Available online at www.sciencedirect.com

SCIENCE @ DIRECT®

Physica B 338 (2003) 92–96

PHYSICA B

www.elsevier.com/locate/physb

Using terahertz pulses to study light scattering

Jeremy Pearce, Daniel M. Mittleman*

Department of Electrical and Computer Engineering, Rice University, MS 366, 6100 Main Street, Houston, TX 77005, USA

Abstract

We describe a new experimental technique for studying the propagation of light in scattering media. We use terahertz time-domain spectroscopy to generate and detect sub-picosecond pulses of far infrared radiation, and employ these pulses in spectroscopic studies of synthetic random media. This technique offers a number of unique advantages in comparison with other experimental methods, including the coherent detection of the electromagnetic field, the broad bandwidth, and the ease of sample preparation. As a first demonstration, we study the ballistic transport through a dense collection of spheres. These results are compared with the predictions of the quasi-crystalline approximation.

Keywords: Terahertz; Scattering; Random media

The propagation of electromagnetic waves in random media is a topic of current interest in many research communities. Multiple scattering can lead to a rich array of phenomena, many of which are still not fully understood. Issues such as the nature of localized light [1,2] and the transition from ballistic to diffusive transport [3–5] are quite controversial, and more experimental studies are required. Many of the relevant areas of research focus specifically on the propagation of visible or near-infrared radiation, including such examples as the use of infrared light in diffuse photon tomography [6], and random lasers [7]. However, because of the scale-invariance of classical wave equations, such studies need not be performed at optical frequencies. One can instead study a well-characterized macroscopic model system, using longer wavelength radiation. Then, by a simple scaling, one can transfer these results to smaller dimensions and provide insight and guidance for future optical studies [8–11].

Here, we describe a new experimental technique for studying wave propagation in complex media. We employ terahertz time-domain spectroscopy (THz-TDS) for the generation and detection of sub-picosecond pulses of far-infrared radiation. This technique provides ready access to a wavelength range that is ~500 times larger than the wavelength of visible light. This spectral range represents the highest frequencies for which the electric field can be measured directly (rather than inferred from interferometric measurements). Terahertz measurements can therefore exploit some of the advantageous features of both optical and microwave techniques.

The THz-TDS system used for these measurements is quite similar to that described previously (see Fig. 1) [12,13]. This technique affords a

---

*Corresponding author. Tel.: +1-7133485452; fax: +1-7133485686.

E-mail address: daniel@rice.edu (D.M. Mittleman).

0921-4526/$ - see front matter © 2003 Elsevier B.V. All rights reserved.
doi:10.1016/S0921-4526(03)00467-8

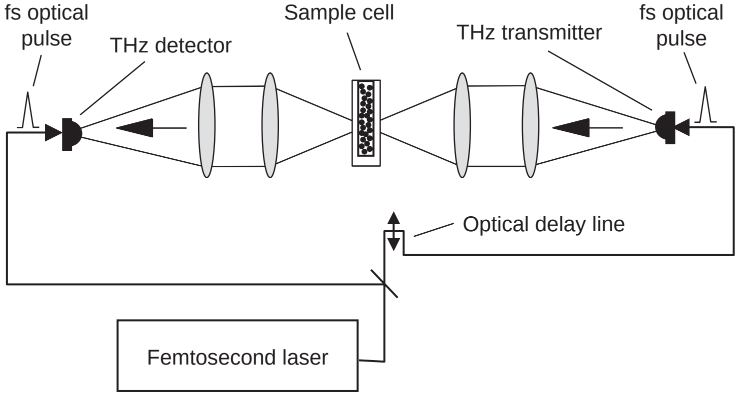

Fig. 1. Schematic of the transmission experiment.

