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CALCULATION OF THE EFFECTIVE DIELECTRIC PERMITTIVITY OF A COMPOSITE MATERIAL CONTAINING A FILLER WITH NEGATIVE DIELECTRIC PERMITTIVITY

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Abstract. An explanation is provided for why the Bruggeman equation is unsuitable for calculating the effective permittivity of a composite material containing a filler with negative permittivity. Formulas have been derived for calculating the effective permittivity of a composite containing spherical nanoparticles of filler with negative permittivity. These formulas can be used in producing composite materials with specified permittivity when metal nanoparticles are used as fillers in composites. The existence of a non-monotonic "resonant" dependence of the effective dielectric permittivity on the concentration of filler nanoparticles is predicted in these cases.

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1. INTRODUCTION

One of the important advantages of composite material is that by varying the volume concentration of filler nanoparticles in its matrix, it is possible to manufacture artificial material with a specified value of effective permittivity. In particular, the use of such composite materials in multilayer bandpass filters, which belong to frequency-selective surfaces of microwave and optical ranges, allows to significantly reduce the number of dielectric layers in the structure and thereby considerably enhance their frequency-selective properties [1]. This enhancement is expressed in the expansion of the lower and upper stopbands of the filter at a fixed bandwidth.

For calculating the effective dielectric permittivity of composite materials, the Bruggeman equation is widely used [2]. In the case of isotropic composites containing spherical isotropic particles, this equation takes the form

$$c_f \frac{\varepsilon^f - \varepsilon^e}{\varepsilon^f + 2\varepsilon^e} + (1 - c_f) \frac{\varepsilon^m - \varepsilon^e}{\varepsilon^m + 2\varepsilon^e} = 0, \qquad (1)$$

where c_f is the relative volume concentration of filler particles in the composite material matrix, ε^m and ε^f are the relative dielectric permittivities of the composite matrix material and its filler material respectively, ε^e is the relative effective dielectric permittivity of the composite material. This equation was obtained in the quasi-static approximation, which assumes that the dimensions of each particle of the composite material are small compared to the wavelength both in the particle itself and in the composite material. The dipole interaction between composite particles was taken into account in the effective field approximation, describing the effect on a selected particle from other surrounding particles. This approximation became known as the effective medium approximation [3] or effective medium theory [4]. The solution to equation (1) is expressed by the formula

$$\varepsilon^e = \frac{H + \sqrt{H^2 + 8\varepsilon^m \varepsilon^f}}{4}, \qquad (2)$$

where

$$H = (2 - 3c_f)\varepsilon^m + (3c_f - 1)\varepsilon^f.$$

The generalization of equation (1) to the case when the sizes of spherical particles are comparable to the wavelength in the filler material, but remain much smaller than the wavelength in the composite material, is most easily accomplished by making the substitution in equation (1) [5]

$$\varepsilon^i \to \varepsilon^i J(k^i a), \tag{3}$$

where $k^i = \sqrt{\varepsilon^i \mu^i}\omega/c$ is the wave number for the i-th material, $(i = m, f)$, a is the particle radius, and the function $J(x)$ is given by the formula

$$J(x) = 2\frac{1 - x\mathrm{ctg}(x)}{x^2 + x\mathrm{ctg}(x) - 1}. \tag{4}$$

This function becomes equal to one when $x = 0$.

The generalization of equation (1) for the case of anisotropic composite materials containing co-directed ellipsoidal particles made of isotropic materials is written as a system of equations [6–8]

$$\frac{c_f(\varepsilon^f - \varepsilon_j^e)}{\varepsilon_j^e + N_j(\varepsilon^f - \varepsilon_j^e)} + \frac{(1 - c_f)(\varepsilon^m - \varepsilon_j^e)}{\varepsilon_j^e + N_j(\varepsilon^m - \varepsilon_j^e)} = 0, \tag{5}$$

where ε_j^e are the diagonal elements of the tensor of relative effective permittivity ε^e of the composite material, index j numbers the coordinate axes x, y, z, coinciding with the axes of co-directed ellipsoidal particles, N_j are the depolarization coefficients of ellipsoidal particles of the composite. Equations (5) express the requirement that the sum of electric dipole moments $\mathbf{p}^i$ of all particles, both matrix and filler, contained in any sufficiently large selected volume of composite material, equals zero. Note that the components of the complex vector $\mathbf{p}^i$ of dipole moment of the i-th particle in an anisotropic medium with relative permittivity tensor ε^e are expressed by the formula [9]

$$p_j^i = V^i \frac{\varepsilon_j^e(\varepsilon^i - \varepsilon_j^e)}{\varepsilon_j^e + N_j(\varepsilon^i - \varepsilon_j^e)}\varepsilon_0 E_j^0, \tag{6}$$

where ε_0 is the absolute permittivity of free space, E^0 is the complex amplitude of the electric field in the anisotropic surrounding medium, V^i is the volume, ε^i is the relative permittivity of the material of the i-th particle.

Further, harmonic electromagnetic oscillations will be described by the multiplier $\exp(-i\omega t)$. In this case, the imaginary part of any of the considered permittivities cannot take negative values.

