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Dielectric Function of Matrix Disperse Systems with Metallic Inclusions. Account of Multipole Interaction between Inclusions

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Dielectric function of small density system of fine metallic particles soluted in dielectric matrix is calculated. For that purpose, the improved variant of Maxwell-Garnett approach is developed taking into account the pair multipole interaction between inclusions. The exact solution of two metallic spheres electrostatic problem is brought in. The exact account of the pair interaction leads to the appearance in the dielectric function of non-analyticity which is result of summing up of all the series of perturbation theory. Singularities of dielectric function connected with this non-analyticity give rise to the origin of new excitation branches in a system.

Processes of propagation and absorption of electromagnetic radiation (EMR) in two-component disperse systems present a problem that has a long history [1-3]. Promoted interest to the technological aspects of such systems had led to revival of theoretical [4-21] and experimental [22-29] researches in this classical problem.

In this and also aftergoing paper we will turn attention to the study of some features of the EMR absorption in matrix disperse systems (MDS), i.e. in the systems which present a continuous matrix with the other phase inclusions adopted in a way that the volume part of the fraction intruded is small compared with the volume matrix part. The MDS with metallic inclusions (the most often of spherical form) intruded in dielectric matrix are of pronounced interest, because mainly on the basis of such MDS the composite materials are created of different aiming purposes with predicted values of dielectric and magnetic permeabilities.

Upon the theoretical study of the processes of EMR interaction with such systems the method of the effective medium is widely used. This method in the simplest case of non-magnetic media consists in that the MDS with dielectric permeability values distributed between matrix and inclusions is replaced by homogeneous and continuous medium with some effective dielectric permeability ($\tilde{\varepsilon}$) that depends as on dielectrical permeability both of a matrix (ε_0) and of intrusions (ε), as well on the intrusion concentration, and their statistical distribution in a matrix. This approach gives results being in a good agreement with an experiment only in the case if the wavelength of the radiation interacting with MDS greatly exceeds the mean particles dimensions and the distance between them (the longwave approachment). The review of early researches on this method one may find in [1-3], and its various modifications - in [1,5,7,9,12,16-18,20]. The following schemes of $\tilde{\varepsilon}(\omega)$ calculation are traditionally used for MDS with spherical inclusions of the radius r:

ATA - approximation [1,30]

$$\frac{\tilde{\varepsilon}-\varepsilon_0}{\tilde{\varepsilon}+2\varepsilon_0}=f\frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0};$$

CPA - approximation [1,30]

$$f\frac{\tilde{\varepsilon}-\varepsilon_0}{\tilde{\varepsilon}+2\varepsilon_0}=(1-f)\frac{\varepsilon-\varepsilon_0}{\varepsilon+2\varepsilon_0};$$

IDA - approximation [1,9]

$$\frac{\tilde{\varepsilon}-\varepsilon_0}{\tilde{\varepsilon}+2\varepsilon_0}\left(\frac{\varepsilon_0}{\tilde{\varepsilon}}\right)^{1/3}=(1-f);$$

where $f=\frac{4\pi}{3}r^3n$, n is the inclusion concentration.

ATA (average t-matrix approximation) is more known in a literature as Maxwell-Garnett approximation (MGT) [31], and CPA - coherent potential approximation - is often called the effective averaged approximation or mean field approach [32]. IDA (iterated dilute approximation) is based on successive application of either ATA or CPA. These treatments in some sense are complementary to each other, because they are used in different cases, that depends on MDS topology, inclusion concentration and so on. There, MGT is applied mainly to the calculation of MDS $\tilde{\varepsilon}$ with a small inclusion concentration, whereas CPA is used for statistical mixtures, i.e. when both disperse system components are mixed in equal parts. Lately, IDA-approximation has been used for calculation of electrodynamical characteristics of small particles fractal aggregates. Nevertheless, the following regulations are common for all these approaches:

- they are applicable only in a longwave approximations;
- the information about the system's structure contents in them only through a parameter f, i.e. the high

order statistical correlations in the inclusion location are not used.

In the present paper we developed the improved variant of MGT for $\tilde{\varepsilon}(\omega)$ of the matrix disperse systems with spherical inclusions. By this, in a distinction from standard MGT approachment we will account exactly the pair multipole interaction between inclusions belonging to MDS. In Supplement 1 the exact solution is adduced of electrostatic problem of two spheres behaviour in an external field. Note, that the similar problems were considered in papers [8-14, 17-21], and also in [33-38].

