Determination of nanoscale inhomogeneities in scattering and absorbing medium

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Abstract. In this paper, we propose an original method for determining the size of nanoscale inhomogeneities in scattering and absorbing medium by comparing the theoretical and experimental dependences of the transmission medium on the wavelength of light. The method was tested on a sample of nanoporous glasses and gave satisfactory results. Were also carried out numerical evaluation of this phenomenon by using the software package CST Microwave Studio.

1. Introduction

Determination of the characteristic size of inhomogeneities of the medium is an important scientific and technical challenge that attracts the attention of many researchers [1,3,4,11]. Currently known methods of solving this problem are often unsuitable because of its complexity or high cost (for example, small-angle x-ray scattering). We have proposed a simple and cheap method for determining the size of the inhomogeneities in scattering and absorbing media. The method was tested on a sample plate of absorbing nanoporous, in which nanoscale inhomogeneities (pores with an average diameter of 17 nm, filled with water) served as the scatter-particles.

To implement this method was obtained by the theoretical dependence of the transmittance of the scattering and absorption coefficients and the experimental dependence of transmittance on the wavelength of light.

The main idea of this method is as follows: at long wavelengths the absorption of the medium becomes negligible and the attenuation of light occurs due to scattering by inhomogeneities. Thus it is possible to determine experimentally the scattering coefficient by examining the transmission of the plate in the range of longer wavelengths. Since the characteristic size of inhomogeneities of the medium is much smaller than the wavelength, the scattering is Rayleigh in nature. Consequently, the scattering coefficient is inversely proportional to the fourth power of wavelength. Further, the proportionality coefficient and the absorption coefficient chosen in such a way as to ensure the best agreement between theoretical and experimental dependences of the scattering coefficient on wavelength. Since Rayleigh scattering is character, then the resulting proportionality factor depends on the sixth degree of pore diameter. From the results obtained is determined by the average pore size.

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2. Theoretical derivation of the transmission coefficient of the system parameters

To receive our dependence transmittance from scattering and absorption coefficients, one must understand the process of light propagation in scattering and absorbing medium. We first derive for this equation of radiation diffusion.

Assuming that the size of scattering particles is much smaller than the wavelength of light [4], the radiative transfer equation has the form:

\[ \frac{\Omega \nabla I}{I} = - (\gamma_a + \gamma_s)I + \frac{\gamma_s}{4\pi} \int I d\omega, \]

(1)

Where \( I \) - the intensity of radiation, \( \Omega \) - the unit vector in the direction of the beam, \( \gamma_a \) and \( \gamma_s \) - absorption and scattering coefficients of the medium (which are assumed to be constant), \( d\omega \) - an element of solid angle. The value of a constant damping system \((\gamma_a + \gamma_s)^{-1}\), that is a characteristic spatial scale of intensity. We assume that this quantity is small compared with typical dimensions of the system. We introduce the total intensity of radiation incident on this element of the medium:

\[ Q = \int I d\omega \]

(2)

and apply to (1) Fourier transform. Then we have:

\[ ik\Omega I_k = - (\gamma_a + \gamma_s)I_k + \frac{\gamma_s}{4\pi} Q_k, \]

where \( I_k = \int e^{-ikx} d^3x \). Thus:

\[ I_k = \frac{\gamma_s}{4\pi} \frac{Q_k}{\gamma_a + \gamma_s + i k \Omega}. \]

(3)

Integrating (3) of the total solid angle and using (2), we obtain:

\[ Q_k = \frac{\gamma_s}{4\pi} Q_k \int \frac{d\omega}{\gamma_a + \gamma_s + i k \Omega}, \]

or:

\[ 1 - \frac{\gamma_s}{k} \text{arctg} \left( \frac{k}{\gamma_a + \gamma_s} \right) Q_k = 0, \]

(4)

where \( k = |k| \). Equation (4) represents the Fourier-representation of pseudodifferential equation [7] for the value \( Q \). In the short-wave approximation \( k \ll 1 \), this equation becomes:

\[ \frac{1 + k^2 \gamma_s}{3 \gamma_a \gamma_s} Q_k = 0. \]

(5)
Confine ourselves to the media in which the scattering coefficient $\gamma_s$ is much higher than the absorption coefficient $\gamma_a$. Then, moving (5) in coordinate representation, ie performing the inverse Fourier transform, we obtain the desired equation of radiation diffusion [4]:

$$\nabla - 3\gamma_a\gamma_s \overrightarrow{Q} = 0. \quad (6)$$

Under the same assumptions can be obtained by the boundary condition to equation (6):

$$Q - \frac{1}{2\gamma_s} \frac{\partial Q}{\partial n} = 2Q_e, \quad (7)$$

where $\frac{\partial}{\partial n}$ - derivative along the outward normal to the area, $Q_e$ - the total intensity of radiation incident outside the border area.

2.1. Transmittance equation

Now we get directly the transmittance of the medium in the case of plane-parallel plate nanoporous glass of thickness $h$. Then we can consider the change in light intensity in only one spatial direction (along the axis $x$).

