Effective Dielectric Permittivity of Matrix Disperse Systems in Differential Medium Approximation

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Theoretical approach is proposed to description of dielectric properties of matrix disperse systems which consist of dielectric matrix with embedded metallic inclusions. On the basis of effective differential medium approximation the analytical expressions are obtained for the effective dielectric permittivity $\tilde{\epsilon}$ of the matrix disperse system with inclusions of spherical and ellipsoidal shape. The analysis of limits of possible values of real and imaginary parts of $\tilde{\epsilon}$ is carried out depending on system parameters.

1. Introduction

Under theoretical study of processes of electromagnetic radiation interaction with matrix disperse systems (MDS) which represent a continuous matrix (commonly, dielectric one) with the imbedded inclusions of the various form and nature, the effective medium approximation is widely used. The core of this method consists in that the MDS with distributed values of the dielectric permeability of a matrix ε_1 and inclusions ε_2 , is substituted by continuous medium with effective dielectric permittivity $\tilde{\epsilon}$, which depends both on ϵ_1 and ε_2 , and on the value $f = V/V_0$ (where V is a volume engaged by inclusions, V_0 is a total volume of a system) as well as on its statistical distribution in a matrix. Such approximation is in a close relation to the experiment in a case when wavelength of the incident electromagnetic radiation is large compared to the inclusion sizes and the mean distance between them (long wave approximation). There are many calculation methods for such systems. The literature review concerning this problem can be found in [1-2]. In the present paper the calculation of effective dielectric permittivity $\tilde{\epsilon}$ is carried out under the large concentration of metallic inclusions of spherical and ellipsoidal shape in the approximation of differential effective medium (DEM) [3-5]. The estimation of possible limits of values $\tilde{\epsilon}'$ and $\tilde{\epsilon}''$ ($\tilde{\epsilon} = \tilde{\epsilon}' + i \tilde{\epsilon}''$) is obtained with use of common methods of electrodynamics of inhomogeneous medium.

2. The MDS effective dielectric permittivity in DEM approximation

A correct calculation of effective dielectric permittivity ($\tilde{\epsilon}$) of MDS under the large volume fraction of inclusions represents a very complicated problem. One of the methods of $\tilde{\epsilon}$ calculation is the differential effective medium approximation (DEM) [3-4].

Generally, DEM follows from the average field approximation (Bruggemann approximation [6]). In case of inclusions of ellipsoidal shape the Bruggemann approximation for calculation of $\tilde{\epsilon}$ has a form:

$$F(f,\varepsilon_{1},\varepsilon_{2},\widetilde{\varepsilon}) = \frac{(1-f)(\widetilde{\varepsilon}-\varepsilon_{1})}{(1-L)\widetilde{\varepsilon}+L\varepsilon_{1}} + \frac{f(\widetilde{\varepsilon}-\varepsilon_{2})}{(1-L)\widetilde{\varepsilon}+L\varepsilon_{2}} + \frac{4(1-f)(\widetilde{\varepsilon}-\varepsilon_{1})}{(1+L)\widetilde{\varepsilon}+(1-L)\varepsilon_{1}} + \frac{4f(\widetilde{\varepsilon}-\varepsilon_{2})}{(1+L)\widetilde{\varepsilon}+(1-L)\varepsilon_{2}} = 0,$$
(1)

where L is a depolarization factor along the large semi-axis $(L_1 = L; L_2 = L_3 = (1 - L)/2)$, ε_2 and ε_1 are dielectric permittivity of inclusions and a matrix, respectively. Consider first the variation of the effective dielectric permittivity at the expense of adding a small portion of particles of the second phase with relative volume Δf . This change will be:

$$\Delta \widetilde{\varepsilon} = -\frac{\left(\partial F/\partial f\right)_{f=0}}{\left(\partial F/\partial \widetilde{\varepsilon}\right)_{f=0}} \Delta f = g(\varepsilon_1, \widetilde{\varepsilon}) \Delta f .$$
⁽²⁾

