Study of Morphological Effects on Terahertz Spectra using Ammonium Nitrate

Amartya Sengupta, Aparajita Bandyopadhyay, John F Federici

Department of Physics, New Jersey Institute of Technology, Newark, New Jersey 07103, United States of America

Robert B Barat

Department of Chemical Engineering, New Jersey Institute of Technology, Newark, New Jersey 07103, United States of America

Abstract: The effect of morphology on Terahertz spectra in the region from 0.2 to 1.2 THz was studied using Ammonium Nitrate of different grain sizes. The results agree with Mie scattering theory for small grain sizes.

© 2005 Optical Society of America

OCIS codes: (290.5820) Scattering Measurements; (300.6500) Spectroscopy, time resolved

1. Introduction

Most of the solids under THz spectroscopic study have grain sizes which are comparable to the wavelength of THz waves and size dependent characteristics (like complex refractive index, optical depth) play a major role in determining the material transmission and extinction [1]. The usefulness of scattering measurements with respect to explosives in the THz region thus bears particular interest, as it would set the fundamental limitations on the absorption signature of these materials in the THz domain.

Ammonium Nitrate (AN), a well known fertilizer and a key ingredient in Ammonium Nitrate Fuel Oil (ANFO) bombs, was chosen for the current study as previous work in the THz range has shown that it has a monotonically increasing absorption spectra up to 1.5 THz in the pelletized form [2]. Thus any effect of scattering on the material transmission was expected to be apparent in the lower frequency range of 0.2 to 1.2 THz.

2. Experimental Methods

The experimental set – up for the study of morphological effects on THz spectra using AN followed the standard arrangement for THz-Time Domain Spectroscopy (THz – TDS) and is described in [3].

During the sample preparation, dry crystalline form of AN having an average grain size of 700 - 750 μm was used as the starting material and consequently grounded to two different samples of average grain sizes of 350 - 400 μm and 100 - 150 μm. Polyethylene envelopes were used as sample holders and the empty envelop was used as the reference for the measurements.

3. Results and Analysis

Time resolved THz spectroscopy provides both amplitude and phase information about the samples under study. In Fig. 2(a), THz electric field amplitude of the reference and the AN samples of three different morphologies. The phase shift gives the broadband refractive index of AN as 1.804 which compares well with the literature value of 1.816 [4]. The corresponding amplitude spectrum is shown in Fig. 2(b) which has sharp absorption dips at 0.57, 0.77 and 1.13 THz due to water vapor present in the atmosphere [5].
Fig. 2. Plots of the THz signal for the reference and samples in (a) time domain and (b) frequency domain

The transmission spectrum of AN samples were calculated using the Fast Fourier Transform (FFT) of sample and reference measurements:

\[
\text{Transmittance, } T(\omega) = \frac{|E_{\text{sample}}(\omega)|}{|E_{\text{reference}}(\omega)|}
\]  

(1)

Fig. 3(a) shows the transmission spectra of AN for different morphologies. A significant trend was observed where the width of the transmission spectra decreases with the increase in grain size of AN. A subsequent numerical fit in Fig. 3(b) revealed that the transmission has frequency dependence such that, when the grain size is small it follows the inverse square law. However, with increase in grain size the dependence tends to follow an inverse cube law. The numerical fit was generated using the Curve Fitting Toolbox in MATLAB where the two fitting coefficients have 95% confidence bounds.

Fig. 3. Plots of (a) THz transmission spectra for Ammonium Nitrate samples of different morphologies and (b) comparison of experimental and numerical fits for THz transmission

The extinction of a material is defined as the sum of intrinsic or true material absorption and the extrinsic losses associated with scattering. In this study, the extinction is defined as:

\[
\text{Extinction, } \varepsilon(\omega) = -\ln \left( \frac{|E_{\text{sample}}(\omega)|}{|E_{\text{reference}}(\omega)|} \right)
\]  

(2)

Fig. 4. Plot of material extinction for Ammonium Nitrate obtained in current study and comparison with Ref [2]
Fig. 4 shows the comparison of material extinction for AN in the current study with Ref [2]. A discrepancy at higher frequencies can be associated with the fact that the methods of sample preparation in the two cases were different and also the current experimental set–up is not optimized for higher frequencies.

Mie scattering theory suggests that the optical transmittance of a collection of spherical scatterers in the forward direction, that is in $\theta = 0$ direction depends inversely on the square of the frequency and grain size [6]. From the numerical fits, it can be seen that when the grain size of AN is small (100 – 150 $\mu$m), the result agrees well with the theoretical prediction. However, when the grain size increases, there is a deviation in the frequency dependence. This can be attributed to the fact that larger grains have arbitrary shapes which are not necessarily spherical and thus Mie theory does not appear to hold for them. In a more physical sense, one can argue that larger grains introduce the effects of dependent scattering, multiple scattering and other factors making the radiation transfer problem more complex. The future scope of this study would be to model this problem analytically using more involved scattering theories.

References