1. Dielectrical function

We will consider a system that consists of continuous dielectric matrix with intruded spherical particles of different kinds (noted below by indices *a, b, c*...). The dielectric permeability of a matrix is ε_0, and of particles - $\varepsilon_a, \varepsilon_b, \varepsilon_c$ ..., correspondently. Let the number of spheres of a kind a is n_a, b - n_b, c - n_c and so on. The whole particles number is $n = \sum_a n_a$, the concentration of particles of kind a is $c_a = \frac{n_a}{V}$, the whole one is $c = \frac{n}{V}$. All the system is located in an external field proportional to $e^{-i\omega t}$, and the wavelength $\lambda = \frac{2\pi c}{\omega}$ is large compared to sphere radius and the mean distance between them. For calculation of the response of such a system to the external field we will enter a following way.

Let us numerate all the particles by an index i ($1 \le i \le N$). Let $\vec{p}_i(1,\dots N)$ be the dipole moment which is acquired by the i-th particle when the system is located in an external field. It is clear, that the input in $\vec{p}_i(1,\dots N)$ comes out both from the dipole moment produced in this particle due to its polarization in a field $\vec{E}$ (the field in a medium in a place of location of the i-th inclusion in the absence of other particles), as well as from polarization of the i-th particle by a field of the rest $(N-1)$ particles. Note, that in all three calculation schemes of $\tilde{\varepsilon}$ that were indicated in an Introduction in $\vec{p}_i(1,\dots N)$ only the polarization induced in every particle by the field $\vec{E}$ is accounted. The consequent account of the influence of the field of other particles can be carried out in a following way:

$$\vec{p}_i(1,2,\dots N) = \vec{p}_i + \sum_{j\ne i} \vec{p}_{i,j} + \sum_{j<k;\, j,k\ne i} \vec{p}_{i,jk} + \vec{p}_{i,1,2,\dots i-1,i+1,\dots,N}, \tag{1}$$

where $\vec{p}_{i,j}$ is the input to $\vec{p}_i(1,\dots N)$ from two-particle interaction, $\vec{p}_{i,jk}$ - from three-particle one and so on. To find these quantities one should put consequently $N=1,2,\dots$ and solve the obtained equations for $\vec{p}_i, \vec{p}_{i,j}, \dots$. There one finds:

$$\vec{p}_i = \vec{p}_i(i);\ \vec{p}_{i,j} = \vec{p}(i,j) - \vec{p}_i(i);$$
$$\vec{p}_{i,jk} = \vec{p}(i,j,k) - \vec{p}(i,k) - \vec{p}(i,j) - \vec{p}_i(i);\ \text{and so on.} \tag{2}$$

In the present work we restrict ourselves in (1) only by first two members, i.e. we account only the pair interaction. For two spheres the quantity $\vec{p}_i(i,j)$ is found in Supplement.

To determine the effective dielectric permeability $\tilde{\varepsilon}(\omega)$ of the system we will hold the scheme first proposed by Brown [33]. In the beginning, using (1), we calculate the macroscopic medium polarization $\langle \vec{P}(\vec{r}) \rangle$ averaged over system configuration which because of the linearity of Maxwell equations will be connected with $\vec{E}$ by the relation:

$$\langle \vec{P}(\vec{r}) \rangle = \hat{K} \cdot \vec{E}. \tag{3}$$

where $\hat{K}$ is a linear operator.

In the second, as follows again from Maxwell equations the mean Lorentz field $\langle \vec{F} \rangle$ in a electrostatic limit is connected with the polarization by the relation:

$$\langle \vec{F}(\vec{r}) \rangle = \vec{E} + \int' G(\vec{r} - \vec{r}\,') \langle \vec{P}(\vec{r}\,') \rangle d\vec{r}\,' \tag{4}$$

where

$$G_{\alpha\beta}(\vec{r}) = \frac{\partial^2}{\partial x_\alpha \partial x_\beta} \frac{1}{r}, \tag{5}$$

and the prime by an integral means that the last is considered in a sense of the principal value, i.e. the integration is produced over all the space excluding a sphere of a radius δ ($\delta \to 0$) near the Green function pole $G(\vec{r} - \vec{r}\,')$ which origins when $\vec{r} \to \vec{r}\,'$ [35-38].

Forth, after excluding $\vec{E}$ from (3) and (4) with account of relation [33-36]

$$\langle\vec{P}\rangle=\frac{3}{4\pi}\cdot\frac{\tilde{\varepsilon}-\varepsilon_0}{\tilde{\varepsilon}+2\varepsilon_0}\langle\vec{F}\rangle \qquad (6)$$

one can obtain the needed expression for effective dielectric permeability $\tilde{\varepsilon}(\omega)$. Note that a given calculation scheme appears the more preferable as compared with one proposed in [11,12] because in the final result $\tilde{\varepsilon}$ comes in immediately through Clausius-Mosotti factor [36]

$$\tilde{\chi}=\frac{3}{4\pi}\cdot\frac{\tilde{\varepsilon}-\varepsilon_0}{\tilde{\varepsilon}+2\varepsilon_0},$$

which in the scheme of papers [11,12] may appear only after summing up the infinite series of a certain class of diagrams of the perturbation theory over particles polarizability [17].

