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Passive Measurements of the Refractive Index of Liquid Nitrogen and Free-flowing Substances by the Prism Method in the Millimeter Wavelength Range

Valery A. Golunov, Konstantin V. Gordeev, Konstantin N. Rykov

Kotelnikov Institute of Radioengineering and Electronics of RAS, Fryazino Branch, <http://fire.relarn.ru/>
Fryazino 141190, Moscow region, Russian Federation

E-mail: golsnow@mail.ru, gordeev@yandex.ru, constrykov@yandex.ru

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Abstract: It is proposed to use the classical prism method in the millimeter wavelength range for measuring the refractive index of liquid and free-flowing substances, including mixtures with particle sizes comparable to the wavelength. The method is implemented using a hollow radio transparent rectangular prism filled with a test substance. The measurements were carried out in the thermal radiation mode using radiometers with horn-lens antennas at frequencies of 37.5 and 94 GHz. To measure the deflection of the refracted beam, a linear scanner with a black body mounted on it, cooled with liquid nitrogen, was used. The distance between the prism and the scanner was 1 m. The refractive index of liquid nitrogen, sand, gravel, marble chips and granular polyethylene were measured. Using the refractive formula and the Landau-Lifshitz-Loeong formula for calculating the dependence of the refractive index of binary mixtures on the bulk density of particles, estimates of the refractive index of the material of the particles that make up the substances under study are obtained. They are in satisfactory agreement with the known experimental data for quartz, feldspar, granite, and marble.

Keywords: prism method, refractive index, millimeter waves, thermal radiation, mixtures of particles commensurate with the wavelength

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1. INTRODUCTION

The refractive index is one of the most important characteristics of solid, liquid and gaseous media that determine their electrodynamic properties [1]. To measure the refractive index, a variety of methods have been developed that take into account both the features of the media under study and the technical capabilities of the used

wavelength range. For example, in the range of millimeter (MM) waves, methods are used that are implemented using active waveguide, resonator, quasi-optical and open systems [2-7]. In the optical range, since Newton's time, the prism method has been widely used, based on its property, due to refraction, to decompose white light into spectral components.

The possibility of evaluating the dielectric constant (DC) of a solid from the measured values of the permittivity of a mixture of its particles with air, and vice versa, is of certain practical importance. The widely used formulas for calculating the dependence of the DC of binary mixtures on the volume density of particles, such as Maxwell Garnett, Polder-van Santen, were developed within the framework of the effective medium theory in the approximation of particles small in comparison with the wavelength [8,9]. This condition is satisfied, for example, by the results of measurements performed on coaxial, waveguide and resonator devices, since negligible particle sizes are a necessary condition for correct measurements using these devices. However, even the fulfillment of this condition does not guarantee satisfactory agreement of the experimental data with the results of calculations performed according to any of the above formulas. The reason is that at average values of the volume density, these formulas lead to diverging values of the dielectric constant [10].

With increasing particle size, multiple scattering occurs between individual particles, that affects the dielectric properties of the medium. However, strict accounting of the scattering effect, carried out within the framework of the theory of strong fluctuations [11,12], the quasicrystalline approximation (QCA), and the quasicrystalline approximation with a coherent potential (QCA-CP) [13], insignificantly increases the permissible ratio of particle size to wavelength. When MM waves propagate in such granular media as

sand, snow, gravel, the particle sizes can be commensurate with the wavelength that, on the one hand, can lead to the appearance of the multiple scattering effects, and, on the other hand, limits the use of waveguide and resonator devices for measuring their DC. In this work, to measure the refractive index of liquid and granular weakly absorbing media, including mixtures with particles whose sizes are commensurate with the wavelength, a variant of the implementation of the prism method on MM waves in the passive mode is proposed. Liquid nitrogen, sand, granular polyethylene, marble chips and gravel were used as media under study. Refractive index was measured at 37.5 and 94 GHz.

2. SCHEME, TECHNIQUE AND EQUIPMENT FOR MEASUREMENTS

Using the refractive properties of a prism to measure the refractive index of substances assumes that these substances have a prismatic shape. If, in the study of solids, such a form is made due to their appropriate processing, then in the study of liquids and loose substances, a preliminary preparation of a prismatic container is necessary. In the proposed study, in order to give such samples the required prismatic shape, triangular hollow prisms were created with input and output edges for radiation, made of 20 mm thick Penoplex foam sheet, which provided both the necessary stiffness of the prism and almost absolute transparency for MM waves. The base of the prism is made of wood in the shape of a right-angled triangle. A drawing of a prismatic cell is shown in **Fig. 1**.