After differentiation of Eq. (1) we find:

$$g(\tilde{\varepsilon},\varepsilon_{1}) = \frac{\tilde{\varepsilon}}{3} (\varepsilon_{1} - \tilde{\varepsilon}) \left[\frac{1}{(1-L)\tilde{\varepsilon} + L\varepsilon_{1}} + \frac{4}{(1+L)\tilde{\varepsilon} + (1-L)\varepsilon_{1}} \right].$$
(3)

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(5)

In the result of addition to the effective medium of Δf portion of particles of the second phase, $f\Delta f$ particles of the second phase will be replaced. Therefore, the actual variation of a fraction of particles of the second phase in a new composite will be: $\Delta f_{eff} = (1 - f)\Delta f$, i. e. in the relation (2) it is necessary to replace $\Delta f \rightarrow \Delta f/(1 - f)$. In view of it, the relation (2) will have a form:

$$\frac{\mathrm{d}\widetilde{\varepsilon}}{g(\widetilde{\varepsilon},\varepsilon)} = \frac{\mathrm{d}f}{1-f} \,. \tag{4}$$

The solution of the Eqs. (3)-(4) with the initial condition $\tilde{\varepsilon} = \varepsilon_1$ at f = 0 gives:

$$1 - f = \left(\frac{\varepsilon_1}{\widetilde{\varepsilon}}\right)^a \left(\frac{\varepsilon_1 + \gamma \varepsilon_2}{\widetilde{\varepsilon} + \gamma \varepsilon_2}\right)^b \left(\frac{\widetilde{\varepsilon} - \varepsilon_2}{\varepsilon_1 - \varepsilon_2}\right),$$

where

$$a = \frac{3L(1-L)}{1+3L}; \ b = \frac{2(1-3L)^2}{(1+3L)(5-3L)}; \ \gamma = \frac{1+3L}{5-3L}$$

Thus, for the case of particles of spherical form at L = 1/3 and a = 1/3, $\gamma = 1/2$, b = 0, this formula takes a form [4]:

$$1 - f = \left(\frac{\varepsilon_1}{\widetilde{\varepsilon}}\right)^{1/3} \left(\frac{\widetilde{\varepsilon} - \varepsilon_2}{\varepsilon_1 - \varepsilon_2}\right).$$
(6)

Let us analyze the case of symmetric replacement $f \Leftrightarrow \psi = (1 - f)$ in the formula (1). Really, such replacement can take place when considering the dielectric in MDS as inclusions, and metal particles as a matrix. The physical sense of the formula does not alter from such replacement. Then, from Eq. (1) we obtain:

$$f = \left(\frac{\varepsilon_2}{\widetilde{\varepsilon}}\right)^a \left(\frac{\varepsilon_2 + \gamma \varepsilon_1}{\widetilde{\varepsilon} + \gamma \varepsilon_1}\right)^b \left(\frac{\widetilde{\varepsilon} - \varepsilon_1}{\varepsilon_2 - \varepsilon_1}\right).$$
(7)

For the case of particles of spherical shape with L = 1/3 this formula acquires the form [3]:

$$f = \left(\frac{\varepsilon_2}{\widetilde{\varepsilon}}\right)^{1/3} \left(\frac{\widetilde{\varepsilon} - \varepsilon_1}{\varepsilon_2 - \varepsilon_1}\right).$$
(8)

From formulas (6) and (8) it follows the relation for calculation of $\tilde{\epsilon}$:

$$\frac{1-f}{\varepsilon_1^{1/3}} + \frac{f}{\varepsilon_2^{1/3}} = \frac{1}{\widetilde{\varepsilon}^{1/3}} \,. \tag{9}$$

The equation (9) belongs to a common class of relations of a kind:

$$\widetilde{\varepsilon}^{k} = f \varepsilon_{2}^{k} + (1 - f) \varepsilon_{1}^{k} , \qquad (10)$$

which, as shown in [7], at $|k| \leq 1$ completely satisfies to the theorems on the limits of possible values of the effective dielectric permittivity for MDS and statistical mixtures [7-12]. The further consideration of behavior $\tilde{\epsilon}$ depending on parameters of system we will carry out using just these methods and restrict with the MDS, consisting of the dielectric matrix with the metal inclusions of spherical shape.