Now, we determine the explicit sight of operator $\hat{K}$. To make it, we fulfil the averaging of (1) using the n-particle distribution function $\Phi(\vec{r}_1,\vec{r}_2,\ldots r_N)$ of system configurations where

$$\frac{\Phi(\vec{r}_1,\vec{r}_2,\ldots r_N)}{V^N}d\vec{r}_1 d\vec{r}_2\ldots d\vec{r}_N \qquad (7)$$

is the probability to find the first particle center in a volume $d\vec{r}_1$, the second particle center - in $d\vec{r}_2$, and so on. When accounting that the i-th particle is fixed and restricting in (1) with only first two members, we find

$$\langle\vec{P}_i(1,\ldots n)\rangle=\vec{P}_i+\frac{1}{V}\sum_{j\neq 1}\int_V\vec{P}_{i,j}(\vec{r}_i,\vec{r}_j)\Phi_{i,j}(\vec{r}_i,\vec{r}_j)d\vec{r}_j \qquad (8)$$

where $\Phi_{i,j}(\vec{r}_i,\vec{r}_j)$ is the pair distribution function [39].

Now, having summarized in (8) over i from 1 to n and dividing both parts of the equation by V, after some transformations we obtain:

$$\langle\vec{P}\rangle=c\sum_a c_a\vec{p}_a+c^2\sum_{a,b}c_a c_b\int d\vec{r}_{a,b}\vec{p}_{a,b}(\vec{r}_a,\vec{r}_b)\Phi_{a,b}(\vec{r}_a,\vec{r}_b) \qquad (9)$$

where

$$\Phi_{a,b}(\vec{r}_a,\vec{r}_b)=\left(1-\frac{\delta_{ab}}{N}\right)\Phi_{ij}(\vec{r}_i,\vec{r}_j),$$

and $\vec{p}_i=\vec{p}_a$; $p_{a,b}(\vec{r}_a,\vec{r}_b)=p_{i,j}(\vec{r}_i,\vec{r}_j)$ when the index i corresponds to particle's kind a, and j - to the kind b.

Inverting now the expression (9) with the same accuracy one can find also the field $\vec{E}$.

$$\vec{E}=\frac{\langle\vec{P}\rangle}{c\sum_a c_a\alpha_a}-\frac{1}{\left(\sum_a c_a\alpha_a\right)^2}\sum_{a,b}c_a c_b\int d\vec{r}_{a,b}\Phi(\vec{r}_a,\vec{r}_b)\hat{\beta}_{a,b}(\vec{r}_a,\vec{r}_b)\langle\vec{P}\rangle \qquad (10)$$

When obtaining (10) it was taken into account that

$$\vec{p}_a=\alpha_a\vec{E},\ \alpha_a=\frac{\varepsilon_a-\varepsilon_0}{\varepsilon_a+2\varepsilon_0}r_a^3,$$

and the tensor $\hat{\beta}_{a,b}(\vec{r}_a,\vec{r}_b)$ which determines the bond between $\vec{p}_{a,b}(\vec{r}_a,\vec{r}_b)$ and $\vec{E}$ has a form

$$p^{\gamma}_{a,b}(\vec{r}_a,\vec{r}_b)=\sum_\sigma\beta^{\gamma\sigma}_{a,b}(\vec{r}_a,\vec{r}_b)E_\sigma, \qquad (11)$$

γ and σ number the space variables x, y, z. Note, that from (2) follows

$$\vec{p}_{a,b}(\vec{r}_a,\vec{r}_b)=\vec{p}_a(a,b)-\vec{p}_a, \qquad (12)$$

i.e.

$$\beta^{\gamma,\sigma}_{a,b}(\vec{r}_a,\vec{r}_b)=\beta^{\gamma\sigma}_a(\vec{r}_a,\vec{r}_b)-\alpha_a\delta_{\gamma\delta}, \qquad (13)$$

and the tensor $\beta^{\gamma\sigma}_a(\vec{r}_a,\vec{r}_b)$ appearance is found from the solution of the correspondent two-particle problem (Supplement 1):

$$p^{\gamma}_a(a,b)=\beta^{\gamma\sigma}_a(\vec{r}_a,\vec{r}_b)E^\sigma. \qquad (14)$$

After exclusion of $\vec{E}$ from relations (10) and (4), we obtain:

$$\langle\vec{F}\rangle=\frac{\langle\vec{P}\rangle}{\sum_a c_a\alpha_a}-\frac{\sum_{a,b}c_a\alpha_a\int d\vec{r}_a\Phi_{a,b}(\vec{r}_a,\vec{r}_b)\hat{\bar{\beta}}_{a,b}(\vec{r}_a,\vec{r}_b)\langle\vec{P}\rangle}{\left(\sum_a n_a\alpha_a\right)^2}. \qquad (15)$$