Depolarization coefficients N_j are known as elements of the depolarization tensor $\hat{\mathbf{N}}$, which describe the relationship between the electric field $\mathbf{E}^i$ inside the sample and the field $\mathbf{E}^0$ in the space surrounding the sample. When the sample has an ellipsoidal shape, and its axes $\boldsymbol{\varepsilon}^e$ are directed along the tensor axes, describing the dielectric properties of the surrounding medium, this relationship is expressed by the formula [9]

$$E_j^i = \frac{\varepsilon_j^e}{\varepsilon_j^e + N_j(\varepsilon^i - \varepsilon_j^e)}E_j^0, \tag{7}$$

where ε^i is the relative dielectric permittivity of the isotropic material of the th sample. For a spherical sample in an isotropic medium, the depolarization coefficient N_j does not depend on dielectric permittivities e^i, ε^e and takes a fixed value for any direction j. In this case, formula (7) has a simple form [9]

$$\mathbf{E}^i = \frac{3\varepsilon^e}{2\varepsilon^e + \varepsilon^i}\mathbf{E}^0. \tag{8}$$

The mathematical solution of the system of equations (5) can be written as

$$\varepsilon_j^e = \frac{H_j \pm \sqrt{R_j}}{2(1 - N_j)}, \tag{9}$$

$$R_j = H_j^2 + 4N_j(1 - N_j)\varepsilon^m\varepsilon^f,$$

$$H_j = (1 - c_f - N_j)\varepsilon^m + (c_f - N_j)\varepsilon^f.$$

As noted in [8], traditionally values of coefficients N_j are substituted into equation (5), which are considered as constants independent of ε_j^e and defined by integrals

$$N_j = \int_0^\infty \frac{a_x a_y a_z\, ds}{2(s + a_j^2)\sqrt{(s + a_x^2)(s + a_y^2)(s + a_z^2)}}, \tag{10}$$

where a_x, a_y, a_z are semi-axes of ellipsoidal composite particles. From this formula, it follows that

$$N_x + N_y + N_z = 1. \tag{11}$$

It is also evident that coefficients N_j in formula (10) depend only on the shape of particles, but not on their sizes and dielectric permittivities ε^m and ε^f. In the particular case when the ellipsoid is a spheroid, i.e. $a_x = a_y \neq a_z$, formula (10) is significantly simplified and takes the form [9]

$$N_z = \frac{1 + \kappa^2}{\kappa^3}(\mathrm{arctg} - \kappa),$$

$$N_x = N_y = \frac{1 - N_z}{2}, \tag{12}$$

$$\kappa = \sqrt{a_x^2/a_z^2 - 1}.$$

However, formula (12) was obtained for a spheroidal sample located only in an isotropic medium [9]. This means that its use for samples in anisotropic media is invalid. Therefore, in [8] for the depolarization coefficients N_j, it is proposed to use another formula, which is rigorous but more complex. It coincides with formula (10) if instead of semi-axes a_x and a_z one uses the semi-axes of the reduced spheroid [10]

$$a'_x = a_x/\sqrt{\varepsilon^e_x}, \qquad a'_z = a_z/\sqrt{\varepsilon^e_z}. \tag{13}$$

Thus, the depolarization coefficients N_j in equations (5) are not constants but functions of concentration c_f. They must be found together with functions $\varepsilon^e_j(c_f)$ by jointly solving the system of equations (9), (10), and (13). Note that coefficients N_x and N_z, according to formulas (12) and (13), can take complex values when the ratio $\varepsilon^e_x/\varepsilon^e_z$ ceases to be real. These facts were pointed out in work [8]. Comparison of calculation results ε^e_j, performed using both the traditional and rigorous formula for depolarization coefficients N_j, was conducted in work [11].

Further, we will consider the case of a composite material that is isotropic with respect to a uniform electric field $\mathbf{E}^0$. For definiteness, we will assume that this field is directed along the axis z. Therefore, we will call the effective dielectric permittivity of the composite ε^e the diagonal element ε^e_z of tensor $\boldsymbol{\varepsilon}^e$, which describes the properties of the medium with respect to inhomogeneous local fields of particle scattering $\mathbf{E}^s$. The depolarization coefficient N, corresponding to element ε^e_z, will be called the diagonal element N_z. Then the equation for the effective dielectric permittivity ε^e, according to formula (9), takes the form

$$\varepsilon^e = \frac{H \pm \sqrt{R}}{2(1-N)}, \tag{14}$$

$$H = (1 - c_f - N)\varepsilon^m + (c_f - N)\varepsilon^f,$$

$$R = H^2 + 4N(1-N)\varepsilon^m\varepsilon^f.$$

This ambiguous formula, when choosing the plus sign and $N = 1/3$, coincides with formula (2).

2. UNSOLVED PROBLEM

A significant disadvantage of the Bruggeman equation (1) is that for negative values of ε^f there exists a range of concentration values c_f, in which the effective dielectric permittivity ε^e, calculated using formula (2), takes complex values even with real values of ε^m and ε^f. The lower and upper boundaries of this range for real ε^m and ε^f are expressed by the formula

$$c_{1,2} = \frac{(\sqrt{2\varepsilon^m} \mp \sqrt{-\varepsilon^f})^2}{3(\varepsilon^m - \varepsilon^f)}. \tag{15}$$

However, a complex value of ε^e in the absence of dielectric losses is inadmissible, as it indicates an unstable state of the composite material, manifested in the change of amplitude of a plane electromagnetic wave as it propagates. This drawback of equation (1) was pointed out in work [12].

Another disadvantage is that at $2\varepsilon^m + \varepsilon^f < 0$ formula (2) at point $c_f = 0$ instead of value $\varepsilon^e = \varepsilon^m$ gives value $\varepsilon^e = -\varepsilon^f/2$, and at $\varepsilon^m + 2\varepsilon^f < 0$ at point $c_f = 1$ instead of value $\varepsilon^e = \varepsilon^f$ gives value $\varepsilon^e = -\varepsilon^m/2$. However, this drawback is easily eliminated if in formula (2) we choose the opposite sign before the square root sign, i.e., choose another root of the quadratic equation (1).

