ELECTRODYNAMIC PROPERTIES OF MATRIX DISPERSED SYSTEMS WITH TWO-LAYER INCLUSIONS

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Abstract

Theoretical approach for calculation of the effective dielectric permittivity $(\tilde{\varepsilon})$ of matrix dispersed systems (MDS) that consist of a dielectric matrix with randomly arranged two-layer spherical inclusions of different radiuses has been proposed and departures from the Maxwell-Garnet formula due to increasing of an inclusion volume fraction are studied. It is shown that the effects of direct dipole-dipole interactions between inclusions become important in this case. In the electrostatic approximation we have exactly solved the problem of a response of this N–particle system on the external electric field and obtained corrections to the Maxwell-Garnet formula for $\tilde{\varepsilon}$ with account of the pair dipole-dipole interaction between inclusions.

Introduction

An increasing interest to study of interaction of an electromagnetic radiation and matrix dispersed systems (MDS) displayed recently is associated first of all with the fact that these systems have some features that are absent in corresponding continuous media [1-11]. The simplest and the most efficient method of calculation of the electrodynamics characteristics of MDS is the method of an effective dielectric permittivity $(\tilde{\varepsilon})$ and an effective conductivity $(\tilde{\sigma})$. A detailed review of some methods of calculation of these quantities for different MDS are given in [1-4]. It is necessary to note that MDS constituted of a dielectric matrix with randomly embedded inclusions of different physicochemical nature and shape (spheres, ellipsoids, cylinders, and so on) are ones of the best studied. It is clear that these MDS simulate good enough real systems including soils, rocks, and even biological live objects such as suspensions of cells. Moreover, MDS and statistical blends are the basement for creation of different composite materials with the beforehand given electrodynamics properties.

In this paper, we performed calculations of $\tilde{\varepsilon}$ for MDS with two-layer spherical inclusions of different size. With the help of the method of expansion of polarizability of these systems with respect to group of particles (inclusions) [5-7] and the theorem of spherical harmonics [8-9], in the first section we solved the problem of obtaining of electric potential at an arbitrary point of the matrix consisting of N inclusions in the external electric field $\sim \vec{E}_o e^{-i\omega t}$ (in the electrostatic approximation when a wave length of the field $\lambda \sim \frac{2\pi c}{\omega}$ is much larger than a typical size of particles and an average distance between them). We obtained the relations that allow one to perform calculation of the potential of electric field with account of any order of the multipole interaction between inclusions. In the second section, we obtained the expression $\tilde{\varepsilon}$ of our MDS with account of only pair interaction between inclusions.

1. N–particle problem

We consider a system of N particles in a volume V and in the homogeneous varying with time electric field $\vec{E}_0(\vec{r},t)=\vec{E}_0(\vec{\omega})e^{-i\omega t}$. In the electrostatic approach, the field $\vec{E}(\vec{r})$ at any point of the system may be obtained from the relation $\vec{E}(\vec{r})=-grad\ \varphi(r)$, where the potential $\varphi(\vec{r})$ satisfies the equation

$$\Delta\varphi=0 \qquad (1)$$

The inclusions are two-layer spherical particles of an external radius b_i, and radius of the inclusion nucleus is a_i; $\varepsilon_1^{(i)}(\omega)$, $\varepsilon_2^{(i)}(\omega)$, ε_0 are the dielectric permittivities of the inclusion nucleus, shell and the matrix respectively (Fig.).

