Terahertz scattering by dense media

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(Received 16 March 2012; accepted 3 May 2012; published online 14 June 2012)

Frequency dependent absorption of a given material at distinct frequencies in the terahertz (THz) range is commonly used as a *spectral fingerprint* for material identification and classification. However, in the presence of strong scattering, these features can often become distorted or altered. Thus, there is an important need to understand how scattering from a sample alters the THz signal. In this letter, we propose an iterative algorithm that builds on the effective field theory proposed by P. C. Waterman and R. Truell [J. Math. Phys. **2**, 512–537 (1961)] and offers a rather simple and computationally efficient method for accurately explaining the multiple scattering response of a medium. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4720078]

Scattering from granular samples measured in transmission mode has been investigated by various authors.[2,9–12] When the volume fraction of scattering particles in a composite medium is low, i.e., when the average inter-particle distance is greater than the incident wavelength, multiple-scattering processes are unlikely to occur. The field that impinges on each particle can be identified with the primary incident wave.[1,3] For such cases, the macroscopic optical constants of the medium can be described in terms of the response of single particles to the incident field.

Bandyopadhyay *et al.*[9] used the Mie formalism under the independent scattering approximation to separate the scattering effects from the measured extinction spectra of granular salt, flour and ammonium nitrate. The independent scattering approximation is found to be valid only for very low concentrations ($<1\%$). Meanwhile, Foldy's effective field approximation, which takes into account the first order multiple scattering process, is found to produce reasonable results for volume densities up to $\approx 10\%$.[4,5] However, when the density of the scatterers in the medium increases ($>10\%$), the field incident on each particle is a superposition of the incident primary wave and of the field that has previously been scattered by the other particles in the medium. In other words, multiple scattering processes become dominant and independent scattering assumptions are no longer valid. Zurk *et al.*[11] applied the dense medium theory under the quasi-crystalline approximation (QCA) to estimate the scattering attenuation for granular polyethylene (PE) pellets with air voids acting as scatterers and occupying a volume of 20% of the entire medium.

Recently, Kaushik *et al.*[12] applied the generalized self-consistent effective medium theory, proposed by Chýlek *et al.*,[14] to estimate the frequency dependent scattering loss of terahertz by two component composite granular materials, with scatterers occupying a volume fraction up to 44%.

In this letter, we propose an iterative algorithm using the effective field approximation proposed by Waterman and Truell[6] to estimate the frequency dependent scattering loss from three different granularities of polyethylene and air samples and compare the theoretical analysis with the transmission THz-TDS measurements of these samples. The proposed technique offers a rather simple and computationally efficient method for estimating the multiple scattering response of a dense medium.

Guisto *et al.*[5] gave a general description of the optical behavior for intralipid solutions in terms of the characteristics of propagation of the coherent field through a random dispersion of particles, using the Foldy-Twersky equation,

$$K = \sqrt{k^2 + 4\pi\eta f(0)}, \tag{1}$$

where, in general, the coherent field can be taken to satisfy the equation,

$$(\nabla^2 + K^2)\langle\psi\rangle = 0, \tag{2}$$

where K is the effective propagation of the medium, as calculated from the Eq. (1), and $\langle\psi\rangle$ is the coherent intensity. They argued that on consideration of Eq. (2) and because Eq. (1) may be solved by iteration, the effective propagation constant K, of a medium with high scatterer density, can be calculated by a simple iteration of Eq. (1),

1. The first step is to start with the Foldy-Twersky equation (Eq. (1)) for calculating the effective propagation constant of the medium with particles embedded in a homogeneous, non-absorbing host with propagation constant k.
2. In the second step, we again solve the above equation by considering the same dispersion of the particles, however, the host medium is now represented by the complex propagation constant K obtained in the first step. The scattering properties of the particles can now be calculated as if they were independent particles embedded into an effective medium with propagation constant K.

Giuston *et al.*[5] argued that the assumption of such a fictitious host medium should account for the multiple-scattering processes (up to second order) that occur among the particles. When calculating $f(0)$, the forward scattering

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amplitude, for the original dispersion, i.e., particles in a homogeneous, non-absorbing host, the scattering cross sections were calculated using the Mie theory, whereas for the second step of the iteration, when the particles are considered within the medium with the complex refractive index K, Giusto *et al.*[5] followed the procedure of Sudiarta *et al.*[8] They applied their iterative scheme (referred to as iterative Effective Field Approximation (EFA) now onwards) to various densities of solutions of stock Intralipid-10% and found their procedure to be highly effective up to 15% volume density and show limited disagreement at densities up to 22% for measurement of scattering coefficient carried out at a single wavelength ($\lambda = 632.8$ nm). However, it must be noted that the Foldy's EFA, given by Eq. (1), assumes the particles to be point scatterers and that the medium is sparse enough, such that the scatterers positions do not influence one another. As a result, it is valid only for the cases where the backscattering from the particles can be neglected and only forward scattering is considered. This may explain the overestimation of the measurements by the theoretical results obtained by Guisto *et al.*[5] at 22% volume density. Second, Guisto *et al.*[5] did not provide any convergence criteria for their algorithm, which is essentially a two step process.