In the optical range, metals possess negative dielectric permittivity [13, 14].

The purpose of this work is to calculate the concentration dependence of the effective dielectric permittivity $\varepsilon^e(c_f)$ of an isotropic non-magnetic composite material containing a filler with negative dielectric permittivity ε^f.

3. CAUSE OF THE PROBLEM

The indicated problem arose as a result of an erroneous assumption that an isotropic composite medium always exhibits isotropic properties not only in relation to a uniform polarizing field $\mathbf{E}^0$, but also in relation to localized inhomogeneous scattering fields $\mathbf{E}^s$ generated by polarized particles of the medium, which have components orthogonal to the field $\mathbf{E}^0$.

In reality, a polarized composite medium, being isotropic with respect to a uniform field $\mathbf{E}^0$, cannot maintain its isotropic properties with respect to micro-inhomogeneous fields $\mathbf{E}^s$ if ε^m and ε^f have opposite signs. Otherwise, i.e., with isotropic depolarization coefficients N_j, the polarized composite medium will be in an unstable state with respect to fields $\mathbf{E}^s$, as indicated by the appearance of an imaginary part at real values of ε^m and ε^f, as noted above.

The dependence of the particle scattering field structure $\mathbf{E}^s$ on the anisotropic properties of the surrounding medium, described by tensor $\boldsymbol{\varepsilon}^e$, can be judged by formulas

$$E_x^p = \frac{\dfrac{2p_x x^2}{\varepsilon_x^e} + \dfrac{3p_y xy}{\varepsilon_y^e} + \dfrac{3p_z xz}{\varepsilon_z^e} - \dfrac{p_x y^2}{\varepsilon_y^e} - \dfrac{p_x z^2}{\varepsilon_z^e}}{4\pi\varepsilon_0\,\varepsilon_x^e \sqrt{\varepsilon_x^e\,\varepsilon_y^e\,\varepsilon_z^e \left(\dfrac{x^2}{\varepsilon_x^e} + \dfrac{y^2}{\varepsilon_y^e} + \dfrac{z^2}{\varepsilon_z^e}\right)^{5/2}}},$$

$$E_y^p = \frac{\dfrac{2p_y y^2}{\varepsilon_y^e} + \dfrac{3p_z yz}{\varepsilon_z^e} + \dfrac{3p_x xy}{\varepsilon_x^e} - \dfrac{p_y z^2}{\varepsilon_z^e} - \dfrac{p_y x^2}{\varepsilon_x^e}}{4\pi\varepsilon_0\,\varepsilon_y^e \sqrt{\varepsilon_x^e\,\varepsilon_y^e\,\varepsilon_z^e \left(\dfrac{x^2}{\varepsilon_x^e} + \dfrac{y^2}{\varepsilon_y^e} + \dfrac{z^2}{\varepsilon_z^e}\right)^{5/2}}}, \quad (16)$$

$$E_z^p = \frac{\dfrac{2p_z z^2}{\varepsilon_z^e} + \dfrac{3p_x xz}{\varepsilon_x^e} + \dfrac{3p_y yz}{\varepsilon_y^e} - \dfrac{p_z x^2}{\varepsilon_x^e} - \dfrac{p_z y^2}{\varepsilon_y^e}}{4\pi\varepsilon_0\,\varepsilon_z^e \sqrt{\varepsilon_x^e\,\varepsilon_y^e\,\varepsilon_z^e \left(\dfrac{x^2}{\varepsilon_x^e} + \dfrac{y^2}{\varepsilon_y^e} + \dfrac{z^2}{\varepsilon_z^e}\right)^{5/2}}}.$$

These expressions follow from formula

$$\varphi^p = \frac{\varepsilon_y^e\,\varepsilon_z^e\,p_x\,x + \varepsilon_x^e\,\varepsilon_z^e\,p_y\,y + \varepsilon_x^e\,\varepsilon_y^e\,p_z\,z}{4\pi\varepsilon_0[\varepsilon_x^e\,\varepsilon_y^e\,\varepsilon_z^e(x^2/\varepsilon_x^e + y^2/\varepsilon_y^e + z^2/\varepsilon_z^e)]^{3/2}}, \quad (17)$$

which describes in the quasi-static approximation the potential of a point dipole moment $\mathbf{p}$ in an anisotropic medium. In turn, formula (17) itself is derived from the known formula [9]

$$\varphi^q = \frac{q}{4\pi\varepsilon_0\sqrt{\varepsilon_x^e\,\varepsilon_y^e\,\varepsilon_z^e(x^2/\varepsilon_x^e + y^2/\varepsilon_y^e + z^2/\varepsilon_z^e)}} \quad (18)$$

for the Coulomb potential of a point charge q in an anisotropic surrounding medium and the known relation [15]

$$\varphi^p = -\frac{\mathbf{p}}{q}\,\mathrm{grad}\varphi^q, \quad (19)$$

expressing the potential φ^p of a point dipole moment $\mathbf{p}$ through the Coulomb potential φ^q of a point charge.

Thus, the error in calculating the effective dielectric permittivity of an isotropic composite material occurred because equation (14) did not account for the dependence of the depolarization coefficient N on the field structure $\mathbf{E}^s$, which, in turn, depends on the concentration c^f. That is, the depolarization coefficient was assigned a fixed value $N = 1/3$, which turned the correct formula (14) into the erroneous formula (2).