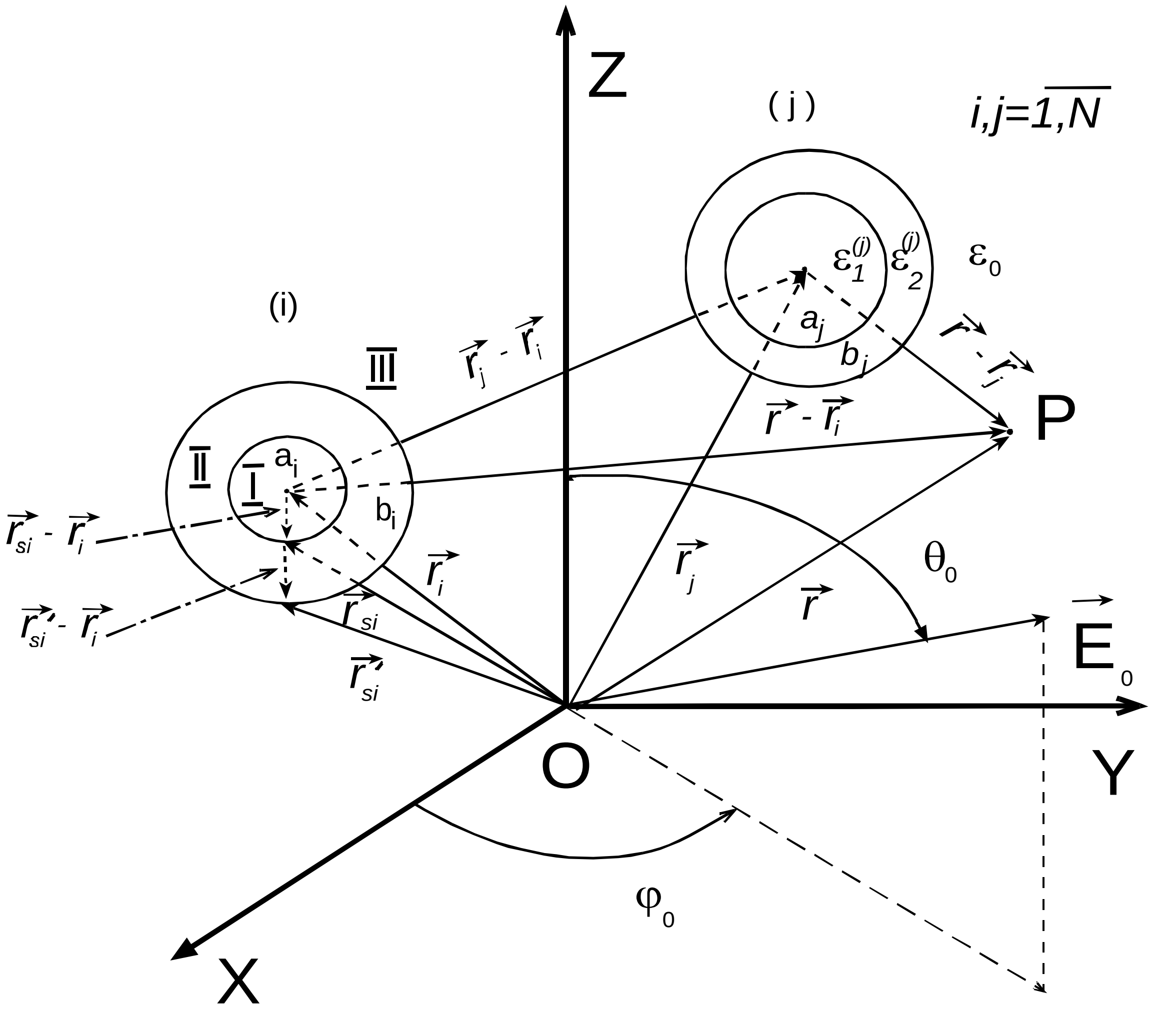

Fig. Mutual disposition of balls.

Taking into account of the problem symmetry, may present a solution of (1) with a center at the i-th particle in the following form [12]:
for the particle nucleus

$$\varphi_i^{(I)}=-E_0\sum_{l,m}A_{lm}^{(i)}\left|\vec{r}-\vec{r}_i\right|^l Y_{lm}\left(\hat{r}-\hat{r}_i\right), \qquad (2)$$

for the particle shell

$$\varphi_i^{(II)}=-E_0\sum_{l,m}\left[C_{lm}^{(i)}\left|\vec r-\vec r_i\right|^{l}+\frac{D_{lm}^{(i)}}{\left|\vec r-\vec r_i\right|^{l+1}}\right]Y_{lm}\left(\widehat{\vec r-\vec r_i}\right), \tag{3}$$

and for the matrix

$$\varphi^{(III)}(\vec r)=-E_0\sum_{lm}d_{lm}^{(i)}\left|\vec r-\vec r_i\right|^{l}Y_{lm}\left(\widehat{\vec r-\vec r_i}\right)-\vec E_0\sum_{lm}\frac{B_{lm}^{(i)}}{\left|\vec r-\vec r_i\right|^{l+1}}Y_{lm}\left(\widehat{\vec r-\vec r_i}\right)-$$

$$-E_0\sum_{\substack{j\neq i\\ p,q}}\frac{B_{p,q}^{(i)}}{\left|\vec r-\vec r_i\right|^{p+1}}Y_{p,q}\left(\widehat{\vec r-\vec r_i}\right), \tag{4}$$