On the other hand, Waterman and Truell[6] provided an expression for the effective propagation constant, for a medium in a concatenated slab formulation with an ensemble of finite sized scatterers, that considered the backscattering from individual particles and included terms up to the second order in η. As a result, this theory is found to produce reasonable estimates of the effective scattering attenuation for a medium with finite size particles (size parameter $a(\omega/c)$ up to 2, where a is the average particle radius) and volume densities up to 30%.[7] Waterman and Truell[6] gave the following expression for the effective propagation constant

$$K = \sqrt{k^2\left(\left[1+\frac{2\pi N f(0)}{k^2}\right]^2 - \left[\frac{2\pi N f(\pi)}{k^2}\right]^2\right)}, \tag{3}$$

where, N is the number of scatterers per unit volume, $f(0)$ and $f(\pi)$ are the forward scattering and backward scattering amplitudes of a single particle, respectively. Now, because of the fact that like Eq. (1), Eq. (3) can also be solved iteratively and that the Waterman and Truell[6] theory considers multiple scattering process for finite sized scatterers up to the second order of the scatterer density, we argue that it will be a better model for estimating effective optical properties of a dense medium with finite sized scatterers.

Here, we employ an iterative scheme similar to the one suggested by Ref. 5, however, instead of using Foldy's EFA approximation, given by Eq. (1), we have used the approximation of Waterman and Truell[6] given by Eq. (3) and we also provide a convergence condition for the algorithm to determine the optimum number of iterations required. The resulting iterative scheme is illustrated in Fig. 1 and will be referred to as *iterative WT-EFA* from now onwards.

As can be seen for the Fig. 1, in the first step, we calculate the forward $f(0)$ and backward $f(\pi)$ scattering amplitudes using the Mie formalism of scattering by a single particle. Then we use Eq. (3), to obtain the first estimate of the effective propagation constant of the medium. In the second step, we again calculate the $f(0)$ and backward $f(\pi)$ scattering amplitudes, however, this time we use the formalism of Sudiarta *et al.*[8] for scattering by a particle in an absorbing medium, this is followed by the second evaluation of K using Eq. (3). Up to this point, our algorithm is the same as that of Giusto *et al.*,[5] with only exception that we have used the Eq. (3) for calculating K. After the second step, we evaluate the self consistency condition for an effective medium given by