The tensor $\hat{\bar{\beta}}_{a,b}(\vec{r}_a,\vec{r}_b)$, as follows from Supplement 1 can be presented in a form

$$\hat{\beta}_{a,b}^{l,m}(\vec{r}_a,\vec{r}_b) = \beta_{a,b}^{\parallel} n_l n_m + \beta_{a,b}^{\perp}(\delta_{l,m} - n_l n_m), \quad (16)$$

where $\vec{n} = \frac{\vec{R}_{a,b}}{R_{a,b}}$; $\vec{R}_{a,b} = \vec{r}_a - \vec{r}_b$; m, l mark the space coordinates x, y, z, and quantities $\beta_{a,b}^{\parallel}$ and $\beta_{a,b}^{\perp}$ are the functions of $R_{a,b}$ equal to

$$\beta_{a,b}^{\parallel} = A_{10}^{+}(R_{ab}) r_a^3 - \alpha_a - 2\frac{\alpha_a \alpha_b}{R_{a,b}^3}, \quad (17)$$

$$\beta_{a,b}^{\perp} = A_{11}^{+}(R_{ab}) r_a^3 - \alpha_a + \frac{\alpha_a \alpha_b}{R_{a,b}^3}.$$

Here A_{10}^{+} and A_{11}^{+} are determined by the system of equations (S.8). When obtaining (17), we have accounted that

$$\frac{\partial^2}{\partial x_m \partial x_l} \cdot \frac{1}{r} = \frac{1}{r^3}\left[2n_l n_m - (\delta_{lm} - n_l n_m)\right]; \ \vec{n} = \frac{\vec{r}}{r}, \quad (18)$$

and also the relation

$$\int d\vec{r}_b \Phi_{a,b}(\vec{r}_a,\vec{r}_b) \hat{G}(\vec{r}_a - \vec{r}_b) \langle \vec{P} \rangle = \int d\vec{r}_b \hat{G}(\vec{r}_a - \vec{r}_b) \langle \vec{P} \rangle,$$

which follows from the fact that distribution function $\Phi_{a,b}(\vec{r}_a,\vec{r}_b)$ for rigid spheres depends only on $|\vec{r}_a - \vec{r}_b|$ when $|\vec{r}_a - \vec{r}_b| \to 0$, and when $|\vec{r}_a - \vec{r}_b| \to \infty$, $\Phi_{a,b}(\vec{r}_a,\vec{r}_b) \to 1$. With account of (6) the equation (15) turns to the form

$$\frac{4\pi}{3} \cdot \frac{\tilde{\varepsilon} + 2\varepsilon_0}{\tilde{\varepsilon} - \varepsilon_0} \langle \vec{P} \rangle = \frac{\langle \vec{P} \rangle}{c \sum_a c_a \alpha_a} - \sum_{a,b} c_a c_b \int d\vec{r}_b \Phi_{a,b}(\vec{r}_a,\vec{r}_b) \hat{\beta}_{a,b}(\vec{r}_a,\vec{r}_b) \langle \vec{P} \rangle.. \quad (19)$$

After integration in relation (19) over the angle we obtain the equation for finding $\tilde{\varepsilon}$:

$$\frac{4\pi}{3} \cdot \frac{\tilde{\varepsilon} + 2\varepsilon_0}{\tilde{\varepsilon} - \varepsilon_0} = \frac{1}{c \sum_a c_a \alpha_a} - \frac{\frac{4\pi}{3} \sum_{a,b} c_a c_b \int_0^\infty R_{a,b}^2 dR_{a,b} \Phi_{a,b}(R_{ab}) \left[\beta_{a,b}^{\parallel}(R_{ab}) + 2\beta^{\perp}(R_{ab})\right]}{\left(\sum_a c_a \alpha_a\right)^2}. \quad (20)$$

This is the ground formula for following investigation. To obtain (20) we assumed that $\Phi_{a,b}(\vec{r}_a,\vec{r}_b) = \Phi_{a,b}(|\vec{r}_a - \vec{r}_b|)$. The tensor $\hat{\beta}_{a,b}(\vec{r}_a,\vec{r}_b)$ in (19) accounts the pair multipole interaction between particles of the system. The formula (20) is the generalization of the known Maxwell-Garnett relation to the case of multicomponent disperse matrix systems with account of pair multipole interaction between inclusions.