3. Estimation of limit value $\tilde{\epsilon}$ for disperse systems

The following inequalities (Wiener relations [9]) for $\tilde{\epsilon}$ are valid for real ϵ_2 and ϵ_1 for two-component system:

$$\varepsilon_{-} < \widetilde{\varepsilon} < \varepsilon_{+},$$
 (11)

$$\frac{1}{\varepsilon} = \frac{f_1}{\varepsilon_1} + \frac{f_2}{\varepsilon_2}, \qquad (12)$$

$$\varepsilon_{+} = f_1 \varepsilon_1 + f_2 \varepsilon_2 \,, \tag{13}$$

where f_1 and f_2 are volume fractions $(f_1 + f_2 = 1)$; ε_1 and ε_2 are dielectric permittivities of medium components; ε_+ is a dielectric permittivity of layered dielectric in electrical field parallel to the border of layers; ε_- is the same for the perpendicilar field orientation. Later in the paper [10] the estimations of a type of Eq. (11) were improved and generalized to the case of complex ε_1 and ε_2 [11-12]. In the case of real ε_1 and complex ε_2 the limits of area of possible values of $\tilde{\varepsilon}'$ and $\tilde{\varepsilon}''$ ($\tilde{\varepsilon} = \tilde{\varepsilon}' + i \tilde{\varepsilon}''$) are given by set of relations (12) and (13) [8], which after introduction of variables $x = \tilde{\varepsilon}/\varepsilon_1$ and $t = (\varepsilon_2 - \varepsilon_1)/\varepsilon_1$ will have a form:

$$x'' = \frac{t''}{t'} (x'-1), \tag{14}$$

$$(x'-a)^{2} + (x''-b)^{2} = R^{2},$$

$$a = \frac{1}{2}; \quad b = \frac{1}{2t''} (t'+|t|^{2}); \quad R = \sqrt{a^{2} + b^{2}},$$
(15)

where x' and x'' (x = x' + ix'') are real and imaginary parts of x; t' and t'' (t = t'+it'') are the same values for t. When obtaining Eqs. (14) and (15) from (12) and (13) the value f was excluded. The straight line of Eq. (14) in a plane of variables (x',x'') passes through a point A (x'=1; x''=0)corresponds to ε_1) and B (x'=1+t'; x''=t' corresponds to ε_2). Through the same points the circle of Eq. (15) passes, which passes, besides, through the coordinate origin. As a result of crossing Eq. (14) and Eq. (15) the segment of the permitted values x' and x'' (Fig. 1) is formed. It should be noted that x' and x'', as it follows from Eqs. (12) and (13) parametrically depend on $f(f = f_2; f_1 = 1 - f).$



Fig. 1. The borders of real and image parts of effective dielectric permittivity in the case of inclusions of spherical shape at f=0.5 and $\varepsilon_1=2$; $\varepsilon_2=5+10i$

- 1) Maxwell-Garnett approximation Eq. (21);
- 2) the approximation Eq. (9);
- 3) Landau approximation Eq. (23);
- 4) DEM approximation Eq. (6)

In the paper [7] the improved estimations of limits of permitted values of $\tilde{\epsilon}$ are obtained. They look like Eqs. (10) and (11) from [7]:

$$(\varepsilon_{1} - \widetilde{\varepsilon})(\varepsilon_{1}^{*} - \widetilde{\varepsilon}^{*}) + f \frac{(\varepsilon_{1} - \varepsilon_{2})(\varepsilon_{1}^{*} - \varepsilon_{2}^{*})(\varepsilon_{1}^{*} \widetilde{\varepsilon} - \varepsilon_{1} \widetilde{\varepsilon}^{*})}{\varepsilon_{1} \varepsilon_{2}^{*} - \varepsilon_{2} \varepsilon_{1}^{*}} \leq 0,$$

$$(16)$$

$$\frac{(\varepsilon_2 - \widetilde{\varepsilon})(\varepsilon_2^* - \widetilde{\varepsilon}^*) + (1 - f) \times}{\times \frac{(\varepsilon_2 - \varepsilon_1)(\varepsilon_2^* - \varepsilon_1^*)(\varepsilon_2^* \widetilde{\varepsilon} - \varepsilon_2 \widetilde{\varepsilon}^*)}{\varepsilon_2 \varepsilon_1^* - \varepsilon_1 \varepsilon_2^*} \le 0.$$
(17)