number of advantages over traditional methods for studying scattering phenomena. First, as noted above, photoconductive sampling provides a direct measurement of the terahertz electric field $E_{\mathrm{THz}}(t)$, rather than merely the intensity. As a result, the phase information is preserved, and one may extract the full complex dielectric function of a sample without relying on Kramers–Kronig analysis. Second, the radiation is in the form of short pulses, containing a broad spectral bandwidth. It is not unusual for the spectrum of a single pulse to span more than one order of magnitude in wavelength. With this source, one can perform measurements which span the entire range from Rayleigh scattering ($ka \ll 1$, where k is the free-space wave vector and a is the particle diameter) through Mie scattering ($ka \geqslant 1$), in a single experiment. Because the measurements are time-gated, issues such as etalon effects and reflections, which can be limiting in many microwave systems, are generally not significant. Finally, unlike most traditional far-infrared detectors, photoconductive antennas operate at room temperature, thus vastly simplifying the instrumentation.

As a first demonstration, we study the ballistic transport of THz pulses through a dense collection of spheres. Such measurements require the preparation of a scale model sample with well-characterized properties. In optical experiments, the most common model scattering medium is a suspension of latex spheres in water, for which characterization of the density and size distribution are not straightforward. In contrast, the THz measurements use commercially available Teflon spheres, with a polydispersity of less than 3% on the diameter. Teflon is an excellent material for these studies since its absorption coefficient is quite low at terahertz frequencies. Also, the refractive index of Teflon, $n_{\mathrm{PTFE}} = 1.433$, is nearly independent of frequency throughout the spectral range of the measurements [14]. We have studied spheres of 0.794 mm diameter. 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 1.5 to 26 sphere diameters. We determine the number density of spheres by weighing each cell on a precision balance. From such measurements we determine a volume fraction in these samples of $\phi = 0.56 \pm 0.04$.

For each path length, we collect both a reference (empty cell) and a sample waveform, to correct for small variations in the thickness of the cell windows. Fig. 2 shows a series of THz waveforms, for several different path lengths. As the path length increases, the pulse takes longer to transit through the sample, and its amplitude decreases. From this data we can extract the frequency-dependent mean free path, by Fourier transform of the measured time-domain waveforms [11,15]. Because the THz fields are measured coherently, we can also determine the effective refractive index $n_{\mathrm{eff}}(\nu)$ of the scattering medium from the spectral phase. This effective index can differ quite substantially from a simple volume-weighted

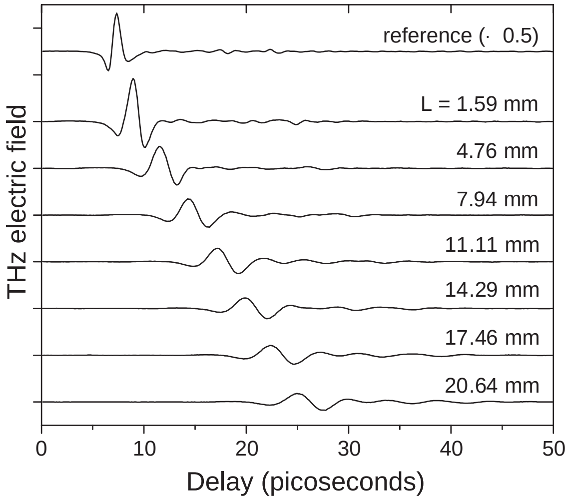

Fig. 2. The upper waveform is a typical reference, transmitted through an empty cell. The subsequent waveforms illustrate the decreasing transmission and increasing delay with increasing optical path length through the scattering medium.

average index, as a result of interference between multiply scattered waves [16].

Numerous theoretical models have been used to compute these effective propagation parameters for waves in dense collections of scatterers. 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. When the multiple scattering equations are averaged over all configurations of the scatterers, a hierarchy of solutions results, relating the conditional average of the Green's function with n particles fixed to that with $n+1$ particles fixed. The simplest solution consists of truncating this hierarchy at first order. This is known as the effective field approximation (EFA), and is equivalent to neglecting all correlations among the locations of scatterers [16]. The EFA is usually only valid for small particle densities. For higher volume fractions, 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 [16]. 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 [17]. For spherical scatterers, the Percus–Yevick pair distribution function provides an adequate description of the positional correlation [18]. Algorithms for performing QCA computations are generally available [19].

