. It allows for the solution of the scattering amplitude function, the scattering cross-section, the absorption cross-section, and other important parameters. The primary parameters for multiple scattering are the mean free path and effective index of refraction. Many different methods have been considered to calculate the complex scattering index of refraction for an inhomogeneous media. The complex scattering index of refractions gives information on parameters such as the mean free path and the effective index of refraction. Complexity arises to solve for these parameters in large volume fractions due to the particle

correlations. Theoretical models, such as EFA and QCA, solve for these parameters at varying volume fractions. Many interesting phenomena, such as the coherent backscattering cone and localization, can be observed in a scattering media. In the experiments described in this thesis, we explore the regime of very high volume fraction, for which even the QCA is not expected to be adequate. The approach to light localization will require the use of high index scatterers, and is the topic of future research

Chapter 3: Terahertz Time-Domain Spectroscopy

3.1 Introduction

Terahertz radiation is an electromagnetic wave with a frequency of 10^{12} Hz. The terahertz (THz) frequency range is also known as the far-infrared range of the electromagnetic spectrum and lies between the microwave and infrared regions. The far-infrared region is one of the least explored ranges of the spectrum, because of difficulties involved in efficient generation and detection of THz radiation. In the past ten years, more practical means of generating and detecting THz radiation have been developed. Many interesting properties of materials have been observed through further exploration of the far-infrared frequency regime.

One effective system to generate and detect THz radiation is known as a terahertz time-domain spectrometer (THz-TDS). A THz-TDS measures the electric field, which provides both the intensity and the phase information of the THz radiation. Imaging systems in which only the intensity is measured are limited to a 2-D image construction; however, with the THz-TDS it is possible to extract thickness and index of refraction parameters from different materials. The ability to capture phase information of a THz pulse opens the door for many practical applications.

The frequency response of many materials spectrum exhibits unique properties in the far-infrared region of the electromagnetic (Hu and Nuss 1995; Mittleman, Gupta et al. 1999). The broad bandwidth and high signal-to-noise ratio makes the THz-TDS system ideal to observe the spectroscopic response of these materials. Numerous applications for detecting materials, parameter extraction, and imaging have been proposed (Mittleman,

Jacobsen et al. 1996; Mittleman, Jacobsen et al. 1998; Dorney, Baranuik et al. 2001). Water absorbs very heavily at approximately 1 THz. This allows for possibilities in biomedical applications (Loffler, Bauer et al. 2001). Cardboard and plastics are virtually transparent to THz radiation, which leads to applications for package inspection in industry (Hu and Nuss 1995). Algorithms exist to measure the thickness and index of refraction of a material (Dorney, Baranuik et al. 2001). Schemes for detecting and sensing of gases have also been developed (Mittleman, Jacobsen et al. 1998). New applications and ideas are continually being explored.

3.2 Overall System

In Fig. 3.1, a typical terahertz time-domain spectrometer system is shown. The main components of a THz-TDS system are a femtosecond laser, a scanning delay line, a detector, a transmitter, an analog to digital converter, and a digital signal processor. The femtosecond laser acts as an optical source for the detector and the transmitter. The emitted THz beam is collimated, and then can be focused down to a focal point to obtain a higher resolution for imaging applications. The scanning delay line is used to down convert the detected waveform into the kilohertz range in order for it to be processed using slow electronics.

A standard femtosecond laser is the mode-locked Ti-Sapphire laser, which can emit pulses of 100 fs in duration. The short time duration implies that the pulse encompasses a large spectrum of wavelengths. The spectrum is centered near 800 nm, the near-infrared region of light. The laser pulse is used to induce current through the transmitter and to measure current in the detector. The delay in time from when the laser pulse interacts with the transmitter to generate the radiation and when the pulse acts on

Figure 3.1: A schematic of a Terahertz Time-Domain Spectrometer system. The transmitted THz beam is collimated, but lenses can be inserted to focus the beam down (Mittleman, Gupta et al. 1999).

the detector to measure the electric field is synchronized. By increasing and decreasing this delay, the phase of the radiation can be measured. The femtosecond is used to synchronize the transmitter and detector by providing a point of reference.

The transmitter and detector can be described in a very complete mathematical model using the Drude-Lorentz equations (Jepsen, Jacobsen et al. 1996). However, only knowledge of antennas and electromagnetic theory is needed for a conceptual understanding. The transmitter and detector are basically antennas and semiconductors with lenses attached to them. A voltage is applied across the transmitter, which then permits a small amount of current to flow. The femtosecond laser interacts with the semiconductor to allow for a larger current to flow through the antenna; this time-varying current then generates a radiated electric field. The electric field is emitted through the

semiconductor and collected by the silicon lens. The radiation propagated through the silicon lens can be approximated by a spherical wave (Rudd, Johnson et al. 2001). The silicon lens directs the radiation to an optical lens where the beam is then collimated. Focal lenses can be inserted for a finer resolution. The radiation is then directed on to the detector by another silicon lens. The current induced in the receiver antenna is then measured and the terahertz pulse can be determined because $E_{THz}(t)$ is proportional to the current.