where $Y_{p,q}\left(\widehat{\vec r-\vec r_i}\right)$ are spherical functions [13], $\widehat{\vec r-\vec r_i}=\dfrac{\vec r-\vec r_i}{\left|\vec r-\vec r_i\right|}$. In equation (4) the first term is the potential of the external field expanded with respect to sherical harmonics with a centered $\vec r_i$ and in the observation point P; the second term is the potential created by the i-th particle due to its polarization at the point P, the third term is sum of potentials that created by rest (N-1) particles at P. While writing expressions (2)-(4), we took into account regularity of the potential $\varphi_i^{(I)}$ at the center of the i-th sphere and at infinity $\varphi^{(III)}(\vec r)$. Unknown constant $A_{lm}^{(i)},B_{lm}^{(i)},C_{lm}^{(i)},D_{lm}^{(i)}$, may be obtained requiring solutions (2)-(4) must satisfy the boundary conditions [13]

$$\left.\varphi_i^{(I)}\right|_{\vec r=\vec r_{si}}=\left.\varphi_i^{(II)}\right|_{\vec r=\vec r_{si}};\ \left.\varepsilon^{(i)}\left(\vec n_i\cdot grad\varphi_i^{(j)}\right)\right|_{\vec r=\vec r_{si}}=\left.\varepsilon_2^{(i)}\left(\vec n_i\cdot grad\varphi^{(II)}\right)\right|_{\vec r=\vec r_{si}} \tag{5}$$

$$\left.\varphi_i^{(II)}\right|_{\vec r=\vec r_{si}'}=\left.\varphi^{(II)}\right|_{\vec r=\vec r_{si}'};\ \left.\varepsilon_2^{(i)}\left(\vec n_i\cdot grad\varphi_i^{(I)}\right)\right|_{\vec r=\vec r_{si}'}=\left.\varepsilon_0\left(\vec n_i\cdot grad\varphi^{(III)}\right)\right|_{\vec r=\vec r_{si}'}$$

where vectors $\vec r_{si},\vec r_{si}'$ are shown at Fig., vectors $\vec r_{si}-\vec r_i$ and $\vec r_{si}'-\vec r_i$ are parallel to the external normal $\vec n_i$; $\left|\vec r_{si}-\vec r_i\right|=a_i$, $\left|\vec r_{Si}'-\vec r_i\right|=b_i$.

Since the spherical harmonics in the third term in (4) are centered at the j-center, we could not meet conditions (5) directly. It is necessary to reduce everything to the i-th center. For this we may use the addition theorems of spherical harmonics. One of these theorems at a domain $\left|\vec r-\vec r_i\right|<\left|\vec r_j-\vec r_i\right|$ gives [8-9]:

$$\frac{Y_{lm}\left(\vec r-\vec r_j\right)}{\left|\vec r-\vec r_j\right|^{l+1}}=\sum_{l'm'}a_{l'm'}^{lm}\left(\vec r_j-\vec r_i\right)\left|\vec r-\vec r_i\right|^{l'}Y_{l'm'}\left(\widehat{\vec r-\vec r_i}\right), \tag{6}$$

where $$a_{l'm'}^{lm}\left(\vec r_j-\vec r_i\right)\frac{Y^{*}_{l+l',m'-m}\left(\widehat{\vec r_j-\vec r_i}\right)}{\left|\vec r_j-\vec r_i\right|^{l+l'+1}}(-1)^{l+m}\left[\frac{4\pi(2l+1)}{(2l'+1)(2l+2l'+1)}\right]K_{l'm'}^{lm}$$

$$K_{l'm'}^{lm}=K_{lm}^{l'm'}=\left[\frac{(l+l'+m-m')!\,(l+l'+m'-m)!}{(l'+m')!\,(l'-m')!\,(l+m)!\,(l-m)!}\right]^{1/2} \tag{7}$$