Therefore, the purpose of this work is reduced to calculating the concentration dependence of the depolarization coefficient $N(c_f)$, which is used in formula (14) when calculating the effective dielectric permittivity $\varepsilon^e(c_f)$ of an isotropic composite material containing a filler with negative dielectric permittivity ε^f.

4. PROBLEM SOLUTION

Let's first consider the case when dielectric losses in an isotropic composite material are absent and therefore the dielectric permittivities ε^m, ε^f and ε^e take real values. This case is interesting because there are several points of concentration values c_f, where the values of N and ε^e are known in advance.

The most important of these points is the concentration point c_f, where ε^e takes a zero value. The existence of a "zero" point in the region $0 < c_f < 1$ follows from the continuity condition of the real function $\varepsilon^e(c_f)$ and its boundary conditions

$$\varepsilon^e(c_f)\,|_{c_f=0} = \varepsilon^m > 0,$$

$$\varepsilon^e(c_f)\,|_{c_f=1} = \varepsilon^f < 0.$$

The simultaneous zeroing of the function $N(c_f)$ at the "zero" point follows from formulas (12), (13). From equation (5), we find the value c_f for the "zero" point:

$$c_f = \varepsilon^m/(\varepsilon^m - \varepsilon^f). \quad (20)$$

We note that at this point, the expression under the radical $\mathcal{R}$, used in formula (14), becomes zero.

Two other important values of c_f are the boundary points $c_f = 0$ and $c_f = 1$, where ε^e takes values ε^m and ε^f respectively. To find the value of N at such points, we can use the formula

$$N = \frac{c_f\varepsilon^e}{\varepsilon^e - \varepsilon^m} + \frac{(1-c_f)\varepsilon^e}{\varepsilon^e - \varepsilon^f}, \quad (21)$$

which is derived from equation (5). From this, we find the values N:

$$\lim_{\substack{c_f\to 0\\ \varepsilon^e\to\varepsilon^m}} N = \frac{\varepsilon^m}{\varepsilon^m - \varepsilon^f}, \quad \lim_{\substack{c_f\to 1\\ \varepsilon^e\to\varepsilon^f}} N = \frac{\varepsilon^f}{\varepsilon^f - \varepsilon^m}. \quad (22)$$

We note that at these two extreme points, as at the "zero" point, the expression under the radical $\mathcal{R}$ becomes zero.

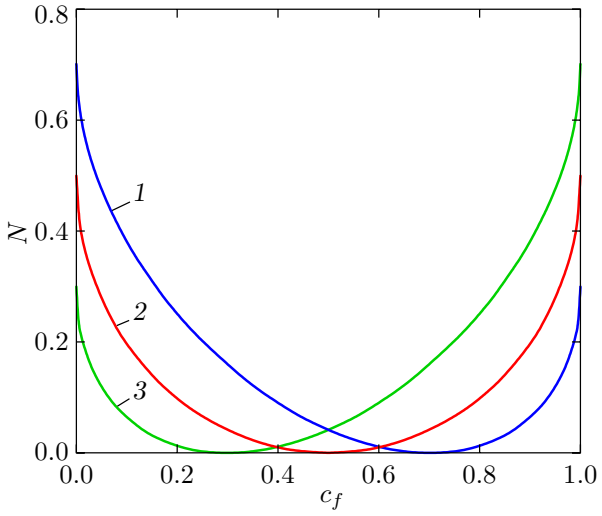

Fig. 1. Concentration dependencies of particle depolarization coefficient. 1 – $\varepsilon^m = 7$, $\varepsilon^f = -3$; 2 – $\varepsilon^m = 5$, $\varepsilon^f = -5$; 3 – $\varepsilon^m = 3$, $\varepsilon^f = -7$

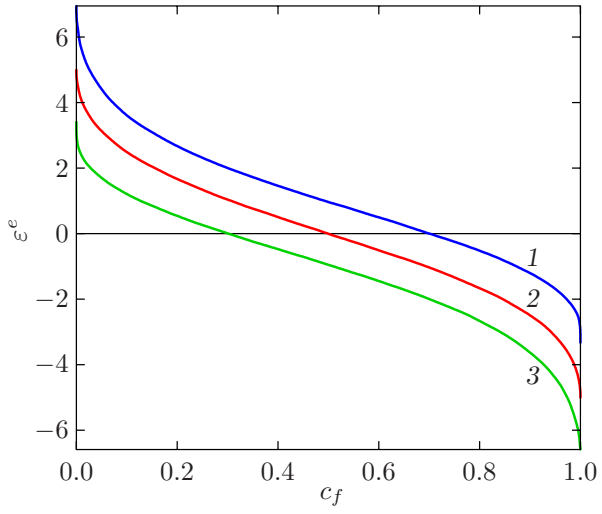

Fig. 2. Concentration dependencies of effective dielectric permittivity. 1 – $\varepsilon^m = 7$, $\varepsilon^f = -3$; 2 – $\varepsilon^m = 5$, $\varepsilon^f = -5$; 3 – $\varepsilon^m = 3$, $\varepsilon^f = -7$

Let's consider two more points c_1 and c_2, given by formula (15). At these points, as we already know, the real solutions of the partial equation (1), expressed by formula (2), coincide with solutions (14) of the generalized equation (5) at $N = 1/3$. It means that the expression under the radical $\mathcal{R}$ becomes zero at these two points of values c_f.