Fig. 2 shows the path of rays in a rectangular prism filled with a substance with a refractive index n at normal incidence of a plane wave from above. The angle of incidence on the lower face α and the angle of refraction $\alpha + \beta$ are related by the Snell relation:

$$n \sin \alpha = \sin (\alpha + \beta) \quad (1)$$

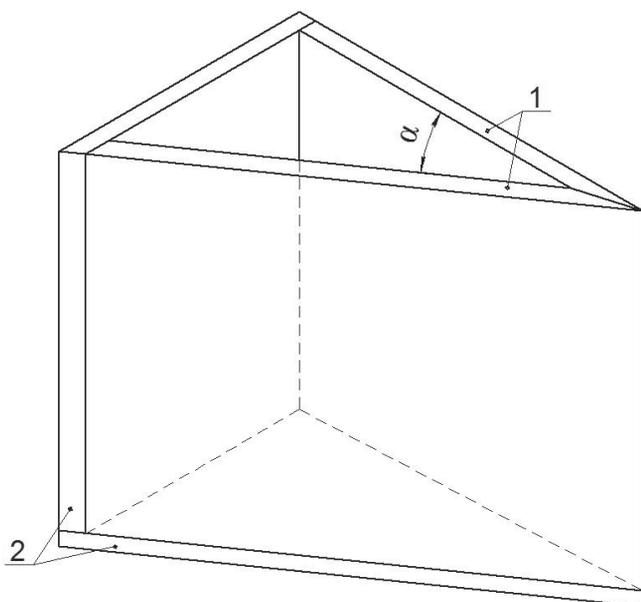


Fig. 1. Rectangular prism design: 1 – foam, 2 – wood.

At a distance $OB = L$ from the refracting face, the beam is displaced in the transverse direction by a distance $AB = x$. If L and x are known, then the refractive index in accordance with (1) is determined as follows:

$$n = \frac{\sin(\beta + \alpha)}{\sin \alpha} = \frac{\sin[\arctg(x/L) + \alpha]}{\sin \alpha}. \quad (2)$$

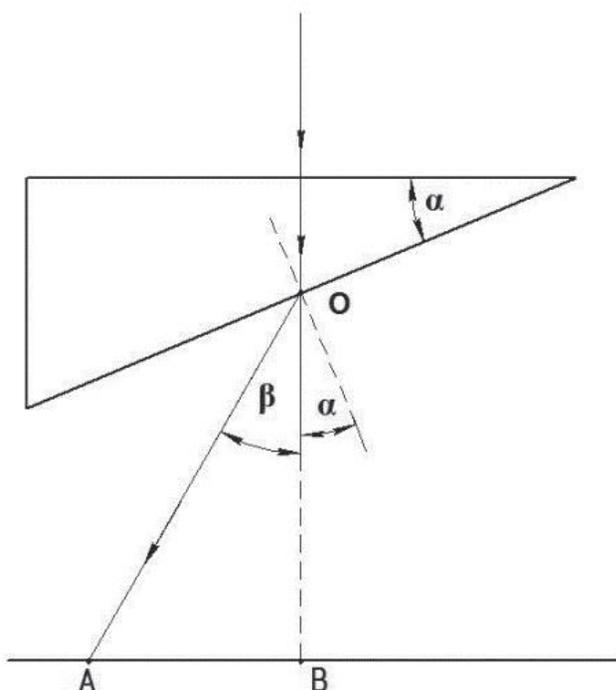


Fig. 2. Ray path in a prism.

Relation (2) is, in fact, a classical algorithm for measuring the refractive index of substances. The novelty of the measurement technique proposed below is the implementation of the algorithm in the thermal radiation mode.