The femtosecond laser generates pulses of approximately 100 fs in duration at a repetition rate of 80 MHz. In order to increase the signal-to-noise ratio (SNR), the pulses are averaged using a scanning optical delay line. A scanning optical delay line measures and averages at one time delay point and then moves to another time delay and averages. It essentially allows for the waveforms to be sampled with a high SNR at the cost of data collection time. The acquisition of THz waveforms used to take minutes, but new methods have been developed to acquire a waveform in milliseconds while maintaining a high SNR (Hu and Nuss 1995).

3.3 Generation and Detection of Terahertz Radiation

Electrically generating an electromagnetic wave that is picoseconds in duration with a broad bandwidth ranging from 50 GHz to as high as 5 terahertz is challenging, but has been accomplished very efficiently with a femtosecond laser, a semiconductor, a metal antenna, and a silicon lens as shown in Fig. 3.2. The emitter antenna consists of a high-resistivity chromium compensated GaAs substrate wafer with a standard mixture of gold-titanium for the metal leads on the wafer. Attached to the GaAs substrate is a silicon lens. The choice for silicon is based on two main reasons. First, silicon has an

Figure 3.2: Simple schematic of a THz setup with the emitter and detector antenna geometry shown (Jepsen, Jacobsen et al. 1996).

index of refraction of 3.42 and GaAs has an index of refraction of 3.60. Because the indices of refraction of both materials are fairly close, this decreases the amount of radiation that is reflected between the two different surfaces, thus increasing the transmission. Second, the effects of dispersion and absorption for silicon in the terahertz regime are minimal.

The antenna structure of a typical emitter as shown in Fig. 3.2 is composed of two metal leads that are separated by $50 \mu\text{m}$ attached to a GaAs substrate. There is an applied bias voltage across the two metal leads in order to induce a small current across the antenna. When the femtosecond laser interacts with the GaAs substrate the density of carriers increases in the semiconductor and current flows through the antenna. The femtosecond laser is a very intense short pulse and when it is applied to the substrate, the current increases very rapidly and then drops down to zero before the next pulse arrives.

Figure 3.3: A terahertz time signal and the magnitude of the frequency response.

The electric field generated by the emitter is proportional to the derivative of the current flowing through the antenna. Notice in Fig. 3.3 that this is essentially the description of the terahertz pulse.

There are several interesting features to observe from the terahertz signal and frequency response shown in Fig. 3.3. Notice the oscillations in the frequency response of the terahertz pulse. Water absorbs very heavily in the far-infrared regime and those oscillations are due in part to water vapor in the air (van Exter, Fattinger et al. 1989). In the time signals, there are several bumps that follow the main transient pulse. Those are inherent to the terahertz system and are caused by optical or electrical effects, such as impedance mismatches in the antennas. The frequency response shows that there is signal energy as far out as 1.5 THz. There have been systems reported to reach frequencies up to 5 THz (Ralph, Perkowitz et al. 1994).

The silicon lens is an important element in the antenna that allows for the broad bandwidth. After the radiation is emitted from the substrate, it is directed radially

Figure 3.4: THz reflection image of a circular burned region on a tissue sample (chicken breast). Darker shades indicate less reflected radiation (Mittleman, Gupta et al. 1999).

outward from the GaAs. The silicon lens is attached to the GaAs in order to collect the radiation and direct it towards the collimating lens. It is cemented to the GaAs substrate to reduce reflections between the two surfaces. Due to the small sizes of the apertures, diffraction effects prevent THz radiation of less than 50 GHz to be efficiently generated. The silicon lens effectively gathers all the radiation that is generated into a spherical wave and directs it toward a lens where it can be collimated.

The THz detector, shown in Fig. 3.2, is essentially the same as the emitter with the exception of a current meter instead of a DC bias. The detector consists of a silicon lens, a GaAs substrate wafer, a metal antenna, and a current meter. The GaAs wafer acts as a photoconductive switch to allow current to flow through the antenna. The silicon lens focuses the THz radiation on to the antenna. The femtosecond laser interacts with

Figure 3.5: THz image of a packaged semiconductor integrated circuit (Hu and Nuss 1995).

the GaAs substrate to short the photoconductive switch. The THz radiation induces a current in the antenna that is proportional to its electric field. The current that is measured is the average electric THz field over the detector area. A scanning delay line is used to average over several pulses of THz radiation as well as sample the electric field.

3.4 Applications

Since the development of the terahertz time-domain spectrometer, numerous applications have been proposed and implemented. Several biomedical applications have been suggested, such as skin cancer detection, burn tissue diagnostic, and tooth cavity detection (Mittleman, Gupta et al. 1999; Jaeyoung, Yichul et al. 2000; Loffler, Bauer et al. 2001). Algorithms for determining the thickness and index of refraction of various materials have been developed (Dorney, Baranuik et al. 2001). Gas sensing experiments have been performed to detect the presence and determine the composition of different gases (Mittleman, Jacobsen et al. 1998). Applications for package inspection have been proposed because of the transparency of THz radiation to plastics and cardboards (Mittleman, Gupta et al. 1999). Defects in semiconductor integrated chips can be

detected using a THz-TDS (Hu and Nuss 1995). There are a variety of different and interesting applications being worked on and many more that are being suggested.

