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Evidence of Mie scattering at terahertz frequencies in powder materials

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We present experimental results as well as modelling concerning terahertz (THz) wave scattering by powder materials with grain size of the order of the wavelength. The studied material is a mixture of high density polyethylene and fructose powders, whose grain size is varied from a sample to another one while the volume concentration is kept constant. Experimental data are recorded with a THz time-domain spectroscopy set up. The scattering contribution to the total propagation loss in such powders shows a resonant behaviour for grain size roughly equal to the THz wavelength, for which it reaches 80%–90% of the total loss. Modelling using Mie theory allows us to well describe the absorption and scattering phenomena. © 2014 AIP Publishing LLC.
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During the last decade, many studies have been performed in the terahertz (THz) frequency domain in view of identifying suspect dangerous or prohibited materials like explosives¹ or drugs.² The identification of such materials is based on reliable spectral signature databases serving as references to compare the measured results with the data of the materials to be identified. In most of cases, these materials are absorbent, and thus, in order to be precisely characterized, they must be diluted in a transparent host medium, like high density polyethylene (HDPE) or poly-tetra-fluoro-ethylene (PTFE). Also, in the case of explosives or bacteria, the dilution makes the samples less dangerous than the pure material. Moreover, threat materials are often made of powders that can be either pure or mixed with a concealing substance. Previous studies have shown that scattering of THz waves by powder grains is responsible for both an overestimation of the absorption and a distortion of the spectral resonance peaks of the powder material. In materials with a low density of scattering particles, Shen *et al.*³ reported that the extinction coefficient of the mixture depends on the square of the THz frequency, whereas Franz *et al.*⁴ brought out a Christiansen effect in such inhomogeneous samples. On the other hand, multiple scattering process occurs in densely packed mixtures of dielectric powders, which can be modelled by the quasi-crystalline approximation.⁵ Apart from this latter case or from large non-spherical particles, Mie theory is generally used to well predict the size dependent effect of scattering in the THz region. Bandyopadhyay *et al.*⁶ successfully employed Mie theory to separate intrinsic absorption from scattering losses in different powder materials.

Here, we study, both experimentally and numerically, the THz scattering in moderately dense mixtures, namely mixtures of powders of fructose and HDPE. We mostly investigate the role of the size of the scattering fructose particles. We define the dimensionless size-parameter x as

$$x = \pi a / \lambda, \quad (1)$$

where a is the average diameter of the scattering particle and λ the relative wavelength in the medium ($\lambda = \lambda_0 / n$, λ_0 is the wavelength in vacuum and n is the refractive index of the mixture). We observe that the scattering contribution to the total value of the losses increases continuously with the particle size as far as x remains smaller than 2π . Over this value, the scattering contribution decreases. For small values of x , simple predictive models, like the model of the Christiansen effect proposed by Raman,⁴ allow one to nicely describe the scattering process, whereas it is no more the case for higher values of x , typically larger than 2π . Thus, a more rigorous theory, like the Mie theory, is necessary to fit the experimental results.

We use a mixture of fructose powder, which is the fingerprint material,⁷ and of HDPE (MIPELON XM-220), which serves as transparent host medium. The samples are fabricated by mixing the two components while keeping constant their mass ratio (50%–50%), corresponding to a volume concentration of about 30% of fructose and 70% of HDPE. Different pellets are fabricated using a hydraulic press (10 ton) and varying the fructose grain diameter a from 50 μm to 550 μm by step of about 50 μm . The fructose grains are sorted by size using a set of sieves. Typically, about 90% of the grain size is spread between the biggest and smallest sieve sizes (typically $\Delta a = 50 \mu\text{m}$) used to select the grains, while the 10% resting grains exhibit a smaller size. A fine analysis by microscopy reveals that the grains exhibit a square-like shape whose side length is typically 10% smaller than the size expected from the sieve selection process. The thickness of the pellets ranges from 0.5 to 1 mm to optimize the signal-to-noise ratio of the transmitted THz signal.