The measurement scheme is shown in Fig. 3. The receiving system consisted of a radiometer with a horn-lens antenna with an aperture of 180×160 mm and a lens focal length of 400 mm. As a radiation source, a black body (BB) with a size of 100×200 mm, cooled with liquid nitrogen, was used. The BB was installed on a moving platform of a linear scanner with a stroke length of 1 m. Scanning was carried out perpendicular to the optical axis of the antenna at a distance of up to 1.5 m from a prism mounted almost close to the lens. The dimensions of the prism face adjacent to the lens were 250×200 mm. To study substances, depending on their dielectric and attenuating properties, three prismatic cells were used, differing only in the values of the angle α (10° , 25° , and 40°) shown in Fig. 1.

The BB thermal radiation was received by broadband radiometers with sensitivities no worse than 0.2 K at a time constant of 1 s, operating at 37.5 and 94 GHz. The output signals of the radiometers were recorded using an ADC and a computer.

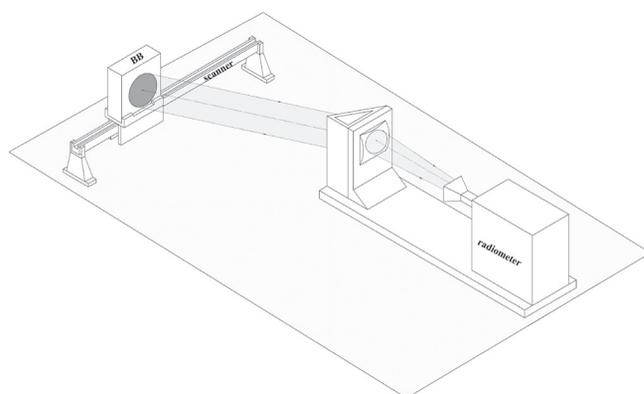


Fig. 3. Refractive index measurement circuit.

3. MEASUREMENT RESULTS

The prism cell, as noted above, is designed to measure the refractive index of liquids and loose materials. Let us first consider the results of measuring the refractive index of liquid nitrogen, the dielectric properties of which are of particular interest in connection with its wide application in radiometry for cooling black bodies used to calibrate radiometers. Fig. 4 shows the records of the output signals of the radiometers at frequencies of 37.5 and 94 GHz when the BB moved along the perpendicular to the optical axis of the lens antenna (see Fig. 3). At each frequency, two records of the output signal were performed, one of which (calibration) corresponded to the case of a hollow prism, the other - the case of the prism filled with liquid nitrogen. The cell angle $\alpha = 25^\circ$, the distance L at 37.5 and 94 GHz was equal to 1.28 m and 1.31 m, respectively (see Fig. 2).

The displacements x of the refracted rays relative to the direct rays, determined as the difference between the maximum of the corresponding output signals of the radiometers, were 11.55 cm at 37.5 GHz and 11.84 cm – at 94 GHz. As a result of calculations using formula (2) at both frequencies, practically coinciding values

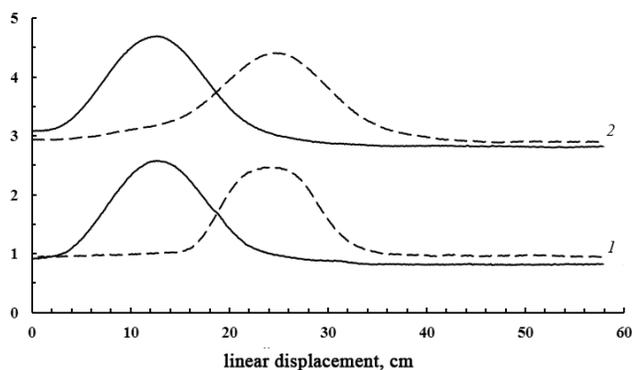


Fig. 4. Output signals of radiometers at frequencies of 37.5 GHz (1) and 94 GHz (2) when the FT moves along the perpendicular to the optical axis of the lens antenna with an hollow prism (solid lines) and prism is filled with liquid nitrogen (dashed lines).