Substituting (2)-(4) and (6) in (5), we obtain:

$$\begin{cases} A_{lm}^{(i)} - C_{lm}^{(i)} = \dfrac{D_{lm}^{(i)}}{a_i^{2l+1}}; \\ A_{lm}^{(i)} - \dfrac{\varepsilon_2^{(i)}}{\varepsilon_1^{(i)}} C_{lm}^{(i)} = -\dfrac{\varepsilon_2^{(i)}}{\varepsilon_1^{(i)}} \cdot \dfrac{(l+1)}{l} \cdot \dfrac{D_{lm}^{(i)}}{a_i^{2l+1}}; \\ C_{lm}^{(i)} + \dfrac{D_{lm}^{(i)}}{b_i^{2l+1}} = N_{lm}^{(i)} + \dfrac{B_{lm}^{(i)}}{b_i^{2l+1}}; \\ C_{lm}^{(i)} - \dfrac{(l+1)}{l} \cdot \dfrac{D_{lm}^{(i)}}{b_i^{2l+1}} = \dfrac{\varepsilon_0}{\varepsilon_2^{(i)}} \left[N_{lm}^{(i)} + \dfrac{B_{lm}^{(i)}}{b_i^{2l+1}} \cdot \dfrac{(l+1)}{l} \right]; \end{cases} \tag{8}$$

where

$$N_{lm}^{(i)} = d_{lm}^{(i)} + \sum_{\substack{l'm' \\ j \neq i}} B_{l'm'}^{(j)} a_{lm}^{l'm'} \left(\vec{r}_j - \vec{r}_i \right). \tag{9}$$

From (8) and (9), we may obtain the system of algebraic equations for the coefficients $B_{em}^{(i)}$

$$\frac{B_{lm}^{(i)}}{\alpha_l^{(i)}} + \sum_{j \neq i}^{N} \sum_{l'=1}^{\infty} \sum_{m'=-l}^{l} B_{l'm'}^{(j')} \; a_{lm}^{l'm'} \left(\vec{r}_j - \vec{r}_i \right) = -d_{lm}^{(i)} \quad , \tag{10}$$

where $d_{lm}^{(i)}$ are coefficients of expansion of the external electric field with respect spherical functions that are centered at the i-th inclusion, and

$$\alpha_l^{(i)} = \frac{l b_i^{2l+1} \left\{ \left[\varepsilon_2^{(i)} (2l+1) + l \varepsilon_1^{(i)} \right] \cdot \left(\varepsilon_2^{(i)} - \varepsilon_0 \right) + \left(\varepsilon_1^{(i)} - \varepsilon_2^{(i)} \right) \cdot \left[(l+1) \varepsilon_2^{(i)} + l \varepsilon_0 \right] q_i^{2l+1} \right\}}{\left[\varepsilon_2^{(i)} (l+1) + l \varepsilon_1^{(i)} \right] \cdot \left[(l+1) \varepsilon_0 + l \varepsilon_2^{(i)} \right] + l(l+1) \left(\varepsilon_1^{(i)} - \varepsilon_2^{(i)} \right) \left(\varepsilon_2^{(i)} - \varepsilon \right) q_i^{2l+1}}, \tag{11}$$

$q_i = \dfrac{a_i}{b_i}$, $a_{lm}^{l'm'} \left(\vec{r}_j - \vec{r}_i \right)$ are given by (7). After obtaining $B_{lm}^{(i)}$, we may easily get coefficients $A_{lm}^i, C_{lm}^i, D_{lm}^i$ from (8).

Relations (7)–(8) and (10)–(11) completely solve the problem of electric response of the system of N two-layer spheres on the field $\vec{E}_0$ accounting of the direct multipole interaction between particles. Keeping in (10) terms of the order $l = l' = 1$, we take into account only the dipole-dipole interaction between them. Picking up terms with $l = l' = 2$, we may take into account the quadrupole interaction, and so on.

It is worth noting that $\alpha_l^{(i)}$ is the polarization of the i-th particle of the order l in the field $\vec{E}_0$. At $l = 1$ (dipole polarization) formula (11) transforms into formula (5.36) [2], and at $q_i = 0$, $\alpha_l^{(i)}$ coincides with the l order multipole polarization of the i-th sphere–balls [6].