These relations can be easily written down in variable t:

$$(x'-a_1)^2 + (x''-b_1)^2 = R_1^2,$$

$$a_1 = 1; \quad b_1 = \frac{f|t|^2}{2t''}; \quad R_1 = b_1;$$

$$(x'-a_2)^2 + (x''-b_2)^2 = R_2^2.$$
(18)

$$a_{2} = \left(1 + t' + \frac{1 - f}{2}|t|^{2}\right);$$

$$b_{2} = \frac{(1 - f)(1 + t')|t|^{2}}{2t''};$$
(19)

$$R_2 = \frac{(1-f)|1+t'| |t|^2}{2t''}.$$

As a result of intersection of circles (18) and (19) the small segment is formed, which tops lay on a straight line (14) and a circle (15), whereas its position in a sector depends on f. One can show [7] that relations for calculation of $\tilde{\epsilon}$ in a form (10) under $|k| \leq 1$ satisfy to the restrictions given above. It should be noted that a number of known relations belongs to the condition (10). When $k = \pm 1$ we have Eqs. (12) and (13), when k = 1/3 there is the relation of L. D. Landau [13], and when k = -1/3 we have the relation (9), etc. The case k = 0 is a special one and $\tilde{\epsilon}$ can be found from the relation of Lichtenecker [14]:

$$\ln \tilde{\varepsilon} = f \ln \varepsilon_2 + (1 - f) \ln \varepsilon_1.$$
⁽²⁰⁾

The relation (20) also satisfies to above mentioned restrictions. On Fig. 1 the area of the permitted values of $\tilde{\epsilon}'$ and $\tilde{\epsilon}''$ is shown for the case f = 0.5 at $\epsilon_1 = 2$; $\epsilon_2 = 5 + 10i$. The points in the figure correspond to values of $\tilde{\epsilon}$ calculated by the formulas:

the point (1) is Maxwell-Garnett approximation [2]

$$\frac{\widetilde{\varepsilon} - \varepsilon_1}{\widetilde{\varepsilon} + 2\varepsilon_1} = f \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1};$$
(21)

point (2) is the average field approximation [6]

$$(1-f)\frac{\widetilde{\varepsilon}-\varepsilon_1}{\varepsilon_1+2\widetilde{\varepsilon}}+f\frac{\widetilde{\varepsilon}-\varepsilon_2}{\varepsilon_2+2\widetilde{\varepsilon}}=0; \qquad (22)$$

the point (3) is the formula (8), the point (4) is the formula (9), the point (5) is the formula of L. D. Landau [13]:

$$\tilde{\varepsilon}^{1/3} = f \varepsilon_2^{1/3} + (1 - f) \varepsilon_1^{1/3}.$$
(23)



Fig. 2. Dependence of dielectric permittivity $\tilde{\epsilon}$ of MDS on the volume fraction of inclusions *f*, calculated by the DEM approximation with $\epsilon_1=2$; $\epsilon_2=40$

- 1) the upper border of value $\tilde{\epsilon}$, Eq. (13);
- 2) Bruggeman approximation Eq. (22);
- 3) Landau approximation Eq. (23);
- 4) DEM approximation Eq. (6);
- 5) the approximation Eq. (9);
- 6) Maxwell-Garnett approximation Eq. (21);

7) the lower border of value $\tilde{\epsilon}$, Eq. (12)

