Development methodology definition diameter of particles in the composite medium using pulsed terahertz spectroscopy

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Abstract. In this paper, a method for determination of the diameter of spherical particles in composite medium on the basis of the Mie scattering theory using terahertz spectroscopy in the range from 0.1 THz to 1 THz was proposed.

1. Introduction
Terahertz (THz) light is a million times lower frequency than X rays and is non-ionizing. Therefore, it would be safe and desirable to use THz light for medical therapy and diagnostic purposes [1-4] for which it needs to develop new THz components [5-7]. Spectroscopic measurements using terahertz radiation are highlighted as they probe the scattering properties of granular media, which are sensitive to the packing structure. The wavelength of terahertz emission ranges from 30 μm to 3 mm and correlates with the particle size in many typical granular (composite) media. Many parameters obtained with angular scattering and spectral attenuation or in the measurement of reflection become sensitive to particle size and to other geometric characteristics of the samples, such as the degree of polarization or the intensity of scattering and damping. There is possibility of estimating the characteristics of composite media using THz Time-Domain spectroscopy (TDS). The applying of TD spectroscopy is not limited to the study of the quality of manufacturing of artificial media: it is used to analyzing both biological media and food products. A large number of studies make it possible to estimate the accuracy of the obtained results and the possibility of their application for improving quality control or monitoring of manufacturing process [8]. In this paper, the THz technique for analyzing composite media which contain spherical particles of a certain radius was proposed. The dependence of the scattering and absorption cross section on the radius of spherical particles for metals, semiconductors, and dielectrics was investigated based on the theory of Mie.

2. Theory
The analytical method is based on the Mie Theory. The Mie theory of the scattering of radiation on sphere in a homogeneous medium is based on the expansion of the electromagnetic field along the cylindrical harmonics and the “cross-linking” of the tangential components of the electric and magnetic field strengths at the boundary of the sphere. To satisfy these boundary conditions, it is necessary to assume that, in addition to the field incident on the sphere and the field inside the sphere, there is also the field of the secondary (scattered) wave [9]. Following formula (1) is used to calculate the cross section scattering of radiation for metal sphere according the Mie theory.
\[
\sigma_{\text{scatt}} = \frac{2\pi c^2}{\omega^2 \varepsilon_m} \sum_{n=1}^{\infty} (2n + 1) \left\{ |a_n(x, mx, m)|^2 + |b_n(x, mx, m)|^2 \right\},
\]
where \( c \) is the speed of light in vacuum, \( \varepsilon_m \) is the permittivity of the medium, \( \omega \) is the cyclic frequency of the incident radiation, \( a_n \) and \( b_n \) are the Mie coefficients. The formula (2) is used to calculate the coefficient \( x \)
\[
x = kr_s,
\]
where \( r_s \) is the radius of the sphere, \( k \) is the wave vector of radiation. Next formula (3) is used to calculate the coefficient \( m \):
\[
m = \sqrt{\frac{\varepsilon_s(\omega)}{\varepsilon_m}},
\]
where \( \varepsilon_s \) is the permittivity of the material of the sphere, \( \varepsilon_m \) is the permittivity of the matrix. The formula (4) and (5) used to calculate the Mie coefficients:
\[
a_n(x, y, m) = \frac{\psi_n'(y) \psi_n(x) - m \psi_n'(x) \psi_n(y)}{\psi_n(y) \zeta_n(x) - m \zeta_n'(x) \psi_n(y)}, \quad (4)
\]
\[
b_n(x, y, m) = \frac{m \psi_n(y) \psi_n(x) - \psi_n'(x) \psi_n(y)}{m \psi_n(y) \zeta_n(x) - \zeta_n'(x) \psi_n(y)}, \quad (5)
\]
where \( \psi_n(z) \) and \( \zeta_n(z) \) are the Debye functions. The formula (6) and (7) are used to calculate the Debye functions:
\[
\psi_n(z) = zJ_n(z) = \frac{\sqrt{\pi z}}{2} J_{n+\frac{1}{2}(z)}, \quad (6)
\]
\[
\zeta_n(z) = zH^{(1)}_{n+\frac{1}{2}(z)} = \frac{\sqrt{\pi z}}{2} H^{(1)}_{n+\frac{1}{2}(z)}, \quad (7)
\]
where \( J_n(z) \) is the spherical Bessel function, \( J_{n+1/2}(z) \) and \( H^{(1)}_{n+1/2}(z) \) are the functions of Bessel and Hankel for half-integral order. The formula (8) is used to calculate the cross-section of extinction. Extinction describes the decrease of the intensity of radiation as a result of absorption and scattering on particles.
\[
\sigma_{\text{ext}} = \frac{2\pi c^2}{\omega^2 \varepsilon_m} \sum_{n=1}^{\infty} (2n + 1) \text{Re}\{a_n(x, mx, m) + b_n(x, mx, m)\}. \quad (8)
\]
The formula (9) is used to calculate the cross section of absorption. The radiation absorption cross section is obtained from the difference between the extinction and scattering cross sections.
\[
\sigma_{\text{abs}} = \sigma_{\text{ext}} - \sigma_{\text{scatt}}. \quad (9)
\]
This theory can help analytically study the dependence position of the scattering peak and the radius of a spherical particle.