2. Effective dielectric permittivity of MDS with two-layer inclusions

Comparing relations (10) and (11) with corresponding formulas of papers [7, 10, 11] (for example, formulas (10) and (11) from [10]) we may note that the system of equations for $B_{lm}^{(i)}$ in the case of MDS with two-layer inclusions differ from equations of MDS for one-layer inclusions by the magnitude of the multipole polarization of the l-th order ($\alpha_l^{(i)}$) of a particular inclusion. In other words, to obtain $\tilde{\varepsilon}$ of the MDS systems under consideration one may use directly by formulas of papers [7, 10, 11] changing relation $\alpha_l^{(i)}$ with (11). For example, by using formula (1) of paper [11], we may straight forward obtain $\tilde{\varepsilon}$ for MDS with two-layer spherical inclusions accounting the pair dipole interaction between them:

$$\frac{\tilde{\varepsilon}+2\varepsilon_0}{\tilde{\varepsilon}-\varepsilon_0}=\frac{1}{\frac{4}{3}\pi\sum_k n_k\alpha_1^{(k)}}-\frac{1}{\left(\sum_k n_k\alpha_1^{(k)}\right)^2}\times \quad . \tag{12}$$

$$\sum_{k,p} n_k n_p \int_0^\infty R_{kp}^2\phi\left(R_{kp}\right)\cdot dR_{ab}\left[\beta_{kp}^{II}\left(R_{kp}\right)+2\beta_{kp}^{\perp}\left(R_{kp}\right)\right]$$

Here coefficients $\alpha_1^{(i)}$ are given by relation (11) at $l=1$, $R_{kp}=\left|\vec{r}_k-\vec{r}_p\right|$, $n_K=\frac{N_K}{V}$ is a density number of the k-th type of particles, $\phi\left(R_{kp}\right)$ is the two-particle distribution function of inclusions that was chosen in the form:

$$\phi\left(R_{kp}\right)=\begin{cases}0, & if \quad R_{kp}<b_k+b_p,\\ 1, & if \quad R_{kp}\geq b_k+b.\end{cases} \tag{13}$$

In the case of pair dipole interaction ($l=l'=1$), the coefficients β_{ij}^{II} i $\beta_{ij}^{\perp}$ take the form:

$$\beta_{ij}^{II}=\chi_{10}^{(i)}\left(R_{ij}\right)-\alpha_1^{(i)}-2\frac{\alpha_1^{(i)}\alpha_1^{(j)}}{R_{ij}^3}, \tag{14}$$

$$\beta_{ij}^{\perp}=\chi_{11}^{(i)}\left(R_{ij}\right)-\alpha_1^{(i)}+\frac{\alpha_1^{(i)}\alpha_j^{(j)}}{R_{ij}^3}.$$

Expressions of $\chi_{10}^{(i)}\left(R_{ij}\right)$ and $\chi_{11}^{(i)}\left(R_{ij}\right)$ (at $l=l'=1$) may be obtained from (10)

$$\chi_{10}^{(i)}\left(R_{ij}\right)=\frac{1}{2}b_i^3\left[\frac{\left(\frac{B_i}{B_j}\right)^{1/2}+\Delta_{ij}^{3/2}}{\left(\frac{1}{B_iB_j}\right)^{1/2}-2\Delta_{ij}^{3/2}\delta_i^3}+\frac{\left(\frac{B_i}{B_j}\right)^{1/2}-\Delta_{ij}^{3/2}}{\left(\frac{1}{B_iB_j}\right)^{1/2}+2\Delta_{ij}^{3/2}\delta_i^3}\right], \tag{15}$$

$$\chi_{10}^{(i)}\left(R_{ij}\right)=\frac{1}{2}b_i^3\left[\frac{\left(\frac{B_i}{B_j}\right)^{1/2}+\Delta_{ij}^{3/2}}{\left(\frac{1}{B_iB_j}\right)^{1/2}-\Delta_{ij}^{3/2}\delta_i^3}+\frac{\left(\frac{B_i}{B_j}\right)^{1/2}-\Delta_{ij}^{3/2}}{\left(\frac{1}{B_iB_j}\right)^{1/2}+\Delta_{ij}^{3/2}\delta_i^3}\right],$$

where $\Delta_{ij}=\frac{b_j}{b_i}$, $\delta_i=\frac{b_i}{R_{ij}}$ and $B_i=\frac{\alpha_1^{(i)}}{b_i^3}$.