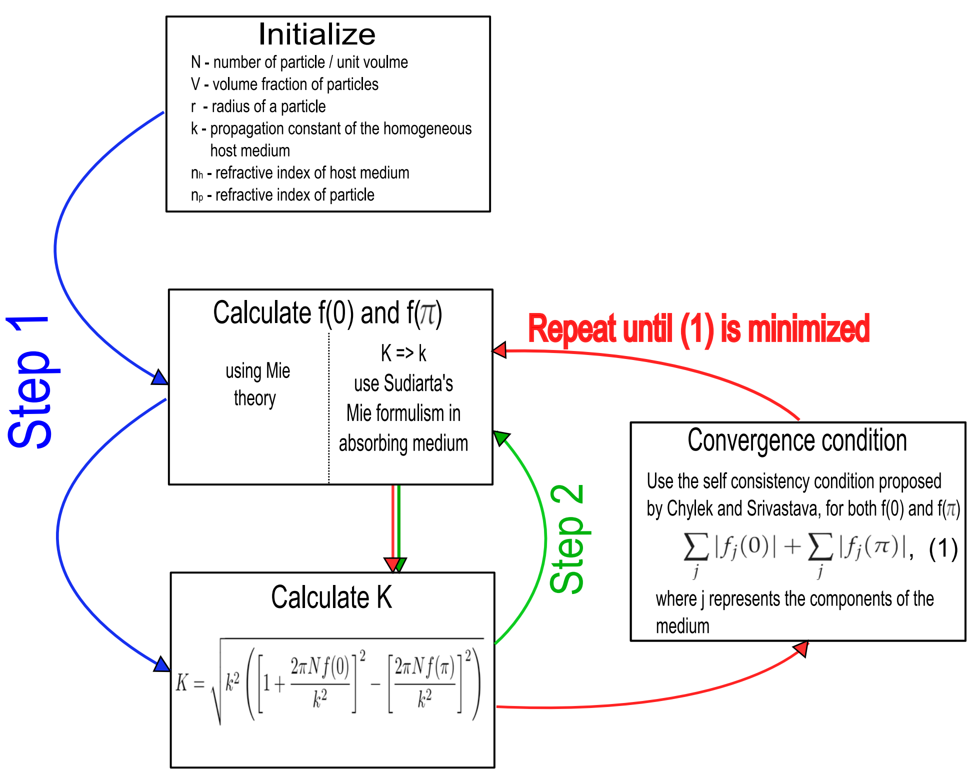

FIG. 1. Flow-chart illustrating the iterative algorithm based on Waterman-Truell approximation.

Chýlek and Srivastava,[13] which states that the forward scattering amplitude $f(0)$ vanishes if the components of the original system are placed back in the effective medium described by the effective propagation constant K. However, in our algorithm we also have the contribution from the backward scattering amplitude $f(\pi)$, accordingly the self consistency condition is modified such that when the components of the original system are placed back in the effective medium, both, $f(0)$ and $f(\pi)$ must vanish. Indeed, it is impossible to conceive a real physical situation in which the forward scattering amplitude of a single object disappears while the backward scattering amplitude still exists. Thus after the second step we calculate $\sum_j |f_j(0)| + \sum_j |f_j(\pi)|$ (where j represents the components of the original medium), and repeat the algorithm as shown in Fig. 2, until the value of $\sum_j |f_j(0)| + \sum_j |f_j(\pi)|$ is minimized.

We apply the iterative WT-EFA algorithm to estimate the frequency dependent scattering loss from three different granularities of polyethylene and air samples and compare the theoretical results with the transmission THz-TDS measurements of these samples. Three different granularities of spectroscopic grade PE powder from two different manufacturers (Sigma-Aldrich and Inducos) were used for our experiments. Two of the PE powders, one from each manufacturer, had relatively small PE grain sizes, with approximately 60 μm (Inducos) and 72 μm (Sigma-Aldrich) diameters, while the third one, again from Inducos, had a larger grain size of approximately 360 μm in diameter. The details of the setup, sample preparation, and internal structure dimensions are the same as described by Kaushik *et al.*[12] We carry out transmission measurements of the three samples. The background is modeled as pure PE with a relative permittivity of 2.13 and the air voids occupy 24%, 25%, and 44.5% by volume, and have a average radius of 24, 28, and 90 μm, for 60, 75, and 360 μm (PE particle diameter) samples, respectively. The effective propagation constant, K, is computed using the iterative WT-EFA algorithm illustrated in Fig. 1. Neglecting any intrinsic attenuation or absorption within the media, we can assume that the total attenuation here is entirely due to the scattering of the incident radiation, and can be obtained from $\alpha_{\text{eff}} = \Im(K)$, where $\Im(\cdot)$ indicates the imaginary part.

In this analysis, we assume a plane wavefront for the terahertz radiation, in the far field. Fig. 2 illustrates the propagation of THz radiation propagates through a sample cell during measurements of the sample and reference data. By analyzing the propagation geometry, and assuming that the reflections are removed from the sample and reference data, the transfer function is given by

$$H(\omega) = \frac{\tau_{\text{ws}}\tau_{\text{sw}}}{\tau_{\text{wa}}\tau_{\text{aw}}} \exp\left(-j(\hat{n}_s - n_0)\frac{\omega l}{c}\right), \tag{4}$$

where the subscripts a, s, and w are for air, sample, and window, respectively, l is the thickness of the sample, and τ represents the Fresnel transmission coefficients. Therefore, τ_{aw} represents the propagation from air to window, τ_{ws} represents the propagation from window to sample, etc. Here, n_0 is the refractive index of free air and $\hat{n}_s$ is the complex refractive index of the sample given by the formula $\hat{n}_s = n_s + jk_s$ with n_s and k_s representing the measured real part of refractive index and the extinction coefficient of the sample, respectively.