Biomedical applications are always very interesting, because there is a large demand for non-invasive procedures. One application in this area is the diagnosis of tissue burn severity. For second degree burns, it is very difficult to determine how serious the burn is without actually making an incision into the patient. An application has been proposed to measure the reflection of THz radiation off of burnt tissue (Mittleman, Gupta et al. 1999). An experiment was performed using burnt chicken breast as the model tissue system. Terahertz rays were projected at the tissue sample and then the energy of the reflected radiation was measured as shown in Fig. 3.4. The circular rings denote varying degrees of reflectivity with the center ring being the least reflective. The center portion of the tissue is the most severely burned. In the experiment, several spectral differences between the dark regions and the white regions were observed that can not be due solely to changes in water content. Therefore, factors such as the chemical changes of the tissue result in the observed spectral change. With more investigation, the causes for the spectral changes of the tissue could be identified and be used in a non-invasive diagnosis of burn depth and severity.

Differences between malignant and benign skin tumors could also be apparent in the spectral response of the tissue. One research group noted several differences of reflectivity verses different wavelengths for various types of pigmented skin lesions using white light (Wallace, Crawford et al. 2000). They were not able to explore the reflectivity in the far-infrared regime, but with the THz-TDS it is possible that larger differences in the spectral response of various types of skin tissues could be observed.

The main limitation that exists for biomedical applications is the high absorption of water in the THz regime. This limits the imaging capabilities to the surface of the skin. These experiments show promising results for future exploration and practical applications in the biomedical industry.

Imaging of plastics and semiconductors show promising results for applications in industry for package inspection and defect detection. For example, air bubbles in integrated chips are very damaging and cause the device to be inoperable. Breaks in the metal leads connecting the semiconductor to an outside circuit board would make the chip unusable. Figure 3.5, shows an image of a semiconductor integrated circuit inside of its plastic packaging. Notice that the metal leads from the semiconductor are plainly visible. The packaging is virtually invisible to the terahertz radiation. The image is very coarse and no image processing techniques have been applied to improve the quality. Methods using a Michelson Interferometer have been performed in conjunction with the THz-TDS to improve the resolution of the image (Johnson, Dorney et al. 2001). A variety of wavelet based de-noising techniques have been suggested for terahertz signals (Mittleman, Jacobsen et al. 1998; Ferguson and Abbott 2001).

The THz-TDS is an effective imaging system when the object of interest is surrounded by plastic or cardboard. This opens opportunities for applications in food inspection, quality control, packaging, and many other areas. Using the THz-TDS, images of raisins in a box of cereal were obtained (Mittleman, Gupta et al. 1999). The raisins absorbed the THz radiation significantly due to the water content and therefore are distinguishable from the surrounding cereal. Imaging experiments to distinguish the portions of meat that are fatty have been performed. The water concentration of cheese

Figure 3.6: (a) Conventional reflective T-ray image of the floppy disk and (b) tomographic image of the same disk at a constant vertical position (Mittleman, Hunsche et al. 1997)

has also been measured. In the area of food inspection, it is very desirable to have a non-invasive method to determine the quality of the food. The THz-TDS provides a safe and non-invasive means of inspection.

Tomography is another area where the THz-TDS provides excellent results. In seismology, tomography is very important for the exploration of the earth for different materials such as oil. Many algorithms have been developed to model the propagation of sound waves through various mediums. Sound waves behave very similar to electromagnetic waves in regards to propagation, refraction, and interference. There is difficulty in determining the accuracy of many of those algorithms due to the expensive cost and challenge of drilling deep into the earth. Many of the same algorithms applied in geophysics can also be applied to THz tomography. Figure 3.6 shows both a reflective

image and a tomographic image of a 3.5-in. floppy disk (Mittleman, Hunsche et al. 1997). Using algorithms developed for tomographic imaging, the various layers of the disk are shown. The accuracy of the algorithm can easily be measured because the composition of the disk is apparent. Notice that the metallic hub does not transmit any of the THz radiation. The surface that appears behind the metal hub is due to a multiple reflection.

Gas sensing is another promising application area (Mittleman, Jacobsen et al. 1998). Many gases exhibit unique resonant frequencies in the THz regime. The absorption lines in the gases are due to transitions between rotational quantum levels. These are unique spectral features and can be used to identify the species. Due to the large bandwidth and the ability to measure both the magnitude and phase of the transmission, the spectral response of the gas can be measured. Not only has it been possible to detect certain gases, but also experiments have been performed that identified the composition of gas mixtures. Gas sensing is of much interest because it would be very useful to detect gases that may be harmful to people.