2. Dielectric permeability of matrix disperse systems with single kind inclusions

In the case of the single kind particles having a radius r it follows from (20) the relation for a finding $\tilde{\varepsilon}$:

$$\frac{\tilde{\varepsilon} + 2\varepsilon_0}{\tilde{\varepsilon} - \varepsilon_0} = \frac{1 - \frac{f}{Ar^6} \int_0^\infty R^2 \Phi(R) \left[\beta^{\parallel}(R) + 2\beta^{\perp}(R)\right] dR}{fA}, \quad (21)$$

where $f = \frac{4}{3}\pi r^3 n$ is the filling power, and

$$\beta^{\parallel}(R) = \left[X_{10}(R) - A\right] r^3;$$

$$\beta^{\perp}(R) = \left[X_{11}(R) - A\right] r^3.$$

Here R - is a distance between two arbitrarily picked spheres, $A = \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0}$, and the coefficients $X_{10}(R)$ and $X_{11}(R)$ are found from the infinite systems of the catching equations (S.11)

$$\sum_{l'=1}^{\infty} T_{ll'}^{m} X_{l'm} = \delta_{1l}, \quad l = 1,2,\ldots,$$

$$T_{ll'}^{m} = \frac{\delta_{ll'}}{A_l} - (-1)^m \frac{(l+l')!}{(l+m)!(l'-m)!} \left(\frac{r}{R}\right)^{l+l'+1}, \quad (22)$$

where $A_l = \frac{l(\varepsilon - \varepsilon_0)}{l\varepsilon + (l+1)\varepsilon_0}$.

The keeping in (22) only of members with $l' = 1$ means the account only of the pair dipole-dipole

(DD) interaction between inclusions, the members with $l' = 2$ the quadrupole (KP) one and so on. In a case of $l' = 1$ we find from (22):

$$X_{10} = \left(\frac{\varepsilon + 2\varepsilon_0}{\varepsilon - \varepsilon_0} - 2\left(r/R \right)^3 \right)^{-1};$$

$$X_{11} = \left(\frac{\varepsilon + 2\varepsilon_0}{\varepsilon - \varepsilon_0} + 2\left(r/R \right)^3 \right)^{-1}; \qquad (23)$$

The expressions (23) multiplied by r^3 are presenting the longitudinal and transverse polarizabilities, correspondently, of two spheres in an external electric field with account of DD interaction between them. For the forthcoming calculation of $\tilde{\varepsilon}$ one should know the pair distribution function $\Phi(R)$. In our case the potential of interaction between spheres is of a kind:

$$u(r) = \begin{cases} \infty, & when \quad R < d = 2r \\ 0, & when \quad R > d = 2r \end{cases};$$

For such a potential it is known the exact solution of the Percus-Jevick [39] equation for the pair distribution function $\Phi(R)$ which had been obtained by Wertheim [40]. This exact solution is following:

$$\Phi\left(\frac{R}{d}\right) = \begin{cases} 1 + \dfrac{1}{12f} \displaystyle\int_0^{\infty} \frac{x^2 H(x) \sin\left(\frac{Rx}{d}\right)}{\frac{Rx}{d}} dx, & when \dfrac{R}{d} > 1; \\ 0, & when \dfrac{R}{d} < 1. \end{cases} \qquad (24)$$

Here,

$$H(x) = \frac{s^2(x)}{1 - s(x)}, \quad s(x) = 2yf \int_0^1 y^2 \frac{\sin(xy)}{xy} C(y) dy,$$

$$C(y) = -\left[\frac{(1+2f)^2}{(1-f)^4} - \frac{6f}{(1-f)^4} \left(1 + \frac{f}{2} \right)^2 y + f \frac{(1+2f)^2}{(1-f)^4} y^2 \right],$$

and f is the filling power.

In the linear over of f approximation we find:

$$\Phi\left(\frac{R}{d}\right) = \begin{cases} 0, & when \quad \dfrac{R}{d} < 1 \\ 1 + 8f\left[1 - \dfrac{3}{4}\left(\dfrac{R}{d}\right) + \dfrac{1}{16}\left(\dfrac{R}{d}\right)^3 \right], & when \quad 1 < \dfrac{R}{d} < 2 \\ 1, & when \quad \dfrac{R}{d} > 2. \end{cases} \qquad (25)$$

Note, that in the simplest case for estimates one may use the expression for $\Phi(R)$, as

$$\Phi\left(\frac{R}{d}\right) = \begin{cases} 0, & when \quad \dfrac{R}{d} < 1 \\ 1, & when \quad \dfrac{R}{d} > 1 \end{cases}; \qquad (26)$$

Thus, the effective dielectric permeability $\tilde{\varepsilon}$ of MDS with monodisperse inclusions of a single kind of particles with the radius r under account of only the pair multipole interaction between particles is completely found from the system of equations (22) and (24). In a case when $\Phi\left(\frac{R}{d}\right)$ is of a kind (26) under account of only the DD interaction we find from (21), (23) and (26):

$$\frac{\tilde{\varepsilon} + 2\varepsilon_0}{\tilde{\varepsilon} - \varepsilon_0} = \frac{1 - \dfrac{2}{3} f \left(\dfrac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} \right) \ln \dfrac{3\varepsilon + 5\varepsilon_0}{2\varepsilon + 6\varepsilon_0}}{f \dfrac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0}}. \qquad (27)$$