The samples are characterized using a classical transmission THz time-domain spectroscopy (THz-TDS) set up.⁸ The THz beam is collimated and focused onto the sample using a set of two parabolic mirrors. A similar optical scheme directs the transmitted THz beam onto the receiver. The THz detecting optics (numerical aperture 0.29) collects about 2% of the scattered THz signal in the case of a Lambertian scattering phenomenon. The optical parameters

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(refractive index n and absorption coefficient α) of the sample material are extracted from the measured THz transmission coefficient using a reliable numerical procedure.⁹ For scattering samples, the total experimental loss, evaluated from the straight transmission through the sample, arises from two contributions: the intrinsic absorption of the material and the scattering loss. We focus here our study on this latter contribution, i.e. on the scattering-induced overestimation of absorption. In our samples, there is no multiple scattering⁴ because we observe that the measured total loss coefficient does not depend on the sample thickness. Thus, the scattering loss can be modelled like absorption and the equivalent total absorption of the mixture writes:

$$\alpha_{total} = \alpha_{absorption} + \alpha_{scattering}. \quad (2)$$

α_{total} is directly obtained from THz-TDS measurements. Conversely, as most of scattered THz light does not reach the receiver, the scattering coefficient $\alpha_{scattering}$ cannot be measured. Nevertheless, $\alpha_{scattering}$ could be determined by estimating $\alpha_{absorption}$ and then subtracting its value from α_{total} (2). The estimation of $\alpha_{absorption}$ would require pellets of very small particles of PEHD and fructose in which scattering is supposed to be negligible. However, this procedure does not lead to a precise value of $\alpha_{absorption}$ as there is still a weak remaining scattering even with particles as small as some tens of micrometers.

The absorption spectra of our fructose-HDPE mixture samples are very similar to those given in Ref. 4, in which the effect of scattering is clearly seen, and thus are not reproduced here. In Fig. 1, we plot α_{total} versus the fructose grain diameter a for 4 frequencies in the spectral range of interest (0.5, 0.8, 1.2, and 1.5 THz). The choice of these frequencies is done for optimizing the precision of the measurement, taking into account both absorption and scattering. Indeed, at the absorption peak frequencies (1.32, 1.73, 2.15, and 2.68 THz)⁷ or at higher frequencies, the precision of the measurements is poor because of low transmission through the sample and/or a weak experimental dynamic. Dots in Fig. 1 are experimental data determined by transmission THz-TDS, while the continuous lines are their corresponding quadratic fit. At low grain diameters, α_{total} increases monotonously with the particles diameter and exhibits a quadratic behaviour whatever is the frequency. Because the mass ratio of the

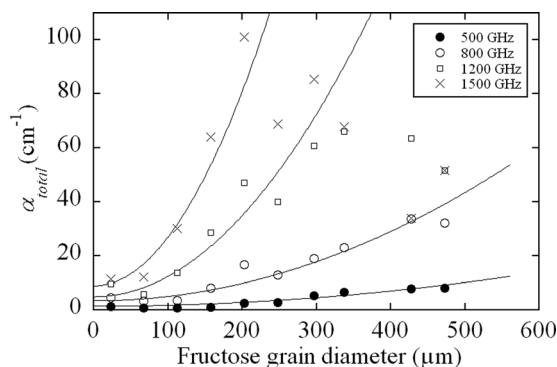


FIG. 1. α_{total} obtained from THz-TDS measurement (dots) versus the fructose grain diameter for 4 frequencies, together with quadratic fits (solid lines).

two components in the mixture is kept constant, the limit of the fit curve, when the fructose grain size tends to zero, permits to evaluate the intrinsic absorption of the mixture, i.e. without scattering loss.

When the grain diameter reaches higher values, α_{total} first saturates and then decreases with the grain diameter. The grain size a_{max} , for which α_{total} is maximum, depends on the frequency. Table I gives a_{max} and the corresponding size parameter value x for different frequencies. The x parameter is calculated using a refractive index of 1.58 for the mixture, measured for the sample with the smallest particles size. Indeed, in such a sample, even if scattering occurs, its influence on the value of the index of refraction remains very weak. The maximum of α_{total} is roughly reached for grain diameters corresponding to twice the wavelength. Therefore, for grain diameter typically smaller than the wavelength, simple models, like the one by Raman of the Christiansen effect, describe well the scattering efficiency while, for bigger grains, more sophisticated scattering theories are compulsory.