The numerous applications described here are only a few of the many suggested areas in which THz-TDS could have an impact. Interestingly, the system is already in commercial use in the area of quality control. In many such applications, the THz beam interacts with a scattering medium, either as the target or as a barrier in front of the target. The research described in this thesis could be of significant value in furthering these types of applications.

Chapter 4: Scattering of Terahertz Pulses

4.1 Experimental Setup

The propagation of classical waves in the presence of 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 (Ishimaru 1978; Robinson 2000). If the scattering probability is high enough, localization effects analogous to the Anderson localization of electrons can occur (Wiersma, Bartolini et al. 1997), although the unambiguous experimental realization of this effect is quite challenging (Scheffold, Lenke et al. 1999). With weaker scattering, diffusive transport of photons has been observed (Graaff and Ten Bosch 2000; Li, Pattanayak et al. 2000). This has recently prompted a great deal of interest in the medical optics community, as one may hope to image the location of objects immersed in scattering media such as tissue (Hyde, Farrell et al. 2001). The usual description of diffusive propagation relies on the fact that the thickness of the sample L is much larger than the Boltzman transport mean free path λ_r (Ishimaru 1978). In this case the incident, coherent light beam is completely randomized, and only multiply scattered photons are transported through the sample. As the sample thickness decreases, there must eventually be a crossover from diffusive to ballistic transport (Chabanov and Genack 1997). Despite several recent studies of this phenomenon, there are conflicting reports as to the nature of this crossover, and even the value of L/λ_r at which it occurs (Kop, de Vries et al. 1997; Boguna, Porra et al. 1999; Ramakrishna and Kumar 1999; Zhang, Jones et al. 1999; Rivas, Sprik et al. 2001).

Here, we describe a new experimental technique for studying the propagation of waves in scattering media, and apply it to the investigation of thin samples, for which $L/\lambda_r < 10$ (Pearce and Mittleman 2001). The measurements are based on terahertz time-domain spectroscopy, in which broadband single-cycle pulses of free-space terahertz (THz) radiation are generated and coherently detected using ultrafast photoconductive sampling. This technique offers several unique advantages over other methods used to study multiple scattering phenomena. First, the results are easier to interpret because the samples are extremely well controlled, and the refractive index and residual absorption coefficient are both known accurately. Difficulties in characterizing the samples has been one of the most significant problems in interpreting experimental results (Scheffold, Lenke et al. 1999; Rivas, Sprik et al. 2001). Second, the broad bandwidth of the THz radiation enables the observation of mean free paths ranging over nearly two orders of magnitude within the pulse spectrum. This permits the characterization of a full range of behavior from $L \ll \lambda_r$ to $L > \lambda_r$ in a single measurement. Finally, because the technique permits the direct measurement of the spectral phase of the transmitted radiation, a great deal of additional information is available. This capability has also proven useful in recent microwave measurements, except that free-space THz measurements avoid the need for a waveguide geometry, which can introduce multimode propagation effects and other complications (Chabanov and Genack 1997).

A basic measurement in the study of scattering phenomena is that of transmission through a random medium, as a function of the length of the medium. We construct a model random medium using commercially available polytetrafluoroethylene (PTFE, teflon) spheres, with size polydispersity $< 3\%$ on the diameter. Teflon is an excellent

Figure 4.1: A schematic of the transmission experiment.

material for these studies since its absorption coefficient is quite low: $\alpha < 0.1 \text{ cm}^{-1}$ for frequencies below 500 GHz, and $\alpha \sim 0.5 \text{ cm}^{-1}$ at 1 THz (Birch, Dromey et al. 1981). Also, the refractive index of teflon, $n_{\text{PTFE}} = 1.4330$, is nearly independent of frequency throughout the spectral range of the measurements (Birch, Dromey et al. 1981). We have studied spheres of 0.794 mm diameter, and also performed a more limited number of measurements on 1.19 mm diameter spheres. The spheres are contained in a teflon sample cell, with windows a fixed distance apart. In order to perform length-dependent studies, we fabricated fifty such cells, with internal path lengths ranging from 1.19 mm to 20.64 mm. For each path length, we collect both a reference (empty cell) and a sample waveform, so shifts in the transmission due to small variations in the cell window thicknesses can be corrected. The sub-picosecond THz pulses are generated and detected using a setup quite similar to the one described by Mittleman *et al.*, (Mittleman, Jacobsen et al. 1996), except that $f = 10 \text{ cm}$ polyethylene lenses are used instead of off-axis

Figure 4.2: (a) THz waveforms transmitted through a random medium, consisting of a collection of teflon spheres. Path lengths are as shown. The reference waveform has been reduced in amplitude by a factor of 2. Each vertical tick mark is one nanoampere. (b) Amplitudes and (c) phases of the Fourier transforms of the reference waveform (dashed), and the first, third, fifth, and seventh waveforms in (a).

paraboloidal reflectors to transport the THz beam, as shown in Fig. 4.1. The confocal parameter for our optical configuration is at least ten times the sample thickness, so the beam approximates a plane wave in the medium. Fig. 4.2(a) shows a series of waveforms, for several different path lengths. As the path length increases, the pulses takes longer to transit through the sample, and their amplitude decreases. This decrease is primarily due to the scattering of high frequency components, as seen in Fig. 4.2(b). The low frequency portions of the waveform scatter much less readily, and therefore persist to much longer path lengths. Also, as seen in Fig. 4.2(c), the spectral phase shows slightly non-linear behavior as the path length increases.