3. Discussion of obtained results

Let us keep briefly on the analysis of the results obtained. We have already noted that the relation (21), as well as (27) take place only in the electrostatic approach with account of the pair interaction between the MDS inclusions. In distinction from (21), in (27) only the DD interaction is accounted, and the function $\Phi\left(\frac{R}{d}\right)$ has the simplest appearance (26). When in (27) one neglects the DD interaction (the second member in a nominator) we will obtain the known relation of MGT-approximation [31]. Provided the expansion in (27) is fulfilled over f within an accuracy of f^2, two equivalent representations for $\tilde{\varepsilon}$ can be obtained:

$$\frac{\varepsilon_0}{\tilde{\varepsilon}} = 1 - 3fA - 6f^2A^2\left[-1 + \frac{1}{3}\ln\frac{8+A}{8-2A}\right],$$
$$\frac{\varepsilon_0}{\tilde{\varepsilon}} = 1 + 3fA + f^2A^2\left[3 + 2\ln\frac{8+A}{8-2A}\right], \qquad (28)$$

where $A = (\varepsilon - \varepsilon_0)/(\varepsilon + 2\varepsilon_0)$.

The first relation coincides with the result of paper [7] (formula (18)), and the second - with the result of [11] (formulas (5,7)). Besides, the relation (27) can be presented in a form which is analogous to Bergman representation for $\tilde{\varepsilon}$ [17]:

$$\tilde{t} = \frac{t - t_0}{f} - \frac{2}{9}\ln\frac{3-8t}{2-8t} = \frac{t-t_0}{f} + \frac{2}{9}\int_{1/4}^{3/8}\frac{du}{t-u}, \qquad (29)$$

where $t_0 = \frac{1-f}{3}$; $t = \frac{\varepsilon_0}{\varepsilon_0 - \varepsilon}$; $\tilde{t} = \frac{\varepsilon_0}{\varepsilon_0 - \tilde{\varepsilon}}$.

Note, that the logarithm in (27) or (28) is not a simple addition accounting for DD interaction, but is connected with the summing up of all the series of perturbation theory over this interaction [7]. The presence of the logarithm, as we will see below, leads to the set of peculiarities in the absorption and radiation processes in such systems. In a case of a proximity of the quantity ε to ε_0 the logarithm can be expanded over small parameter $A = (\varepsilon - \varepsilon_0)/(\varepsilon + 2\varepsilon_0)$. The result is the relation for finding $\tilde{\varepsilon}$ in a form of the series over parameter $x_{ab} = \frac{r_a}{r_a + r_b}$ (for single kind particles case $x_{ab} = 1$). All these results can be obtained from (27) which may be generalized easily to the case of account of the higher multipole interactions between inclusions. Besides, the scheme developed allows also the generalization to the case of three-particle or higher interactions. True, one should upon that solve the problem of behaviour of three, four or more spheres in an external electrical field, as well as know the corresponding distribution functions.

Supplement 1. Polarizability of two spheres in an electric field

Let us calculate the mutual electrostatic response (polarizability) of two spheres $A(r_a, \varepsilon_a(\omega))$ and $B(r_b, \varepsilon_b'(\omega))$ to an external electric field $\vec{E} = \vec{E}_0 \exp(-i\omega t)$. For calculation simplicity, the coordinate system is chosen as it is shown in a Fig.1. The axe Oz goes through spheres centres, the origin of coordinates is in the first sphere center, and the second sphere center coordinate is $z=R$. The spheres are located in a medium with dielectric permeability ε_0. The potential of homogeneous field $\vec{E}_0$ in a point $\vec{r}$ is

$$\psi_0 = -(\vec{E}_0\vec{r}). \qquad (S.1)$$

The expression (S.1) can be written in two equivalent forms

$$\vec{E}_0\vec{r} = E_\parallel r P_1^0(\theta) + E_\perp r P_1^1 \cos\varphi = \\ = E_0 R + E_\parallel r' P_1^0(\theta') + E_\perp r' P_0^1(\theta)\cos\varphi', \qquad (S.2)$$

where $E_\parallel$ is a field $\vec{E}$ component parallel to Oz, and $E_\perp$ is the correspondent component which is transverse to Oz axe (the field $\vec{E}$ lies in a plane xOz).

The potentials created by polarized spheres are the solutions of Laplace equation in spherical coordinates. These solutions can be represented in a form [41]

$$\psi_a^+ = \sum_{l=1}^{\infty}\sum_{m=0}^{l}\frac{A_{lm}^+ P_l^m(\theta)\cos m\varphi}{r^{l+1}};$$
$$\psi_a^- = \sum_{l=1}^{\infty}\sum_{m=0}^{l} A_{lm}^- r^l P_l^m(\theta)\cos m\varphi; \qquad (S.3)$$
$$\psi_b^+ = \sum_{l=1}^{\infty}\sum_{m=0}^{l}\frac{B_{lm}^+ P_l^m(\theta')\cos m\varphi}{r'^{l+1}};$$
$$\psi_b^- = \sum_{l=1}^{\infty}\sum_{m=0}^{l} B_{lm}^-(r') P_l^m(\theta')\cos m\varphi$$

where $P_l^m(\theta)$ are adjoined Legendre polynomials, and $\psi_a^+, \psi_a^- (\psi_b^+, \psi_b^-)$ are the solutions out and inside a sphere $A\,(B)$, correspondently.