Taking into account the explicit form of $\phi\left(R_{ij}\right)$ (13) and relations (14)-(15) and using (12), we obtain the formula of effective dielectric permittivity $\tilde{\varepsilon}$ of MDS with two-layer spherical inclusions in the approximation of dipole-dipole interaction between them:

$$\frac{\tilde{\varepsilon}+2\varepsilon_0}{\tilde{\varepsilon}-\varepsilon_0}=\frac{1}{\frac{4\pi}{3}\sum_k n_k\alpha_1^{(k)}}-\frac{1}{3\left(\sum_k n_k\alpha_1^{(k)}\right)^2}\sum_{k,p} n_k\cdot n_p\cdot\alpha_k^{(1)}\cdot\alpha_p^{(1)}\times\cdot\left\{\left[1+\frac{b_k^3}{b_p^3}\cdot\left(\frac{\alpha_1^{(p)}}{\alpha_1^{(k)}}\right)^{1/2}\right]\times\right.$$

$$\left.\times\ln\frac{(b_k+b_P)^3+\left(\alpha_1^{(k)}\alpha_1^{(p)}\right)^{1/2}}{(b_k+b_P)^3-2\left(\alpha_1^{(k)}\alpha_1^{(p)}\right)^{1/2}}\right]+\left[1-\frac{b_k^3}{b_p^3}\cdot\left(\frac{\alpha_1^{(p)}}{\alpha_1^{(k)}}\right)^{1/2}\right]\ln\frac{(b_k+b_P)^3-\left(\alpha_1^{(k)}\alpha_1^{(p)}\right)^{1/2}}{(b_k+b_P)^3+2\left(\alpha_1^{(k)}\alpha_1^{(p)}\right)^{1/2}}\left.\right]\right\}. \tag{16}$$

Neglecting the second term in the right hand side of (16), we obtain the Maxwell-Garnet relation for MDS with two-layer inclusions which represent mixture of particles with different sizes and different electrodynamics characteristics (different $\varepsilon_{1,2}^{(i)}$). In the case of identical particles, relation (16) gives:

$$\tilde{\varepsilon}=\varepsilon_0\left[1+\frac{3fB}{1-fB-\frac{2}{3}fB\ln\frac{8+B}{8-2B}}\right], \tag{17}$$

where $a_i=a_j=a;\ b_i=b_j=b;\ \varepsilon_1^{(i)}=\varepsilon_1^{(j)}=\varepsilon_1;\ \varepsilon_2^{(i)}=\varepsilon_2^{(j)}=\varepsilon_2$

$$B=\frac{\alpha_1}{b^3}=\frac{(2\varepsilon_2+\varepsilon_1)(\varepsilon_2-\varepsilon_0)+(\varepsilon_1-\varepsilon_2)(2\varepsilon_2+\varepsilon_0)q^3}{(2\varepsilon_2+\varepsilon_1)(2\varepsilon_0-\varepsilon_2)+2(\varepsilon_1-\varepsilon_2)(\varepsilon_2+\varepsilon_0)q^3}$$

$q=\frac{a}{b}$; $f=\frac{4}{3}\pi b^3 n$, n is a density number of inclusions. At $fB<<1$, (17) gives

$$\frac{\varepsilon_0}{\tilde{\varepsilon}}=1-3fB-2(fB)^2\ln\left[\ln\frac{8+B}{8-2B}-3\right]+...; \tag{18}$$

or

$$\frac{\tilde{\varepsilon}}{\varepsilon_0}=1+3fB+f^2B^2\left[3+2\ln\frac{8+B}{8-2B}\right]+... \quad . \tag{19}$$

Formula (18) coincides with formula (18) of paper [14], and formula (19) coincides with formulas (5) and (7) of paper [6], provided that $q=0\ (a\to 0)$.

Conclusion

The results obtained in this paper allow us to make some general conclusions. The form of system of equations (10)-(11) shows that the problem of obtaining the coefficients $B_{em}^{(i)}$ and the effective dielectric permittivity $\tilde{\varepsilon}$ of MDS with complex spherical inclusions (one-layer of different radius, two-layer, inhomogeneous, and so on) is completely equivalent to the problem of obtaining these coefficients and $\tilde{\varepsilon}$ of MDS with continuous spherical inclusions but with a new magnitude of the multipole polarization of an individual inclusion $\alpha_l^{(i)}$. This is a result of the boundary conditions (8), since corrections due to the multipole interaction (coefficients $B_{lm}^{(j)}$ in (18) are added to the interaction coefficients of inclusions with the external field $d_{lm}^{(i)}$, formula (9). An analogous picture takes place for MDS with ellipsoidal inclusions. We would like to stress that the above-developed calculation method of effective dielectric permittivity $\tilde{\varepsilon}$ for similar MDS enables one to take into account even the higher term of mulipole interaction between inclusions. However, this requires knowledge of the many-particle (three, four, and so on) statistical distribution functions of inclusions in a matrix. Unfortunately, exact expressions of these functions are unknown at present with the exception of the two-particle function [15].

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