Thus, the known partial solutions of equation (14) for the five points of value c_f mentioned above can serve as a criterion for verifying the correctness of the sought solution $\varepsilon^e(c_f)$ across the entire range of values f_c.

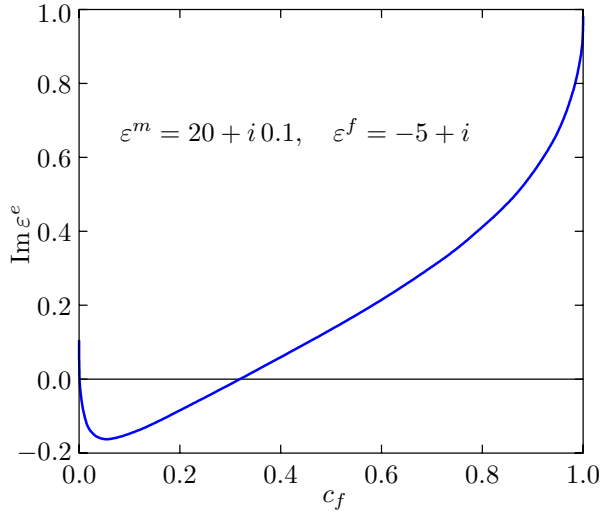

Fig. 3. Dependence $\mathrm{Im}\,\varepsilon^e(c_f)$, calculated using formulas (26) and (23) and subject to recalculation using formulas (27) and (23)

By definition, the effective dielectric permittivity $\varepsilon^e(c_f)$ of an isotropic composite medium is a unique value that can take only one single value. Its imaginary part in the presence of dielectric losses must be only positive at any concentration c_f, and in the absence of losses must equal zero. From the uniqueness $\varepsilon^e(c_f)$ it follows that in formula (14) the expression under the root $\mathcal{R}$ must equal zero. Hence, to find the desired dependence $N(c_f)$ we obtain the missing equation:

$$\varepsilon^e = \frac{(1 - c_f - N)\varepsilon^m + (c_f - N)\varepsilon^f}{2(1 - N)}. \qquad (23)$$

From the equality itself $\mathcal{R} = 0$ follows the equation

$$[(1 - c_f - N)\varepsilon^m + (c_f - N)\varepsilon^f]^2 + \\ + 4N(1 - N)\varepsilon^m\varepsilon^f = 0. \qquad (24)$$

Its solution is expressed by a two-valued formula:

$$N = \frac{\left[\sqrt{(1 - c_f)\varepsilon^m} \pm \sqrt{-c_f\varepsilon^f}\right]^2}{\varepsilon^m - \varepsilon^f}. \qquad (25)$$

In the absence of dielectric losses, as we have already noted, there exists a "zero" point c_f, where $N(c_f)$ becomes zero. Therefore, formula (24) in the absence of dielectric losses takes the form

$$N = \frac{[\sqrt{(1 - c_f)\varepsilon^m} - \sqrt{-c_f\varepsilon^f}]^2}{\varepsilon^m - \varepsilon^f}. \qquad (26)$$

Dependencies $N(c_f)$ and $\varepsilon^e(c)$, defined by formulas (26) and (23), are illustrated by the graphs shown in Fig. 1 and Fig. 2 for three sets of real values ε^m and ε^f.

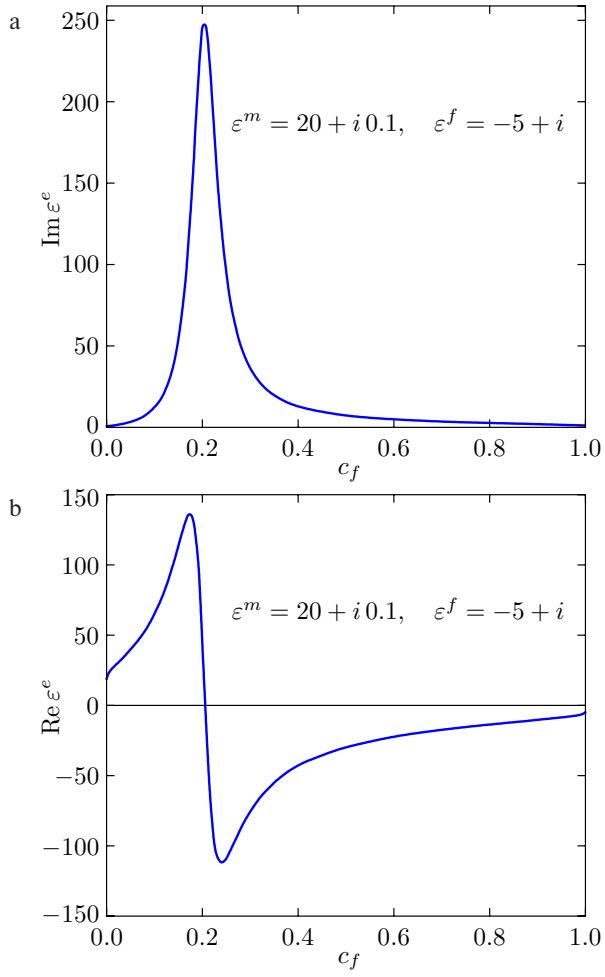

Fig. 4. Dependencies Im $\varepsilon^e(c_f)$ and $\mathcal{R}$ $\varepsilon^e(c_f)$, calculated using formulas (27) and (23)

It can be seen that the dependencies $N(c_f)$ are described by continuous smooth functions with one minimum at the zero level. Dependencies $\varepsilon^e(c_f)$ are described by continuous smooth decreasing functions intersecting the zero level.