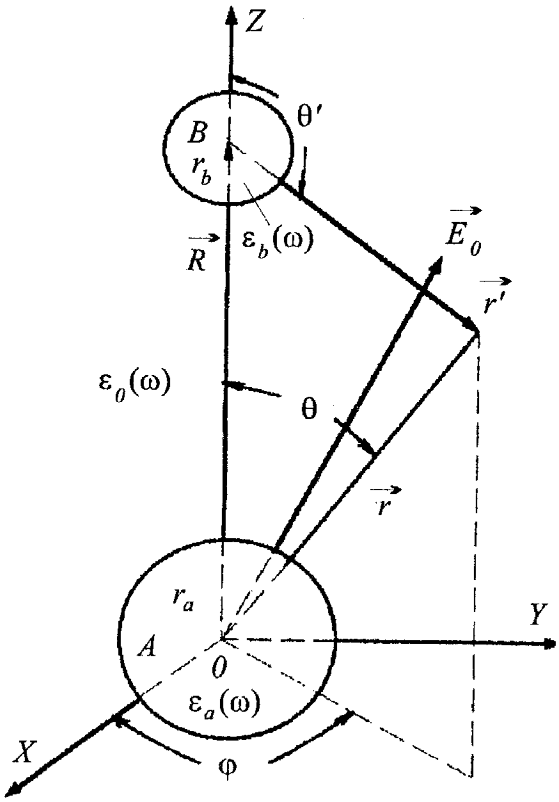

Fig. 1. *Coordinate system used for solution of problem of two spheres in external electrical field* $\vec{E}_0$.

The summing over l begins from $l=1$, provided the spheres are not charged. Representing in accordance with the superposition principle the solutions in three regions:

1. $|\vec{r}| > r_a$; $|\vec{r}\,'| > r_b$; $\psi = \psi_a^+ + \psi_b^+ + \psi_0$;
2. $|\vec{r}| < r_a$; $|\vec{r}\,'| > r_b$; $\psi_a = \psi_a^- + \psi_b^+ + \psi_0$; (S.4)
3. $|\vec{r}| > r_a$; $|\vec{r}\,'| < r_b$; $\psi_b = \psi_b^- + \psi_a^+ + \psi_0$

and taking into account the standard boundary conditions

1. when $r = r_a$ $\qquad \psi_a^+ = \psi_a^-$;

$$\varepsilon_a \frac{\partial \psi_a^-}{\partial r} - \varepsilon_0 \frac{\partial \psi_a^+}{\partial r} = -(\varepsilon_a - \varepsilon_0)\left(\frac{\partial \psi_0}{\partial r} + \frac{\partial \psi_b^+}{\partial r}\right); \tag{S.5}$$

2. when $r = r_b$ $\qquad \psi_b^+ = \psi_b^-$;

$$\varepsilon_b \frac{\partial \psi_a^-}{\partial' r'} - \varepsilon_0 \frac{\partial \psi_b}{\partial r'} = -(\varepsilon_b - \varepsilon_0)\left(\frac{\partial \psi_0}{\partial r'} + \frac{\partial \psi_a^+}{\partial r'}\right), \tag{S.6}$$

and using the transformation rules from the center of sphere A to B and inversely [42-44] which in our case are of a kind

$$\frac{P_l^m(\theta)}{r^{l+1}} = \sum_{l'=|m'|} (-1)^{l+m} \frac{P_{l'}^m(\theta')(r')^{l'}}{R^{l+l'+1}} \binom{l+l'}{l'+m},$$
$(R > r')$; (S.7)

$$\frac{P_l^m(\theta)}{(r')^{l+1}} = \sum_{l'=|m'|} (-1)^{l'+m} \frac{P_{l'}^m(\theta)(r')^{l'}}{R^{l+l'+1}} \binom{l+l'}{l'+m},$$
$(R > r)$;

where $\binom{n}{m} = \dfrac{n!}{m!(n-m)!}$ one can find that the coefficients $A_{lm}^{\pm}$ and $B_{lm}^{\pm}$ satisfy the following system of equations

$$\frac{A_{lm}^{\pm}}{\alpha_a} = E_{\parallel}\,\delta_{m0}\delta_{l1} + E_{\perp}\delta_{ml}\delta_{l1} - -(-1)^{l+m}\sum_{l'=m}\binom{l+l'}{l+m}\frac{B_{l'm}^{+}}{R^{l+l'+1}}, \tag{S.8}$$

$$\frac{B_{lm}^{\pm}}{\alpha_b} = E_{\parallel}\,\delta_{m0}\delta_{l1} + E_{\perp}\delta_{ml}\delta_{l1} - -\sum_{l'=m}(-1)^{l'+m}\binom{l+l'}{l+m}\frac{B_{l'm}^{+}}{R^{l+l'+1}}$$