Here, we use the Mie theory¹⁰ as it is valid even for large size-parameter value. On the other hand, the calculation should be performed numerically, as there is no analytical expression of Mie scattering efficiency.

To evaluate the Mie scattering contribution to the loss, we employ a code based on the formalism by Bohren and Huffman.¹¹ This code computes the 4 Mie coefficients a_n , b_n , c_n and d_n that are, respectively, the efficiencies of extinction, scattering, backscattering, and absorption. In our study, we only focus on the scattering b_n and absorption d_n coefficients, whose sum corresponds to the experimentally evaluated α_{total} . The input parameters of the code are the fructose grain diameter a , the fructose volume ratio, its complex refractive index, the refractive index of the HDPE, which is supposed to be a transparent host material, and the wavelength in vacuum. Let us notice that the Mie theory is based on scattering by perfect spherical objects, whereas the fructose particles in our samples exhibit roughly a square shape, as explained previously.

The refractive indices of the materials used in the calculation have been measured using pure pellets of fructose and HDPE, with grain size smaller than a few tens of μm . For these samples, scattering is almost negligible. We measure $n_{HDPE} = 1.41$ and $\kappa < 10^{-2}$ ($\kappa = \alpha/(4\pi f/c)$) over the whole frequency range 0.1–2 THz. Absorption of HDPE is negligible and, thus, neglected as imposed by the code. For fructose, both refractive index and absorption depend on the frequency: Table II shows the measured values for the 4 frequencies of Fig. 1.

Let us notice that the refractive index of HDPE remains notably smaller than the one obtained for bulk material

TABLE I. Value of the size parameter x calculated at the maximum of α_{total} and using $n_{mixture} = 1.58$.

f (THz)	λ_0 (μm)	a_{max} (μm)	$x = \pi a/\lambda$
0.8	375	430	$1.8 \times \pi$
1.2	250	360	$2.2 \times \pi$
1.5	200	250	$2 \times \pi$

TABLE II. Refractive index and absorption values of pure fructose extracted from THz-TDS data and entered in the Mie scattering computation.

f (THz)	0.5	0.8	1.2	1.5
N	1.97	1.98	2.01	2.04
α (cm ⁻¹)	2.5	6	11	19

($n_{\text{HDPE}} = 1.51 \sim 1.54$).^{12,13} This smaller value is due to air inclusions in the samples. In our case, an estimation based on the pellet density leads to a volume ratio of about 16% of air, which is in good agreement with previously published results,¹⁴ if we consider that the diameter distribution of HDPE grains is centred on 30 μm . Moreover, using a Maxwell-Garnett effective medium approximation (EMA),¹⁵ the expected value of the effective refractive index of such air-HDPE mixture is 1.42, considering a bulk HDPE refractive index $n_{\text{HDPE}} = 1.51$. EMA can be then considered as sufficiently reliable to evaluate the effective refractive index of a mixture in the case of moderate scattering. But, this is not the case for absorption, which is more significantly affected by scattering than the refractive index.

Fig. 2 presents, for the 4 same frequencies as in Fig. 1, the measured α_{total} data versus the fructose grain size, together with the curves calculated with the Mie model considering two values for n_{PEHD} , namely 1.41 (continuous line) and 1.51 (dashed line). For $f = 1.5$ THz (Fig. 2(a)), the