Thus using the above equation and the relation $\alpha_s(\omega) = 2k_s(\omega)\frac{\omega}{c}$, the optical attenuation $\alpha_s(\omega)$ can be found from

$$\alpha_s(\omega) = \frac{2}{l}\left\{\ln\left[\frac{\tau_{\text{ws}}\tau_{\text{sw}}}{\tau_{\text{wa}}\tau_{\text{aw}}}\right] - \ln|H(\omega)|\right\}. \tag{5}$$

For each sample, we compare the measured attenuation loss, calculated using Eq. (5), with the attenuation loss, obtained by applying the iterative WT-EFA algorithm. For the purpose of comparison, we also applied the iterative scheme suggested by Giusto *et al.*[5] to obtain the estimated scattering attenuation loss for the three samples. Fig. 3 shows this comparison. While small details of the attenuation are not captured by the simulation, the overall frequency dependent trends for the three media, as calculated by both algorithms, were in good agreement with the experimental data of the PE sample with average particle diameter 60 μm (air void

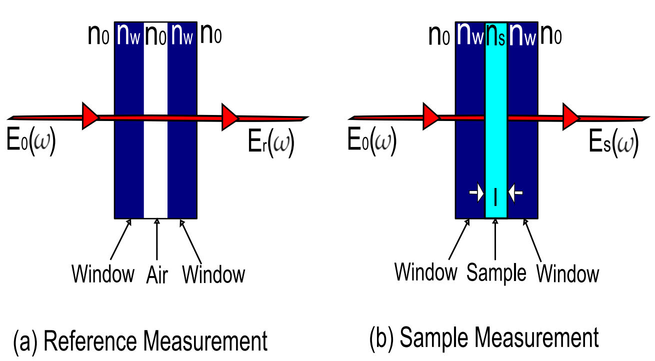

FIG. 2. (a) THz radiation propagates through an empty sample cell, as the reference. (b) THz radiation propagates through an identical sample cell, filled with the powder sample.

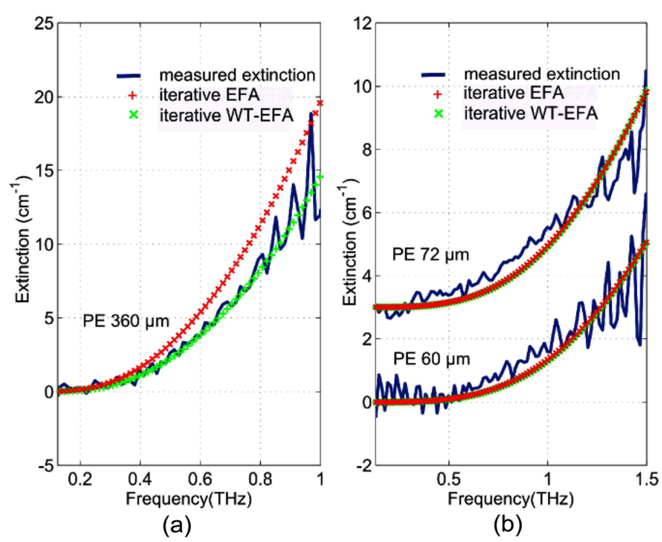

FIG. 3. (a) Comparison of measured (solid) and theory estimated (cross) frequency dependent total attenuation (extinction) loss for PE sample with average grain size of 360 m. (b) Comparison of measured (solid) and theory estimated (cross) frequency dependent total attenuation (extinction) loss for PE samples with average grain size of 60 m and 72 m (with a vertical offset of 3/cm for clarity).

diameter—48 μm) and PE sample with average particle diameter 72 μm (air void diameter–56 μm). However, for the sample with bigger PE particles, the estimations of the iterative WT-EFA algorithm show significant improvement in accuracy than the iterative EFA algorithm proposed by Giusto *et al*.[5] These results clearly indicate that when the dimensions of the scatterers are comparable to the incident wavelength, the scatterers can no longer be assumed to be point sources. As Foldy's approximation assumes the scatterers to be point sources, the two step algorithm proposed by Guisto *et al*.[5] fails to accurately estimate the scattering attenuation for the PE sample with the biggest scatterer dimension ($\approx$180 μm in diameter). Yet, our iterative WT-EFA algorithm using the theory of Waterman and Truell,[6] accurately estimates the scattering attenuation for all the three PE samples.

In conclusion, we propose an iterative algorithm using the multiple scattering theory of Waterman and Truell[6] and the self consistency condition of Chýlek and Srivastava,[13] for calculating the effective propagation constant. From the comparison of simulated and experimental results, it is found that the iterative WT-EFA algorithm reasonably estimated the optical properties of high density (>10%) samples made of non-absorbing granular PE particles of dimensions comparable to the incident wavelength.

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