In the presence of dielectric losses, from the two values $N(c_f)$ in the two-valued formula (24), one should choose the value at which the imaginary part $\varepsilon^e(c_f)$ does not take negative values at any concentration value c_f. An example where the imaginary part of the function $\varepsilon^e(c_f)$, calculated using formula (26), takes a negative value, is shown in the graph in Fig. 3.

In this case, instead of formula (26), one should use formula

$$N = \frac{[\sqrt{(1-c_f)\varepsilon^m} + \sqrt{-c_f\varepsilon^f}\,]^2}{\varepsilon^m - \varepsilon^f}. \qquad (27)$$

The result of alternative calculation of dependence $\varepsilon^e(c_f)$ using formulas (23) and (27), which prevents

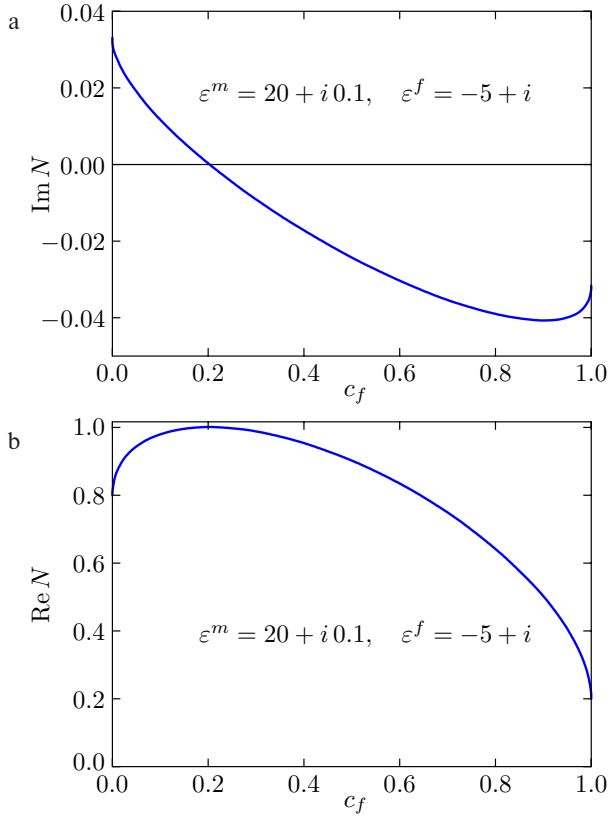

Fig. 5. Dependencies Im $N(c_f)$ and $\mathcal{R}$ $N(c_f)$, calculated using formula (27)

the occurrence of negative values Im ε^e at the same parameter values ε^m and ε^f, is shown in Fig.4. The corresponding dependence $N(c_f)$, calculated using formula (27), is presented in Fig. 5.

The concentration dependency curves in Fig. 4 resemble frequency resonance curves. Let's try to provide not a rigorous but at least a simplified explanation for this unexpected phenomenon. Each nanoparticle of the composite will be simplistically considered as a reactive two-terminal network. For this, we first write the formula for complex conductivity of a parallel oscillatory circuit

$$Y_{par} = 1/\mathcal{R} + i/(\omega L) - i\omega C_0\,\varepsilon \qquad (28)$$

and the formula for complex impedance of a series oscillatory circuit

$$Z_{ser} = \mathcal{R} - i\omega L - i/(\omega C_0\,\varepsilon). \qquad (29)$$

Here ε is the relative dielectric permittivity of the material filling the capacitor, and C_0 is the capacitance when $\varepsilon = 1$.

Both from formula (28) and formula (29), it is evident that when $\mathcal{R}\,\varepsilon < 0$ the sign of the reactive

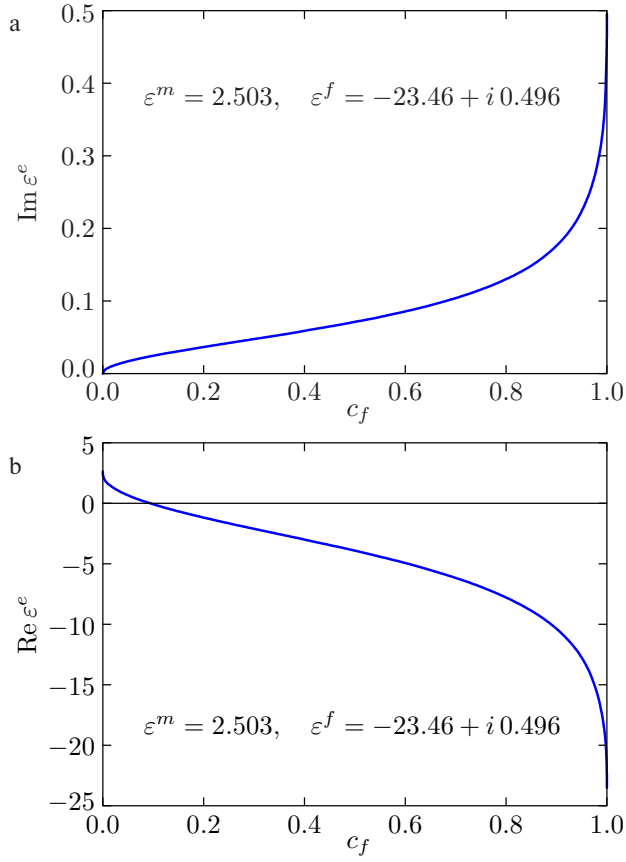

Fig. 6. Dependencies $\varepsilon^e(c_f)$ at frequency 426 THz, calculated using formulas (23) and (26)