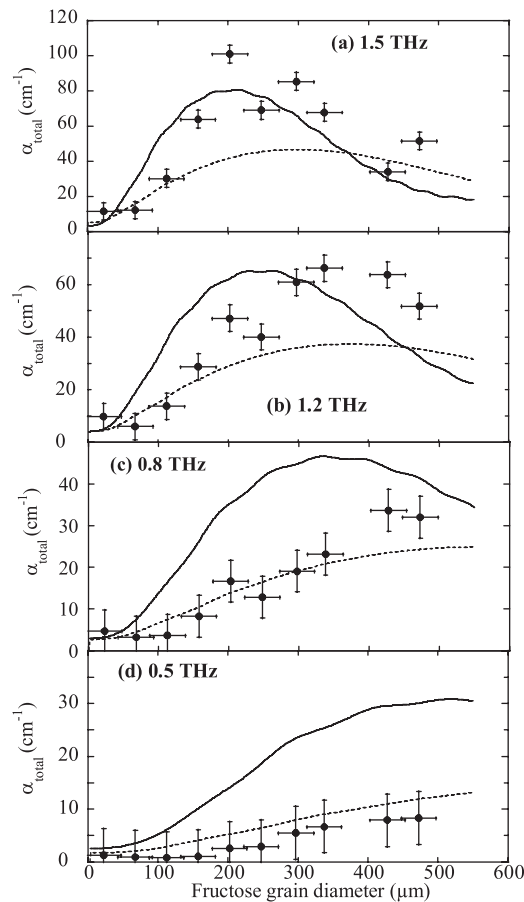


FIG. 2. α_{total} of the fructose-PEHD mixture versus fructose grains size: measured (dots) and calculated using Mie theory with $n_{\text{HDPE}} = 1.51$ (dashed line) and with $n_{\text{HDPE}} = 1.41$ (continuous line).

measured data distribution is bell-shaped. The maximum of absorption ($\sim 100 \text{ cm}^{-1}$) occurs when the grain diameter is equal to twice the wavelength in the grain material (fructose), which could be attributed to the contribution to the loss of a resonant scattering effect involving the grains. The measured absorption is larger than the one (9.3 cm^{-1}) calculated with the EMA model, which does not include any scattering contribution. This means that scattering contribution to the attenuation of the THz beam is by far more important than pure absorption in the fructose material. Similar to the 1.5 THz case, data recorded at 1.2 THz (Fig. 2(b)) exhibit a pronounced maximum around 350 μm and are distributed around the typical bell-shaped curve. For smaller frequencies, namely 0.8 (Fig. 2(c)) and 0.5 THz (Fig. 2(d)), the maximum seems to be shifted outside the experimental range, i.e., over $a = 500 \mu\text{m}$. It would have been interesting to reach the region where the grain size is of the order of millimetre. Unfortunately, such large grain size does not permit to manufacture flat samples of thickness smaller than 1 mm, which is compulsory to detect a signal in transmission. Moreover, the fructose re-crystallisation process leads to grains whose shape becomes flat, far away from the expected spherical one entered in the modelling.

Over the whole experimental spectrum, total absorption is always larger than pure absorption by the fructose grains as calculated with the EMA model. Total absorption and absorption in fructose tend to be equal when the fructose grain size is very small, which is expected as no scattering befalls with small grains.

The theoretical curves, calculated without any adjusting parameter, exhibit an overall shape similar to the experimental data. However, at higher frequencies (1.2 and 1.5 THz), curves calculated with $n_{\text{PEHD}} = 1.41$ (bulk value) are in better agreement with the measured values than with $n_{\text{PEHD}} = 1.51$. The order of magnitude of the total absorption is close to the measured one; however, the calculated curves are slightly shifted towards smaller grain sizes. On the contrary, at lower frequencies (0.5 and 0.8 THz), the best fit is obtained with $n_{\text{PEHD}} = 1.51$. Using the bulk $n_{\text{PEHD}} = 1.41$ value leads to an overestimation of the total absorption.

This observation does not agree with the EMA theory. Indeed, even if made under a high pressure, there are still some resting air inclusions in the pellets. Instead of solving the difficult problem of a mixture of 3 components (air, HDPE and fructose), we suppose here that the mixture behaves as if the air inclusions are located only in HDPE and, thus, lead to a decrease of the HDPE refractive index. This assumption should be especially true for wavelengths longer (lower frequencies) than the particles size, but this is not what is observed. Another tentative explanation could be found in the influence of absorption in fructose.