Fig. 3 shows a comparison of the experimental and theoretical mean free path as a function of wavelength. These data span the range of size parameters from $ka \sim 1.6$–4.7, and show a variation in the mean free path by roughly a factor of 40. The solid lines show the predictions of the QCA and of Mie theory, 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. [14]. Clearly, the QCA underestimates the strength of the scattering, a result of neglecting higher than second-order correlations. Even so, the correspondence to the resonant features in the data is quite good.

The effects of multiple scattering can also be seen in the velocity of the transmitted radiation. The effective refractive index determines the phase velocity, according to $v_P(\nu) = c/n_{\text{eff}}(\nu)$. Fig. 4a shows the measured frequency-dependent phase

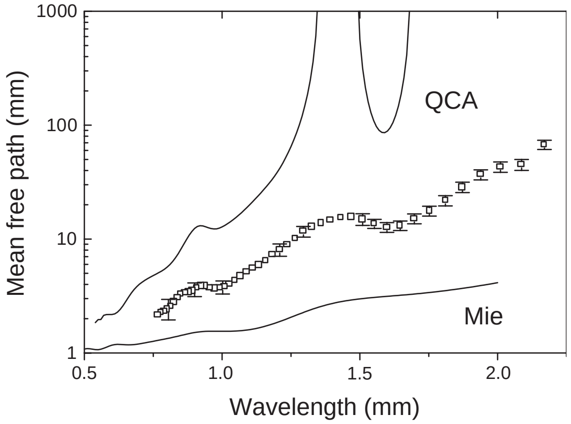

Fig. 3. Mean free path as a function of wavelength (squares), extracted from the time-domain waveforms by Fourier transform. The error bars show typical uncertainties due to variability in repeated measurements. The solid curves shows the prediction of the QCA (upper) and of Mie theory (lower).

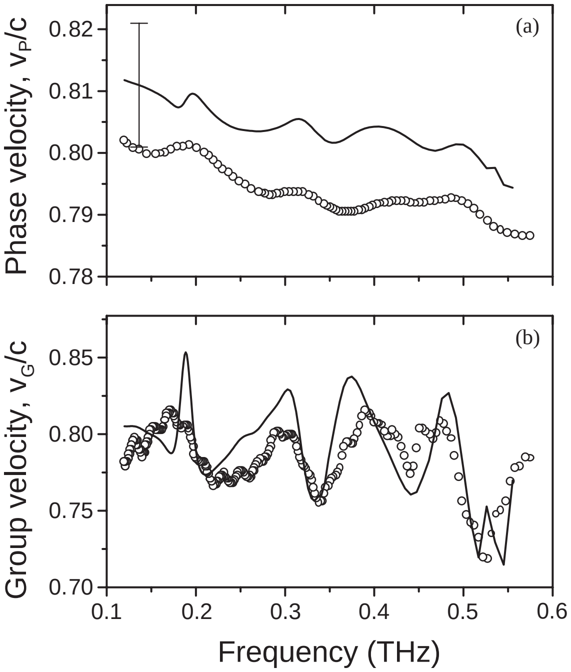

Fig. 4. (a) Measured phase velocity, c/n_{eff}, as a function of frequency (circles), extracted from the phases of the Fourier transforms of the time-domain waveforms. The solid curve shows the prediction of the QCA. The error bar on this curve shows the uncertainty in the computed value arising from the $\pm 4\%$ uncertainty in the volume fraction of the spheres in the samples. (b) Measured group velocity (circles), obtained by numerically differentiating the measured effective index. The QCA prediction (solid curve) matches the data quite well.