Here δ_{mn} is a Kronecker symbol, and $\alpha_{li}(i = a,b)$ is the sphere l-multipole polarizability in an external electric field:

$$\alpha_{li} = \frac{l(\varepsilon_i - \varepsilon_0)}{l\varepsilon_i + (l+1)\varepsilon_0} r_i^{2l+1}, (i=a,b). \tag{S.9}$$

These formulas together with (S.3) give a complete solution of a given problem.

In an onward analysis we consider the more simple case, when spheres A and B are identical, i.e. $r_a = r_b = r$, $\varepsilon_a = \varepsilon_b = \varepsilon$. In this case the system of equations (S.8) simplifies and after involving of the variable

$$A^{+}_{lm} = (-1)^{l-1} B^{+}_{lm} = \left(E_{\parallel}\delta_{m0} + E_{\perp}\delta_{m1}\right)\delta_{l1} r^{l+2} X_{lm} \quad (S.10)$$

comes to the following infinite system

$$\sum_{l'=1}^{\infty} T^{m}_{ll'} X_{l'm} = \delta_{l1}, \; l = 1,2,\ldots, \quad (S.11)$$

where

$$T^{m}_{ll'} = \frac{r^{2i+1}}{\alpha_i}\delta_{ll'} - (-1)^m \binom{l+l'}{l+m}\left(\frac{r}{R}\right)^{l+l'+1}, \quad (S.12)$$

and α_l is given by (S.9).

The expression in the first formula (S.3) for any l gives a potential caused by the l-multipole moment. In particular, for the dipole moment ($l=1$), induced in a sphere A, we have

$$\frac{\vec{p}(1;2)\cdot\vec{r}}{r^3} = \sum_{m=0}^{l} A^{+}_{lm}\frac{1}{r^2}P^{m}_{1}(\theta)\cos(m\varphi). \quad (S.13)$$

Taking into account the appearance of the longitudinal $\left(E_{\parallel} r P^{0}_{l}(\theta)\right)$ and transverse $\left(E_{\perp} r P^{1}_{l}(\theta)\cos\varphi\right)$ parts coming in the expression for A^{+}_{lm} we find from (S.13)

$$p_i(1,2) = a^3\left[X_{10}(R) n_i n_j + X_{11}(R)\left(\delta_{ij} - n_i n_j\right)\right]E_{0j},$$
$$j = x,y,z, \quad (S.14)$$

where $\vec{n}$ is a unit vector equal to $\vec{n} = \frac{\vec{R}}{R}$, and i,j number the space variables x, y, z.

The formula (S.14) gives the value of the dipole moment of any from two spheres A or B in an external electric field E_0 with account of the mutual influence each on other. To find it one should know the quantities $X_{10}(R)$ and $X_{11}(R)$ which are determined from a system of the infinite catching equations (S.11). Note, that by the similar method one may solve also the N-particle problem providing that instead of (S.7) one should use more general relations of the transition from any i-center to any j-center [43,20-21].

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Диэлектрическая проницаемость матричных дисперсных систем с металлическими включениями с учетом мультипольного взаимодействия между ними.

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Найдена диэлектрическая проницаемость системы малой концентрации металлических частиц, растворенных в диэлектрической матрице. Для этой цели развит улучшенный вариант теории Максвелла-Гарнета, позволяющий учесть парные мультипольные взаимодействия между включениями. Точно решена электростатическая задача о поляризуемости двух металлических шаров. Произведенный точный учет парного взаимодействия приводит к неаналитичности в диэлектрической проницаемости, что является результатом суммирования всего ряда теории возмущений. Особенности диэлектрической проницаемости, связанные с указанной неаналитичностью определяют спектр новых ветвей возбуждений системы.

Діелектрична проникність матричних дисперсних систем с металевими включеннями з урахуванням мультипольної взаємодії між ними.

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Знайдено діелектричну проникність системи малої концентрації металічних частинок, розчинених у діелектричній матриці. З цією метою розвинено покращений варіант теорії Максвела-Гарнета, який дозволяє врахувати парну мультипольну взаємодію між включеннями. Точно розв'язано електростатичну задачу про поляризовність двох металічних куль. Проведене точне урахування парної взаємодії призводить до неаналітичності, що є результатом підсумування усього ряду теорії збурень. Особливості діелектричної проникності, які пов'язані з вказаною неаналітичністю, визначають спектр нових гілок збуджень системи.