In Fig. 3 (here plotted for $f = 0.8$ THz), we qualitatively assess the impact of the fructose absorption value on the Mie theory prediction. Decreasing the absorption value of fructose in the calculation leads to a double effect (see Fig. 3): a reduction of the total absorption α_{total} and a shift of the curve toward larger grain values. Thus, for a smaller absorption in fructose, the theory-experiment agreement is better. This could be explained by an overestimation of the measured absorption in pure fructose. Indeed, the fructose absorption

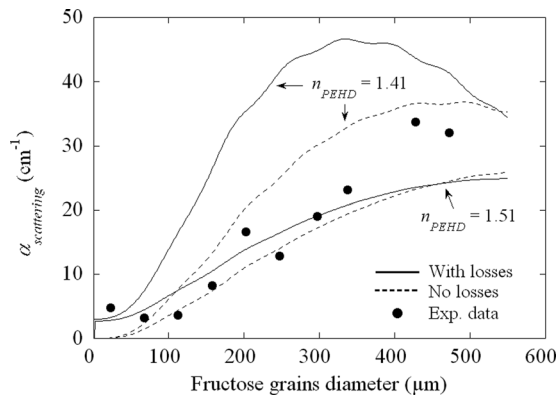


FIG. 3. α_{total} of the fructose-PEHD mixture 0.8 THz: measured (dots) and calculated using Mie theory taking into account intrinsic fructose absorption (dashed line) and without (continuous line).

values, used in the previous modelling (see Table II), were experimentally determined from pure fructose pellets whose diameter of grains is typically smaller than $50 \mu\text{m}$. Even in this case, scattering occurs because of air inclusions and lead to an overestimation of the absorption.

In order to estimate the relative weights of scattering and pure absorption on the propagation loss in the fructose-HDPE mixture, we define the relative scattering contribution to the loss by the ratio η

$$\eta \equiv \frac{\alpha_{scattering}}{\alpha_{total}} = \frac{\alpha_{total} - \alpha_{absorption}}{\alpha_{total}}. \quad (3)$$

We calculate η using the Mie theory code. Fig. 4 presents the relative scattering contribution η versus the particles diameter for the 4 studied frequencies. Continuous and dashed curves are calculated, whereas dots correspond to experimental data. As explained above, α_{total} is determined from direct THz-TDS data, while pure absorption of the mixture is derived from data measured for a mixture of very small particles (extrapolated at small fructose grain sizes from Fig. 1).

Like power attenuation (total absorption), scattering contribution η shows a resonant behaviour versus the grain size, whatever is the THz wavelength. Maximum of power attenuation in the samples occurs for particle diameter comparable to the wavelength. At this maximum, losses induced by scattering represent 80%–90% of the total measured losses.

In summary, we show that scattering effect can not be well predicted by the simple model when the size of the scattering particles is typically of the order or larger than the wavelength. In such a case, more sophisticated models like Mie theory become compulsory to qualitatively and quantitatively describe the scattering process and to evaluate its contribution to the THz power attenuation in the powders. For typical sub-millimetre size particles, the scattering

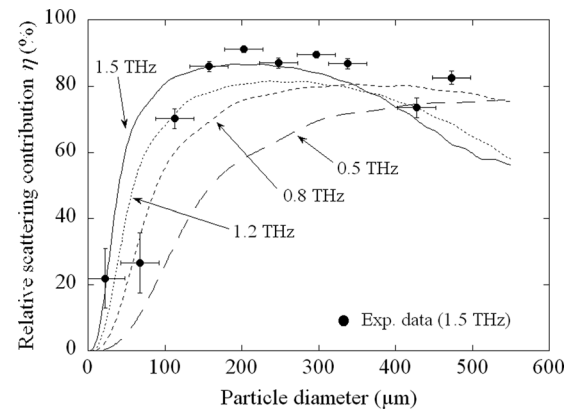


FIG. 4. Relative scattering contribution η versus the particle diameter calculated using $n_{PEHD} = 1.41$ (continuous lines) and directly evaluated from Fig. 1 (dots).

contribution reaches 80%–90% of the total power attenuation, even at frequency for which the particle material shows a rather strong absorption (here at 1.5 THz). The scattering contribution to the total losses greatly depends on the particles characteristics as size, distribution, shape, intrinsic refractive index, and absorption. Let us emphasize that, most of the time, the value of these parameters is unknown. In addition, air inclusions are always present in pellets of powders, even if strongly pressed, and they make the analysis even more complicated.

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