All the calculated values of $\tilde{\epsilon}$ lay in segment area, i. e. in the region of permitted values. On Fig. 2 the dependence $\tilde{\epsilon}$ on f is shown in the case of real values $\epsilon_1 = 2$; $\epsilon_2 = 40$ calculated by relations (21)-(23) and (8)-(9) for the value L = 1/3. From the figure it is visible, that all curves lay in area limited by straight line Eq. (12) and hyperbole Eq. (13):

$$\chi_{+} = 1 + ft , \qquad (24)$$

$$\chi_{-} = \frac{1+t}{(1-f)(1+t)+f} \,. \tag{25}$$

4. Discussion and conclusions

The obtained relations (5)-(9) for the calculation of $\tilde{\epsilon}$ in DEM approximation for the case of spherical and ellipsoidal inclusions do not contradict the conditions of restrictions Eqs. (15) and (18)-(19) for values $\tilde{\epsilon}'$ and $\tilde{\epsilon}''$. However, here are several moments which we would like to discuss. When restricting with the case of spherical inclusions, as follows from relations (21) and (22), the structure of inclusions (spherical shape) is present in expressions (21) and (22) through multipliers

 $\frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1}, \quad \frac{\widetilde{\varepsilon} - \varepsilon_1}{\varepsilon_1 + 2\widetilde{\varepsilon}}, \quad \frac{\widetilde{\varepsilon} - \varepsilon_2}{\varepsilon_2 + 2\widetilde{\varepsilon}}.$ These multipliers

determine within an accuracy of the value a (where a is the particle radius) the polarizability of a particle. It should be noted that the generalization of relations (21) and (22) to the case of ellipsoidal inclusions may be found in the work [1]. The particle's structure comes in relations of DEM approximation Eq. (5) partially, whereas in the case of approximations Eqs. (9), (10) and (20) they are absent at all. The importance of account of inclusions structure is especially seen in processes of absorption of microwave electromagnetic radiation in matrix systems with metal inclusions. Thus, in the case of inclusions of the spherical shape under small f the peak of absorption is found out on a frequency close to the surface plasmon frequency of inclusion $\sim \omega_p / \sqrt{3}$ [2].

The value ω_s can be found from a condition $\varepsilon'_2 + 2\varepsilon_1 = 0$, which in no way enter into relations of DEM approximation. It seems that relations (5)-(8) very well describe electrodynamical properties of various statistical mixtures in low-frequency range under large values of f. Their using makes attainable the description of electrodynamical properties of dump porous systems (sand, ground, rocks, etc...). Proceeding from the relations (5)-(8), the theoretical explanation was obtained of Archi law [15] for effective conductivity ($\widetilde{\sigma}$) of such rocks:

$$\widetilde{\sigma} = \sigma_2 f^{3/2} , \qquad (26)$$

where σ_2 is the fraction conductivity.

Thus, despite of the limitations of relations (9), (20)-(23), they can be used for calculation of electrodynamical properties of two-component systems practically in all the interval of variation of the value f ($0 \le f \le 1$).

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Эффективная диэлектрическая проницаемость матричных дисперсных систем в приближении дифференциальной эффективной среды

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Предложен теоретический подход к описанию диэлектрических свойств матричных дисперсных систем, состоящих из диэлектрической матрицы с введенными в нее металлическими включениями. На основе метода эффективной дифференциальной среды получены аналитические выражения для эффективной диэлектрической проницаемости є матричной дисперсной системы с включениями сферической и эллипсоидальной формы. Проведен анализ границ возможных значений действительной и мнимой частей ε в зависимости от параметров системы.

Ефективна діелектрична проникність матричних дисперсних систем у наближенні диференціального ефективного середовища

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Запропоновано теоретичний підхід до опису діелектричних властивостей матричних дисперсних систем, які складаються з діелектричної матриці з металічними включеннями. На основі методу ефективного диференціального середовища отримано аналітичні вирази для ефективної діелектричної проникності є матричної дисперсної системи з включеннями сферичної та еліпсоїдальної форми. Проведено аналіз меж можливих значень дійсної та уявної частин є в залежності від параметрів системи.