Table
Experimental values of the refractive index of free-flowing substances

	Substance	Particles size, MM	n	ρ_v	ρ , g/sm ³	ρ_0 , g/sm ³	α , angular degree
1	sand	<0.5	1.67	0.67	1.67	2.49	10
2	sand	<1	1.62	0.62	1.59	2.4	10
3	gravel	<1	1.64	0.59	1.55	2.63	10
4	gravel	1-2	1.62	0.56	1.55	2.8	10
5	gravel	3-4	1.665	0.57	1.65	2.9	10
6	marble	1-2	1.748	0.48	1.28	2.65	10
7	polyethylene	3	1.34	0.6	0.575	0.96	30
8	polyethylene	4	1.32	0.64	0.615	0.96	30
9	polyethylene	5	1.23	0.4	-	-	40
10	polyethylene	5	1.32	0.625	0.6	0.96	30

of the refractive index of liquid nitrogen were obtained, equal to $n = 1.189$, which corresponds to the value of the real part of the DP $\epsilon' = 1.41$. The fact that equal values of n were obtained at both frequencies was to be expected, since nitrogen, as it is known, is a neutral gas. For comparison on the Internet for liquid nitrogen values $\epsilon' = 1.4 \dots 1.5$ are given, but there is no information on the methods used to measure DP.

The **Table** lists the values of the refractive index of sand, gravel, marble chips and granular polyethylene measured at 37.5 GHz. In addition, the table lists the values of the volume density of the particles ρ_v , the density of the mixture ρ , the density ρ_0 of the solid material of the particles, and the angle of the prism α at which the measurements were carried out.

The density of the solid material of particles ρ_0 and their volume density ρ_v are related by the ratio:

$$\rho_0 = \frac{\rho}{\rho_v}, \quad (3)$$

where ρ is the density of the mixture, defined as the ratio of the mixture mass M to its volume V_0 :

$$\rho = \frac{M}{V_0}. \quad (4)$$

Thus, if the density of the particle material is known, then the bulk density is determined using the results of measurements of M , V_0 and relations (3)-(4). This approach was used to calculate the volume density of marble chips and polyethylene, since their density has been known.

The volume density of sand and gravel, which could include various minerals with an unknown concentration, was determined as:

$$\rho_V = \frac{V_0 - V_w}{V_0}, \tag{5}$$

where V_0 is the total volume of the mixture, V_w is the volume of pores determined by method of filled with water.

In the theory of random dense media and applications requiring a wide choice of density, a large set of mixing rules was introduced [10], for example, by writing the "power law" approximation, which in the case of a mixture of particles with air is written in the form:

$$\epsilon_{eff}^\eta = 1 + \rho_V (\epsilon_m^\eta - 1), \tag{6}$$

where ϵ_{eff} is the effective DP of the mixture, ϵ_m is the DP of the particle material, η is the exponent. Validation of formulas for mixtures is carried out by comparison with a radiophysical experiment. From a large number of published works, we single out studies [14,16], in which it was shown that the experimental dependences $\epsilon_{eff}(\rho_V)$ both in the case of liquid and powder binary mixtures are best approximated using the refraction formula [17] and the Landau-Lifshitz-Looyenga formula [18,19].

The refraction formula is written in the form (6) at $\eta = 1/2$:

$$\sqrt{\epsilon'_{eff}} = 1 + \rho_V (\sqrt{\epsilon'_m} - 1). \tag{7}$$

For weakly absorbing particles $\epsilon'_m \gg \epsilon''_m$, therefore $\sqrt{\epsilon'_{eff}} = n_{eff}$, $\sqrt{\epsilon'_m} = n_m$ and (7) is transformed to the form:

$$n_{eff} = 1 + (n_m - 1)\rho_V. \tag{8}$$

Thus, the refractive model corresponds to the linear dependence of the refractive index of the mixture on the volume density of the particles, while the proportionality coefficient equal to $(n_m - 1)$ is determined by the refractive index of the material of the particles. Therefore, by measuring the refractive index of the mixture with known volume density of the particles, the refractive index of the particles material can be determined.