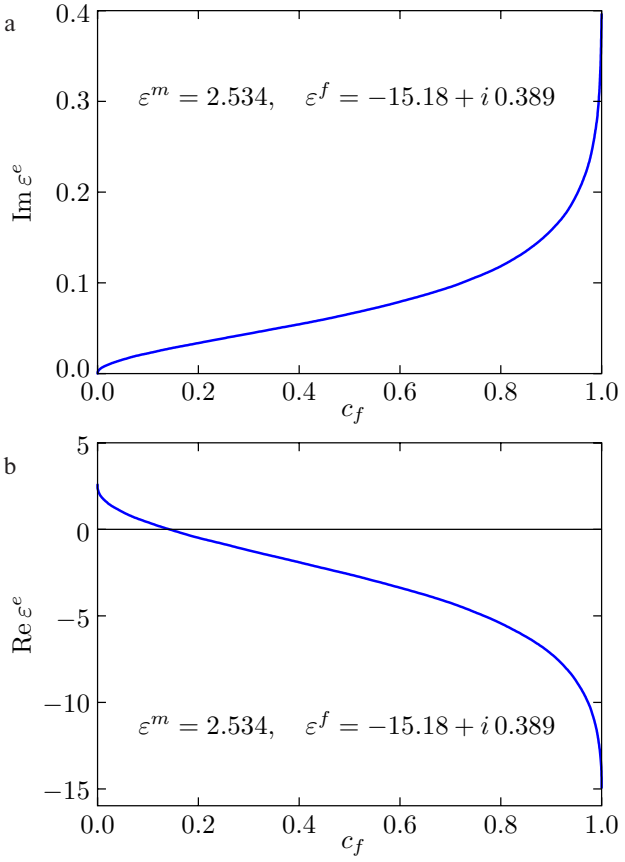

Fig. 7. Dependencies $\varepsilon^e(c_f)$ at frequency 510 THz, calculated using formulas (23) and (26)

part of the capacitor impedance coincides with the sign of the reactive part of the inductance impedance. This means that the nanoparticles of the composite filler with negative dielectric permittivity ε^f will exhibit only inductive properties.

In the case of $\mathcal{R}\,\varepsilon > 0$ the sign of the reactive part of the capacitor impedance is opposite to the sign of the reactive part of the inductance impedance. Therefore, the matrix nanoparticles of the composite, having positive dielectric permittivity ε^m, can exhibit both capacitive and inductive properties, depending on whether the capacitive or inductive term of the reactive part is larger.

In cases where matrix nanoparticles exhibit capacitive properties, they, together with the filler nanoparticles with inductive properties will represent a system whose dielectric properties depend on the filler concentration in a "resonant" manner c_f. At the "resonant" concentration, the real part of the effective dielectric permittivity ε^e becomes zero, while the imaginary part reaches its maximum value. Below the "resonant" concentration, the composite material has an effective dielectric permittivity ε^e where the real part is positive. Above the "resonant" concentration, the real part is negative.

Note that the obtained solution, expressed by formulas (26), (27), is in complete agreement with the previously known solutions of the problem for the five indicated points c_f. This solution also satisfies the boundary conditions

$$\varepsilon^e(c_f)|_{c_f=0} = \varepsilon^m,$$

$$\varepsilon^e(c_f)|_{c_f=1} = \varepsilon^f.$$

5. CALCULATION EXAMPLES FOR POLYSTYRENE FILLED WITH SILVER NANOPARTICLES

As an example, we present the calculation results of the effective dielectric permittivity of a composite, where polystyrene is the matrix and silver nanoparticles are the filler. The calculation will be performed using formulas (26), (27), and (23) at

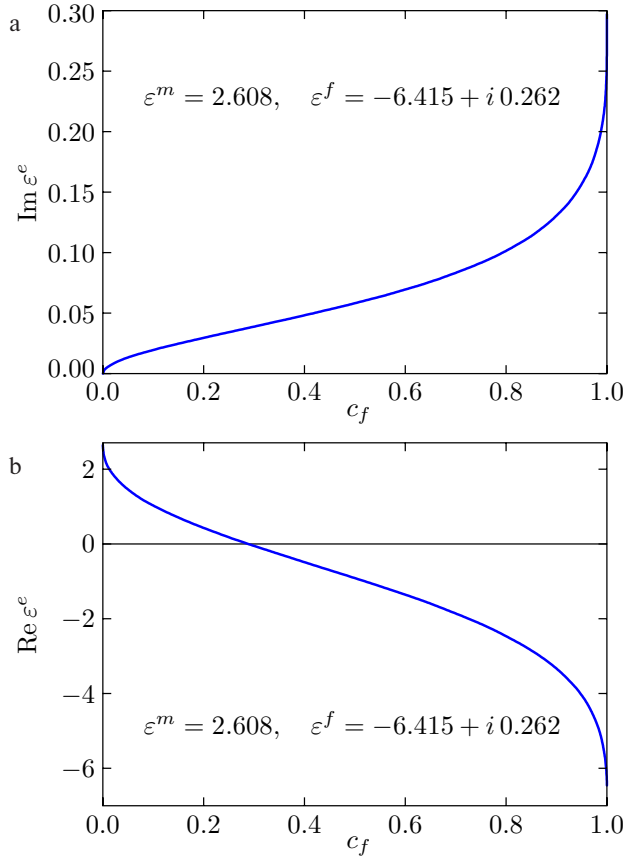

Fig. 8. Dependencies $\varepsilon^e(c_f)$ at a frequency of 688 THz, calculated using formulas (23) and (26)