3. Materials and methods

In the experiment the setup, which scheme is shown in Figure 1, was used. Broadband pulsed THz radiation was generated using an undoped InAs crystal by irradiating it with femtosecond pulses of an ytterbium laser (wavelength - 1040 nm, pulse duration - 120 fs, pulse repetition frequency - 75MHz, power - 1 W). THz radiation had the following output characteristics: spectral range from 0.05 to 2 THz, average power - 30 uW, pulse duration - 2.7 ps. The main power was concentrated in the frequency range from 0.12 to 1.1 THz. THz radiation was generated by a gallium arsenide crystal in the magnetic system, then passed through a Teflon filter (which cuts wavelengths shorter than 50 um). After that, the radiation passed through the sample fixed in a focal plane perpendicularly to the beam. With the simultaneous incidence of the femtosecond probe beam and THz beam passed through the
sample on the electro-optical CdTe crystal, THz pulse induced birefringence of the probe beam in the crystal due to the electro optical effect. The birefringence magnitude is directly proportional to the intensity of terahertz wave electric field in the time point $E(t)$ [10].

![Figure 1](image1.png)

**Figure 1.** Schematic diagram of the set up (FL-1 - femtosecond laser based on potassium-yttrium tungstate crystal activated with ytterbium (Yb: KYW), generating femtosecond pulses; $F_{1,2}$ - a set of teflon filters for IR wavelength range cutting off, BS - beamsplitter, DL - optical delay line, $M_{1,2,3}$ - mirrors, Sam - investigated sample, Wol - Wollaston prism, CdTe - electro optical cadmium-telluric crystal, BD - balanced detector, LA - lock-in amplifier, PC - personal computer, GTP - Glan-Taylor prism, $PM_{1,2}$ - parabolic mirrors, Ch - chopper.

The experiment structure was shown in Figures 2-3. This structure was made of VisiJetM3 Crystal. There are hemispherical depressions of various diameters: 0.2 mm, 0.4 mm, 0.6 mm, 0.8 mm, 1 mm and one section of the structure without depressions.

![Figure 2](image2.png)

**Figure 2.** Image of model the experimental structure

![Figure 3](image3.png)

**Figure 3.** Photo of the experimental structure
During the experiment, the structure was arranged in such a way that the radiation fell on the region in which the spheres of the same diameter were. Knowing the diameters of spherical particles in each region, it is possible to experimentally verify the size of spherical particles in a given composite medium. Since the structure itself should be an analog of the composite medium, the two printed plates of the experimental structure were compressed in such a way that the spherical particles of air were inside the structure.

4. Results and analysis
Using the Mie theory the dependences of the resonance frequency on the radius of air spherical particles for 3 kinds of plastic (TPX, PLA, VisiJetM3 Crystal) were calculated. After carrying out analytical studies the graph of the dependence of the scattering peak frequency on the radius of spherical particles in the experimental medium (Figure 4) was plotted, the obtained graph can be approximated into an exponential dependence. According to the graph, one can see that as the particle diameter increases, the resonant frequency of the scattering peak shifts to the low-frequency range. As a result, the radius of the particles was determined from the experimentally obtained frequency of the scattering peak, using the available analytical calculations.

![Graph showing the analytical dependence of the scattering peak frequency on the radius of air particles in the matrices](image)

**Figure 4.** Analytical dependence of the scattering peak frequency on the radius of air particles in the matrices

When analyzing the reflection spectrum of the composite medium, the reflection peak was found to be at a frequency corresponding to particles of smaller radius. With the help of the conducted analytical modeling it was revealed that the peak for the sector with spherical particles with a diameter of 1 mm is at a frequency corresponding to particles with a diameter of 0.8 mm. Using a microscope, the diameter of the spherical particles was measured, and it turned out that their diameter was not 1 mm, but 0.8 mm (Figure 5). Thereby, it can be argued that this technique can be used to determine the diameter of particles in composite media by means of pulsed terahertz spectroscopy. As a result of the experiment and the analysis of the results obtained using the developed methodology, it was found that the fabricated structure did not correspond to the simulated structure: the particle radii were less than expected.
Figure 5. The reflection spectrum of experimental structure for particles with diameter of 1mm and the scattering cross-section calculated using the Mie theory for particles with diameter of 0.8 mm

5. Conclusion
The Mie theory makes possible to estimate the diameter of spherical particles in matrix using the frequency position of the scattering and absorption peak. In this paper, the diameter of particles in dielectric medium was measured using THz time-domain spectroscopy based on Mie theory.

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