According to (6), (7), the diffusion equation and radiation boundary conditions for him are as follows:

$$\frac{d^2 Q}{dx^2} - 3\gamma_a\gamma_s Q = 0, \quad (8)$$

$$\left[ Q - \frac{1}{2\gamma_s} \frac{dQ}{dx} \right]_{x=0} = 2Q_e, \quad \left[ Q + \frac{1}{2\gamma_s} \frac{dQ}{dx} \right]_{x=h} = 2Q_e. \quad (9)$$

Integrating (8) using (9), we obtain

$$Q = A ch(x) + B sh(x), \quad (10)$$

where

$$A = \frac{sh + \beta ch}{+ \beta^2 sh + 2\beta ch} 2Q_e, \quad B = \frac{ch + \beta sh}{+ \beta^2 sh + 2\beta ch} 2Q_e,$$

$$\alpha = h\sqrt{3\gamma_a\gamma_s}, \quad \beta = 0.5\alpha/\gamma_s$$

Knowing $Q$, we can determine the intensity of the radiation propagating at an angle $\theta$:

$$I = \frac{1}{4\pi} \left( Q - \frac{\cos\theta}{\gamma_s} \frac{\partial Q}{dx} \right) \quad (11)$$

and the total intensity of radiation propagating in the positive direction along the axis $x$:
Using (10), (11), (12), we calculate the desired transmittance $T = \frac{Q_+}{Q_+^\text{plate nanoporous glass}}$:

$$T = \frac{1}{sh \left( \sqrt[3]{3\gamma a\gamma_s} \right) - ch \left( \sqrt[3]{3\gamma a\gamma_s} \right)}.$$  \hspace{1cm} (13)

From this relation together with the experimental results of our proposed method to determine the characteristic pore size.

3. Experimental details

For the experimental measurement of the transmission coefficient of the sample scattering and absorbing medium, as a model object was taken polished disc nanoporous glass NPS-17 15 mm in diameter and 1 mm thick, with an average pore size of 17 nm and a relative pore volume of 58%.

Before the measurements the pores were filled with water. For this sample has been submerged for days in distilled water. The measurements were performed on a standard spectrophotometer Evolution-300. Further measured the transmittance of the sample at various wavelengths in the range 350 - 1100 nm. The measurement results are shown in Fig. 1.

Nanoporous matrices based on silica glass are a unique tool for studying physical and chemical processes to a limited extent commensurate with the scale of the processes and the size of the objects being studied. Space constraints, and effective contact with the pore walls cause significant features of the condition and properties of the filling material compared with the case of its location in the void volume.

The use of nanoporous glasses at the moment, mainly due to the transparency of these glasses in the visible spectrum and the possibility of obtaining samples of optical quality [8,9]. Optical density of samples in the near UV spectral region due to scattering and absorption in the structure of the material.

When considering the optical properties of porous glasses in the visible and near-infrared absorption in the samples can be neglected because of the smallness of this quantity, and the effective optical constants of samples are determined only by attenuation due to scattering by the porous structure and its irregularities. This property of nanoporous glasses and formed the basis of this method.

4. Results and discussion

The experimental and calculated dependences of the transmission and absorption are shown in Fig. 1.

Scattering coefficient is inversely proportional to the fourth power of the wavelength of incident light $\gamma_s = \frac{C}{\lambda^4}$. Coefficient $C$ and the spectral absorption coefficient $\gamma_a$ chosen so as to ensure the best agreement between theoretical and experimental dependences of transmittance on the wavelength of light. Obtained by this coefficient $C$ can approximately estimate the characteristic size of inhomogeneity $D \sim C^{1/6}$.
Estimated value of the average pore diameter \( D = 13.5 \) nm close to the average pore size (17 nm) nanoporous glass. This suggests that the proposed method has a relatively high degree of accuracy in its simplicity and clarity.

5. Numerical Simulation

Numerical simulation using the software package CST Microwave Studio has qualitatively confirmed these results in the approximation of exponential dependence of the medium attenuation coefficient of the wavelength. A small element of the medium (glass), filled the pores with water by 58% in volume terms, was simulated in this package. After this item was held incident electromagnetic wave (port 1 in Fig. 2). Part of the electromagnetic wave, which passing through this element, is recorded on the other side in port 2. Further, the results were processed and compared with theoretical and experimental dependences.

Fig. 1. Experimental and calculated dependence of the transmission and absorption of nanoporous glass on the wavelength of light.

Fig. 2. A small element of the medium (glass), filled the pores with water by 58% in volume terms.
In Fig. 3 is shown numerical dependence of the transmission coefficient, except the wavelength 432 nm (T = 0.80), 1017 nm (T = 0.86) and 1064 nm (T = 0.67), since at these wavelengths are contained plasmon modes, associated, as expected, from the Fano antiresonances. These antiresonances were observed in the numerical simulation due to the not randomized structure (Fig.2), which in a real experiment can’t be observed, and therefore not of interest for our case.

Fig. 2. Qualitatively confirmation of results in the approximation of exponential dependence of the medium attenuation coefficient of the wavelength.

6. Conclusion
Proposed a method for determining the average size of characteristic inhomogeneities of the medium. The method was tested on a sample of nanoporous glasses and gave satisfactory results. Carried out numerical simulation of the process, which also turned out to be in good agreement with experiment. Thus, the method has relatively good accuracy at low cost and complexity, but also allows us to understand the process of light propagation in scattering and absorbing medium.

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