velocity, in units of c, along with the QCA calculation (solid curve). There is a systematic offset between the computation and the measured result. This discrepancy could arise from uncertainty in the volume fraction of the sample, or from the inadequacy of the QCA at volume fractions exceeding 50%. Even so, it is interesting to note that the overall dispersive trend and the resonant features in the spectrum are both well reproduced by the 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 $v_G(\omega) = c/(n_{\mathrm{eff}} + \omega(\mathrm{d}n_{\mathrm{eff}}/\mathrm{d}\omega))$. 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 with respect to frequency. This is shown in Fig. 4b, along with the QCA result (solid curve). The agreement is much improved in comparison with Fig. 4a, demonstrating that the QCA 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.

In conclusion, we note that the QCA is generally assumed to be valid for volume fractions below ~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. Finally, we point out that, in addition to measuring the ballistic component of the wave propagating in the random medium, it is also possible to use THz-TDS to study the diffusive portion of the wave, for which the phase has been randomized by multiple scattering [20,21]. This represents a new method for characterizing classical diffusing waves.

Acknowledgements

This work has been supported in part by the United States National Science Foundation.

References

[1] D.S. Wiersma, P. Bartolini, A. Lagendijk, R. Righini, Nature 390 (1997) 671.
[2] F. Scheffold, R. Lenke, R. Tweer, G. Maret, Nature 398 (1999) 206.
[3] A.A. Chabanov, A.Z. Genack, Phys. Rev. E 56 (1997) 1338.
[4] R.H.J. Kop, P. de Vries, R. Sprik, A. Lagendijk, Phys. Rev. Lett. 79 (1997) 4369.

[5] Z.Q. Zhang, I.P. Jones, H.P. Schriemer, J.H. Page, D.A. Weitz, P. Sheng, Phys. Rev. E 60 (1999) 4843.

[6] D.E. Hyde, T.J. Farrell, M.S. Patterson, B.C. Wilson, Phys. Med. Biol. 46 (2001) 369.

[7] H. Cao, Y.G. Zhao, S.T. Ho, E.W. Seelig, Q.H. Wang, R.P.H. Chang, Phys. Rev. Lett. 82 (1999) 2278.

[8] C.R. Schumacher, J. Appl. Phys. 62 (1987) 2616.

[9] R.A. Cheville, D. Grischkowsky, Appl. Phys. Lett. 67 (1995) 1960.

[10] T.M. Goyette, J.C. Dickinson, J. Waldman, W.E. Nixon, Proc. SPIE 4053 (2000) 615.

[11] J. Pearce, D. Mittleman, Phys. Med. Biol. 47 (2002) 3823.

[12] P.U. Jepsen, R.H. Jacobsen, S.R. Keiding, J. Opt. Soc. Am. B 13 (1996) 2424.

[13] D.M. Mittleman, R.H. Jacobsen, M.C. Nuss, IEEE J. Selected Top. Quantum. Electron. 2 (1996) 679.

[14] J.R. Birch, J.D. Dromey, J. Lesurf, Infrared Phys. 21 (1981) 225.

[15] J. Pearce, D. Mittleman, Opt. Lett. 26 (2001) 2002.

[16] L. Tsang, J.A. Kong, R.T. Shin, Theory of Microwave Remote Sensing, Wiley, New York, 1985.

[17] R. West, D. Gibbs, L. Tsang, A.K. Fung, J. Opt. Soc. Am. A 11 (1994) 1854.

[18] L. Tsang, J.A. Kong, T. Habashy, J. Acoust. Soc. Am. 71 (1982) 552.

[19] J. Kong, Electromagnetic Wave MATLAB Library, Center for Electromagnetic Theory and Applications, Massachusetts Institute of Technology, http://www.emwave.com, 2001.

[20] Z. Jian, J. Pearce, D. Mittleman, Phys. Rev. Lett. 91 (2003) 033903.

[21] J. Pearce, Z. Jian, D. Mittleman, Phys. Rev. Lett. 91 (2003) 043903.