The Landau-Lifshitz-Looyeng formula is applicable for a mixture of dissimilar particles with an arbitrary shape and is written in the form (6) at $\eta = 1/3$:

$$\epsilon_{eff}^{1/3} = 1 + \rho_V (\epsilon_m^{1/3} - 1)$$

or

$$n_{eff} = [1 + \rho_V (n_m^{2/3} - 1)]^{3/2}. \tag{9}$$

It follows from (9) that the refractive index depends nonlinearly on the volume density. Fig. 5 shows the experimental data obtained in this work and plots of the dependence of the refractive index on the volume density of particles, calculated using formulas (8) and (9). It is seen that the nonlinearity of the dependence of the refractive index on the bulk density, corresponding to formula (9),

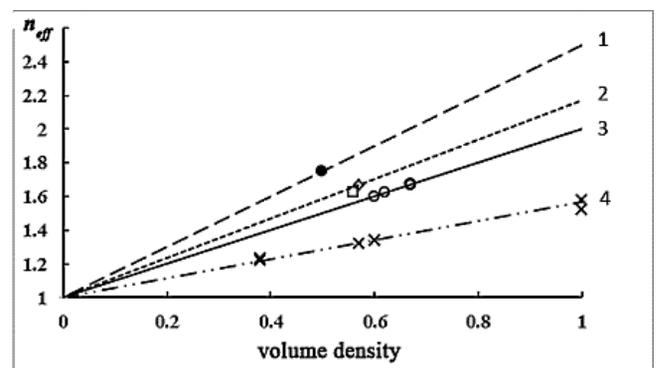


Fig. 5. Graphs of the dependence of the refractive index on the volume density of particles: 1 – marble, 2 – gravel ($d = 2...4$ mm), 3 – gravel ($d < 2$ mm) and sand, 4 – polyethylene ($d = 3...5$ mm). Solid lines – calculated by the refractive formula, dashed lines – by the Landau-Lifshitz-Looyenga formula. Discrete symbols – experimental data.

manifests itself the stronger, the higher the refractive index of the particle material. In the case of polyethylene granules, both formulas lead to practically coinciding graphs at the value of the refractive index of polyethylene $n_m = 1.58$ ($\epsilon_m = 2.49$). This value turned out to be slightly overestimated in comparison with the reference data $\epsilon_m = 2.3 \dots 2.4$.

4. DISCUSSION OF RESULTS

An interesting fact is the coincidence of the experimental data for sand and fine gravel. This means that the sand and fine gravel had similar mineral components. The refractive index of the material of their particles, calculated using the refractive formula, turned out to be equal to $n_m = 2$ ($\epsilon_m = 4$), and calculated using the Landau-Lifshitz-Looyeng formula – $n_m = 2.05$ ($\epsilon_m = 4.2$). Gravel grains can contain quartz, feldspars, and other minerals. From various sources (for example, [20,22]) it follows that the refractive index of quartz is estimated within 1.9...2.1, feldspar – over 2.25. Thus, the results of measuring the refractive index of sand and fine gravel are in satisfactory agreement with the known data on the assumption that quartz is the main mineral in their composition.

The refractive index of gravel with particle sizes $d = 2 \dots 4$ mm turned out to be noticeably higher than for sand and finer gravel ($d = 1 \dots 2$ mm). The reason for this may be the increased concentration in the material of larger particles of such minerals as feldspar and granite, the refractive index of which is higher than that of quartz [20,22].

The refractive index of white marble chips turned out to have the highest value. The refractive index of marble, calculated using the refractive formula, is $n_m = 2.57$ ($\epsilon_m = 6.6$), and calculated by the Landau-Lifshitz-Looyenga formula – $n_m = 2.7$ ($\epsilon_m = 7.29$). It is known [23] that the dielectric properties of marble, even white, which is considered one of the purest calcites, strongly depend on its

chemical composition. The values $\epsilon_m = 6.8 \dots 7.2$ measured at frequencies of 40-50 GHz for marble with unknown composition are given in [24]. These values are in satisfactory agreement with the data obtained in this work and presented above.

Thus, a comparative analysis of the experimental data obtained for a number of granular substances showed that the use of a prismatic cuvette makes it possible to measure their refractive index by the prism method using the mode of receiving thermal radiation.

5. CONCLUSION

In this work, the features of the application of the classical prism method in the range of MM waves for measuring the refractive index of loose materials with particle sizes comparable to the wavelength have been investigated. It is shown that the method is implemented using a hollow radio transparent prismatic cell filled with a test substance. In addition, the proposed method can be implemented in a passive mode. The values of the refractive index of mixtures of sand, gravel, marble chips and polyethylene with air, measured by the prism method at a frequency of 37.5 GHz, are in satisfactory agreement with the known experimental data for quartz, feldspar, polyethylene, and marble.

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