There are several theoretical models that can be used to calculate the various propagation constants. One such model is the effective field approximation (EFA),

Figure 4.3: Scattering mean free path $\lambda_{sc}(\omega)$ versus free-space wavelength, on a log scale. The solid squares are for the smaller spheres, with diameter 0.794 mm, while the open circles are for the larger spheres, $d = 1.19$ mm. The line shows the computed $\lambda_{sc}(\omega)$ for the smaller spheres, using Mie theory. The error bars represent the uncertainties associated with repeated measurements. The inset shows the measured values of $k\lambda_{sc}$ versus wavelength, for the smaller spheres.

which assumes there are no particle correlations (Tsang, Kong et al. 1985). This is valid for situations for small particle densities where the volume fraction is less than $\sim 1\%$. Another model is the quasi-crystalline approximation (QCA) (Tsang, Kong et al. 1985). It considers two-particle correlations and is valid for volume fractions up to $\sim 40\%$. We perform computations of the QCA using Matlab routines available on the internet (Kong 2002). The EFA result can be obtained by computing the QCA with the pair distribution function set equal to one (Tsang, Kong et al. 1985).

4.2 Amplitude

The results of Fig. 4.2(b) can be understood as the frequency-dependent scattering

of photons out of the coherent beam. The transmission of the coherent beam varies exponentially with propagation distance according to

$$|E_{coh}(\omega)| = \exp\{-z/2\lambda_{sc}(\omega)\} \cdot \exp\{-\alpha(\omega)z/2\} \quad (4-1)$$

where $\lambda_{sc}(\omega)$ is the scattering mean free path, related to the transport mean free path by $\lambda_r / \lambda_{sc} = 1/(1 - \langle \cos \theta \rangle)$ (Ishimaru 1978). The measured values of $\lambda_{sc}(\omega)$ are shown in Fig. 4.3, for the two different sphere diameters. Including the absorption factor $\alpha(\omega)$ shifts the estimated mean free path by no more than 5%, so it is neglected in the determination of these results. The more limited data set for the larger diameter is due to the limited availability of these spheres; nevertheless, it is clear that these larger spheres have a distinctly smaller mean free path. For the smaller spheres, $\lambda_{sc}(\omega)$ varies from ~1 mm to ~70 mm across the measurable spectrum. Using these results, we calculate values of $k\lambda_{sc}$ ranging from ~20 to 160 (inset to Fig 4.3). This confirms that, with a small dielectric contrast, one is always in the weak scattering limit $k\lambda_{sc} \gg 1$.

Because these samples are comprised of nearly ideal spheres, it is possible to compare to Mie theory, which can be used to compute the scattering cross-section σ_s for an isolated dielectric sphere. Indeed, this formalism has recently been applied to scattering of THz pulses from isolated spheres (Cheville, McGowan et al. 1998; Reiten, Grischkowsky et al. 2001). Although its applicability to dense collections of spheres is not clear, it is nonetheless instructive to compare to our results. The solid line in Fig. 4.3 shows a numerical computation of $\lambda_{sc}(\omega) = [n_0 \sigma_s(\omega)]^{-1}$ for the smaller spheres, using Mie theory. Here n_0 is the number density of spheres, which can be determined by weighing the cell on a precision balance. From such measurements we determine a

Figure 4.4: The open squares show the measured reduced scattering coefficient as a function of wavelength, extracted from the time-domain waveforms by Fourier transform. The error bars show typical uncertainties due to variability in repeated measurements. The solid curve shows the prediction of the quasi-crystalline approximation.

volume fraction of $\phi = .56 \pm 0.04$, and therefore a number density of $n_0 = 2.14 \pm 0.15$ spheres per mm^3 . Except at the shortest wavelengths, the Mie result is significantly smaller than the measured values. However, the weak oscillatory structure in the calculated curve is echoed in the data. The departure from Mie theory is probably due to the high volume fraction, which leads to correlations in the sphere positions due to excluded volume effects. These are known to increase the mean free path, relative to that predicted by the curve shown in Fig. 4.3 (Fraden and Maret 1990).

Fig. 4.4 shows a comparison of the experimental and theoretical reduced scattering coefficient $1/\lambda_{tr}$ as a function of wavelength using the quasi-crystalline approximation. These data span the range of size parameters from $ka \sim 1.6$ to 4.7, and

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. We have used an approximate form for the complex dielectric of the teflon spheres, obtained by fitting a low-order polynomial to the absorption coefficient reported by Birch *et al.* (Birch, Dromey et al. 1981). Clearly, the theory 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, better than the single-particle Mie calculation shown in Figure 4.2.