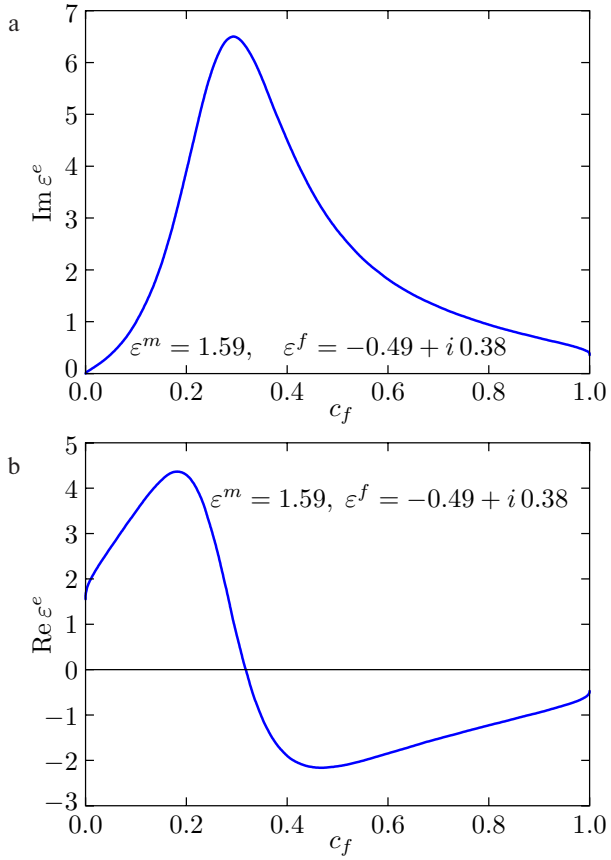

Fig. 9. Dependencies $\varepsilon^e(c_f)$ at a frequency of 914 THz, calculated using formulas (23) and (27)

three frequencies of the visible optical range and one frequency of the invisible ultraviolet range.

The first calculation was performed for a frequency of 426 THz, corresponding to the red color range. At this frequency, polystyrene has a dielectric permittivity $\varepsilon^m = 2.503$ [16], and silver $\varepsilon^f = -23.4 + i0.495$ [14]. The result of calculating the complex effective dielectric permittivity of the composite is shown in Fig. 6.

The second calculation was performed for a frequency of 510 THz, corresponding to the yellow color range. At this frequency, polystyrene has a dielectric permittivity $\varepsilon^m = 2.534$ [16], and silver $\varepsilon^f = -15.18 + i0.389$ [14]. The result of calculating the effective dielectric permittivity of the composite is shown in Fig. 7.

The third calculation was performed for a frequency of 688 THz, corresponding to the violet color range. At this frequency, polystyrene has a dielectric permittivity $\varepsilon^m = 2.608$ [16], and silver $\varepsilon^f = -6.415 + i0.262$ [14]. The result of calculating the effective dielectric permittivity of the composite is shown in Fig. 8.

The fourth calculation was performed for a frequency of 914 THz, corresponding to the invisible ultraviolet range. At this frequency, polystyrene has a dielectric permittivity $\varepsilon^m = 1.59$ [17], and silver – $\varepsilon^f = -0.94 + i0.38$ [14]. The result of calculating the effective dielectric permittivity of the composite is shown in Fig. 9.

The presented results of calculating the effective dielectric permittivity of a composite material, where polystyrene serves as the matrix and silver nanoparticles as the filler, show that across the entire visible optical frequency range, the concentration dependencies $\mathrm{Re}\,\varepsilon^e(c_f)$ and $\mathrm{Im}\,\varepsilon^e(c_f)$ are continuous monotonic functions. Only in the ultraviolet frequency range, the concentration dependence $\varepsilon^e(c_f)$ ceases to be monotonic and exhibits "resonant" properties.

6. CONCLUSIONS

The paper shows that the reason why the Bruggeman equation (1) does not allow calculating the effective dielectric permittivity $\varepsilon^e(c_f)$ of an isotropic composite material containing spherical filler particles with

negative dielectric permittivity is the use of an erroneous assumption in its derivation that the depolarization coefficient N for any of its particles in a polarized composite is always equal to 1/3.

In reality, the depolarization coefficient N at negative values of the real part of the filler's dielectric permittivity ε^f is a function of the volume concentration of the filler c_f in the composite matrix. To calculate the dependence $N(c_f)$, a two-valued formula (24) was obtained. For given parameter values ε^m and ε^f, one should exclude from the two values N the value that does not ensure a stable state of the composite material. The criterion for material state instability in the presence of dielectric losses is a negative value of the imaginary part of $\varepsilon^e(c_f)$ in at least some range of values c_f. In the absence of dielectric losses, in the two-valued formula (25), one should choose the value N expressed by the single-valued formula (26). To calculate $\varepsilon^e(c_f)$ corresponding to the chosen value $N(c_f)$, formula (23) was obtained.

The numerical calculations performed $\varepsilon^e(c_f)$ using the obtained formulas showed that the dependence $\varepsilon^e(c_f)$ is not always monotonic. When the real part ε^m is sufficiently large compared to $|\varepsilon^f|$, the dependence $\varepsilon^e(c_f)$ resembles a frequency resonance curve.

The results of this work may be useful in developing composite materials with specified effective dielectric permittivity values for their use in optical range devices.

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