4.3 Spectral Phase

In addition to the information available from the amplitude of the transmitted waveforms, we also independently determine the spectral phase (Fig 4.2(c)). This can be used to extract an effective refractive index $n_{eff}(\omega)$ for each path length. The solid curve in the lower part of Fig. 4.5 shows the average result for all path lengths. In computing this average, we do not include data points for which the amplitude of the THz field is less than twice the noise level. As a result, not all path lengths contribute to the average at each frequency; the error bars show typical uncertainties associated with this averaging procedure. This measurement of n_{eff} can be compared to several different effective medium approximations (Mishchenko, Hovenier et al. 2000), including the Maxwell-Garnett rule and volume averaging. Using the known dielectric of teflon, and neglecting its imaginary part, we can calculate $n_{eff} = \sqrt{\epsilon_{eff}}$ for these two cases. The results are shown in Fig 4.5.

Figure 4.5: The lower solid curve shows the effective refractive index, which exhibits dispersive effects due to scattering. The error bars represent the uncertainties associated with repeated measurements. The dashed curve is the result of a Monte Carlo simulation of the effective index, using the measured values of $\lambda_{sc}(\omega)$ (Fig. 4.2) and the long wavelength limiting value of $n_{eff}(\omega)$ as inputs. The arrows show two effective medium approximations for the composite sample, using the measured volume fraction. The uncertainty in the volume fraction leads to the error bars shown. The upper portion of the figure shows the measured index of solid teflon, which is not dispersive and agrees well with literature values (Birch, Dromey et al. 1981).

Several aspects of this data set are worth comment. First, at long wavelengths where the scattering is weakest, the index is non-dispersive, and falls at an intermediate value between the predictions of the two effective medium approximations. This value reflects the effective index in the absence of scattering, and demonstrates the inadequacy of effective medium approximations in situations where the filling fraction is near 50%. We also observe that, as the wavelength decreases, the measured index gradually increases, with a fractional shift larger than 3% at the smallest accessible wavelength. This effect is also observed with the larger spheres. To demonstrate that this is not due to intrinsic material dispersion in the sample, we show at the top of Fig. 4.5 measured

values for the index of a solid block of teflon. The index of the bulk material does not vary by more than 0.1% from its average, which is in excellent agreement with the literature value (Birch, Dromey et al. 1981).

One possible explanation for dispersion in $n_{eff}(\omega)$ is the result of the increased path length traversed by photons that have scattered through small angles. The collection optic used in our measurement is approximately $f/2$, so scattered photons whose propagation has been diverted by less than $\sim 14^\circ$ are still collected by the lens. These photons have traveled a slightly longer path through the material, and are therefore delayed somewhat. We can provide support for this explanation with a simple Monte Carlo simulation (Mishchenko, Hovenier et al. 2000). For this simulation, we use the measured values for $\lambda_{sc}(\omega)$, and also the long-wavelength limiting value of $n_{eff}(\omega)$ to convert distance traveled within the sample into transit time. The scattering is assumed to be isotropic. A typical output, shown in Fig. 4.5 (dashed curve), is reasonably consistent with the measured dispersion. It is worth noting that the average additional travel time is less than 2 picoseconds even at the shortest wavelengths and for the longest sample cell. Unlike in the case of strongly scattering samples (Kop, de Vries et al. 1997), this delay is comparable to or shorter than the duration of the incident pulses. As a result, this effect is not readily observable in the time-domain waveforms, but only as phase distortions in the frequency domain.

Although one possible explanation of dispersion is due to the scattering of photons at small angles, perhaps a more rigorous explanation can be described using the quasi-crystalline approximation, which permits an a priori computation of the effective index $n_{eff}(\omega)$. We display this in Figure 4.6, which shows the phase velocity, given by

Figure 4.6: The open circles show the measured phase velocity, c/n_{eff} , as a function of frequency, extracted from the phases of the Fourier transforms of the time-domain waveforms. The dashed curve shows the prediction of the Maxwell-Garnett mixing formula, computed using experimentally measured values for the refractive index of solid teflon. The solid curve shows the prediction of the quasi-crystalline approximation. The error bar on this curve shows the uncertainty in the computed value arising from the $\pm 4\%$ uncertainty in the volume fraction of the spheres in the samples. Because the value of this input parameter is not determined with sufficient precision, it is not possible to conclude from these measurements if the discrepancy between the QCA and the data results from this uncertainty or from shortcomings in the theory.

$v_p = c / n_{\text{eff}}$. The open circles show the measured frequency-dependent phase velocity, in units of c . One can use any of a number of different mixing formulas for computing the effective index of a two-component medium. The most common of these is the Maxwell-Garnett rule, which is equivalent to the QCA in the low frequency limit (Tsang, Kong et al. 1985). For the situation described here, the Maxwell-Garnett result, computed with experimentally determined values for the refractive index of teflon (see Figure 4.4), is shown as a dashed line in Fig. 4.6. It is clear that the dispersion and

resonant features in the experimental data are not reproduced by the mixing rule. The full QCA result is shown as a solid line in Fig. 4.6. As in Fig. 4.4, there is a systematic shift between these computed values and the measurements. This could arise from the uncertainty in the experimentally determined volume fraction, since the phase velocity is essentially linearly dependent on the volume fraction (Tsang, Kong et al. 1985). The error bar on the theoretical curve shows a typical range in the computed phase velocity resulting from this uncertainty; this range is on the same order as the discrepancy between the computation and the experiment. As a result, the discrepancy could be due to the uncertainty in the volume fraction, or to the inadequacy of the QCA at large volume fractions. In order to evaluate the accuracy of the QCA with respect to phase velocity, a more accurate method for experimentally determining the volume fraction will be required. However, despite the apparent discrepancy, it is interesting to note that the overall dispersive trend, as well as the resonant features in the spectrum, are both well reproduced by the QCA calculation.

Because these data are acquired with a broadband spectrometer, it is 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:

$$v_G = c / \left(n_{eff} + \omega \frac{dn_{eff}}{d\omega} \right). \quad (4-2)$$

This is the velocity with which a wave packet moves through the medium. The group velocity is determined experimentally by numerically differentiating the measured effective index. This is shown in Fig. 4.7, along with the QCA result (solid line). The agreement is much improved in comparison with Fig. 4.6, demonstrating that the QCA

Figure 4.7: The open circles show the measured group velocity, obtained by numerically differentiating the measured effective index. The QCA prediction (solid curve) matches the data quite well.

can be used to accurately compute group velocities even when the volume fraction is large enough so that the predicted phase velocity is less reliable.

We note that the QCA is generally assumed to be valid for volume fractions below $\sim 40\%$, whereas our measurements involve volume fractions exceeding 50% . 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. Our results indicate that the QCA provides some indication of the scattering coefficient, and at least a reasonable estimate of the phase velocity of the wave. In the case of the group velocity, the agreement with experiment is in fact quite good. Thus, while not perfect, the QCA is still useful even far beyond its expected limits of the validity. These results indicate the

diminishing importance of higher-order correlations, at least in the case of moderate dielectric contrast studied here.

We report the first use of terahertz time-domain spectroscopy in the study of propagation of waves in random media. The broad spectral coverage enables simultaneous measurements over a large range of parameters. In addition, the spectral phase distortions provide a sensitive measure of scattering statistics. These measurements should eventually lead to the development of THz imaging techniques analogous to diffuse photon density wave imaging, and also hold promise for photon localization experiments.

Chapter 5: Discussion and Future Work

5.1 Summary of Results

The scattering of electromagnetic waves is a common phenomenon that occurs in everyday life. It is a topic of research that has been studied for many years and yields applications in several disciplines. Using the terahertz time-domain spectrometer and its ability to generate and detect broadband pulses, we are able to extract the scattering parameters over a frequency range covering almost two orders of magnitude. Using a low index material like teflon, we are consistently in the weak scattering limit over the entire frequency range. Despite the fact that the volume fraction of the samples is extremely high, the resonant features in both Mie theory and the quasi-crystalline approximation are observed in the data. Due to the terahertz time-domain spectrometer's ability to measure the electric field, the scattering parameter such as phase velocity and the effective index of refraction can be extracted from the data.

The terahertz time-domain spectrometer is a very powerful tool in the investigation of wave scattering. With the broadband pulse and ability to measure the phase of the pulse, each sample measurement yielded much information. There is still much work that needs to be done. Using higher index scatterers, experiments can be performed to observe stronger scattering. For this experiment performed in this thesis, only ballistic scattering was observed. An interesting experiment would be to observe the transition from ballistic scattering to diffusion and to measure the characteristics of diffuse light. In this experiment we only measure the radiation propagating in the

Figure 5.1: The Mie Theory solution for the scattering cross-section of a silicon sphere with a radius of 0.30 mm and a dielectric constant of 3.418.

direction of the incident beam, but much could be gain by investigating the angle dependence of scattering. In fields such as medicine and geophysics, much work has been dedicated to forming images of objects embedded in a scattering media. Similar types of images can be constructed using the terahertz time-domain spectrometer and the same tools that these other fields use to analyze their images can be applied. There is still much work that can be done in this field and there are many more experiments that can be done with the aid of the terahertz time-domain spectrometer.

5.2 Higher Index Scatterers

Using a low index material such as teflon for the scatterers yields a weak scattering material. The unambiguous observation of Anderson localization is very challenging to achieve (Wiersma, Bartolini et al. 1997; Scheffold, Lenke et al. 1999; Wiersma, Rivas et al. 1999). It would be very desirable to observe that in the terahertz

Figure 5.2: THz waveforms transmitted through a random medium, consisting of a collection of silicon cubes for a variety of path lengths. The waveforms are increasing path length from top to bottom.

regime. It has already been observed using microwaves (Genack and Garcia 1991), in a waveguide geometry. In order to create a stronger scattering sample, a higher index material is needed. We have preliminary data of the same experiment performed in this thesis, but using silicon (Si) cubes instead of teflon spheres. Silicon has an effective index of refraction of 3.418 (Grischkowsky, Keiding et al. 1990), in contrast with the lower index of teflon, $n_{PTFR} = 1.4330$. Due to the non-spherical shape of the particles, the Mie theory solution for the sphere is no longer valid. However, it is nonetheless interesting to compute the scattering cross-section for a spherical Si particle. This result is shown in Figure 5.1, and can be compared to Figure 2.2. There are other non-analytical tools available to analyze the scattering of non-spherical particles. It is not

quite certain what the role of the shape of the particle plays in scattering, but that will need to be investigated.

A series of waveforms for the silicon data is shown in Figure 5.2. The top two waveforms correspond to the shortest path lengths. The waveforms are very complex and it is not clear how to describe this result. The rest of the waveforms have similar structure to those shown in Figure 4.2(a). As the path length increases, there is a decrease in the amplitude in the waveforms and it is further delayed in time. However, for the silicon the decay is much more rapid than that of the teflon and the delay is much larger as well. This implies that the silicon has a much shorter mean free path and a higher effective index of refraction. Analysis needs to be done to quantify these statements, but the preliminary data is very promising.

5.3 Angle Dependence Scattering

Coherent backscattering demonstrates the beginnings of localization. There have been several experiments in which this effect has been measured (Albada and Lagendijk 1985; Wolf and Maret 1985). It is a result of light that is backscattered in the direction opposite of the incident light adding coherently as shown in Figure 2.6. It would be very easy to perform similar measurements with our samples using the terahertz time-domain spectrometer. A measurement of the angular dependence of scattering could prove to be very useful as well. The assumption is made that scattering is typically isotropic. In actuality, for wavelengths on the order of the size of the scatterer, forward scattering dominates and the scattering is very anisotropic (Mark 1990). It would be interesting to see if there were different resonances at different angles for spheres similar to the resonances of the cross-sections at varying frequencies.

Figure 5.3: A schematic of the angle dependence scattering experiment.

Figure 5.3 shows a schematic of one of the future experiments that we would like to perform. We will construct several semicircular cells and transmit it with pulses of THz radiation. The detector will be on a rotating arm to swing across a wide range of angles and measure the electric field at each angle. With the data we collect, we can measure the intensity of the radiation at each angle for each frequency. The scattering cross-section can be extracted. Much more information could be gained by also looking at the phase information. It is challenging to measure the diffuse light, because it interferes with the ballistic light. Measuring the electric field at different angles should separate these two effects. Another interesting experiment to perform would be the coherent backscattering experiment using THz radiation. The setup would be similar to Figure 5.3 except we would use square samples and rotate behind the sample. These measurements will give us a better understanding of scattering media.

5.4 Imaging

In the past few years, the imaging power of the terahertz time-domain spectrometer has been demonstrated. In many different applications, there is a demand to make images of objects embedded in various types of media. Some of these media include skin, the earth, the atmosphere, etc. It is a challenging issue to construct images of objects embedded in an inhomogeneous material that scatters the waves that are sent. A future experiment would be to embed an object into one of the samples and transmit THz radiation through the sample. A measurement of either the transmitted or reflected field could be used to locate and image the immersed object.

Many of the algorithms used by the geophysics researchers to construct images of the ground and the biomedical researchers to form images of objects beneath the skin could be directly applied to THz imaging. It is much easier to test the results of the algorithm using the terahertz time-domain spectrometer, because it does not require us to dig up the earth or to cut into the skin. We can perform experiments in a very controlled environment. Using the electric field of the pulse, the thickness of the object can be measured.

Yodh *et al.* has constructed images of heterogeneous turbid media by frequency-domain diffusing tomography (O'Leary, Boas et al. 1996; Li, Durduran et al. 1997; Li, Pattanayak et al. 2000). They generate diffuse photon-density waves (DPDW's), which are exponentially damped spherical waves consisting of randomly walking photons propagation outward from a point source. They place the source and detector in a turbid biological material called Intralipid, which causes the light to strongly scatter. A detector is placed at various positions to detect the DPDW's as a function of position. They place a single spherical object in the center of the region and scan across the object keeping the

source and detector an equidistance apart. This suggests many possibilities of future imaging techniques using the terahertz time-domain spectrometer. However, a better understanding of diffuse light and strongly scattering media is necessary.

There are many more experiments that can be performed that would further the field of wave scattering. A better understanding of the propagation of waves in a scattering media could lead to better imaging techniques and better quality of images. The terahertz time-domain spectrometer gives us powerful imaging capabilities and is a useful tool in investigating the various properties of scattered waves. It grants us many opportunities to perform many experiments and to test current imaging methods and develop more powerful new ones.

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