

# Dependence of the dielectric properties of suspensions on the volume fraction of suspended particles

Constantino Grosse<sup>a,b,\*</sup>, Vladimir Nikolaievich Shilov<sup>c</sup>

<sup>a</sup> Department of Physics, National University of Tucumán, Argentina

<sup>b</sup> Consejo Nacional de Investigaciones Científicas y Técnicas, Argentina

<sup>c</sup> Institute of Bio-Colloid Chemistry, National Academy of Sciences of Ukraine, Kiev, Ukraine

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## Abstract

It is shown that the fundamental expression for the complex permittivity  $\varepsilon_s^*$  of a dilute suspension of monodispersed, spherical particles,

$$\varepsilon_s^* = \varepsilon_c^*(1 + 3\phi d^*),$$

where  $\varepsilon_c^*$  is the complex permittivity of the suspending medium and  $d^*$  the dipolar coefficient, is strictly valid for any value of the volume fraction  $\phi$  of particles in the suspension, provided that  $d^*$  is interpreted as the ensemble average value of the dipolar coefficient of the particles and is defined in terms of the macroscopic electric field in the suspension.

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## 1. Introduction

It is generally accepted [1–7] that the fundamental expression for the complex permittivity  $\varepsilon_s^*$  of a suspension of homogeneous, monodispersed, spherical particles,

$$\varepsilon_s^* = \varepsilon_c^*(1 + 3\phi d^*), \quad (1)$$

where  $\varepsilon_c^*$  is the complex permittivity of the suspending medium and  $d^*$  the dipolar coefficient, is only valid for low values of the volume fraction  $\phi$  of particles in the suspension.

In this work we review three deductions of this expression, taking care to avoid all the usual approximations involving the assumption  $\phi \ll 1$ . We show that all these formulations lead to the above expression, which appears to be valid for any value of the volume fraction, provided that  $d^*$  is interpreted as proportional to the ensemble average value of the dipolar coefficient of the particles and is defined in terms of the macroscopic electric field in the suspension.

In order to simplify the presentation, we first restrict the deductions to the calculation of the permittivity of nonconducting systems. An extension of the obtained results to the calculation of the complex permittivity of conducting systems is presented afterward.

## 2. Deduction based on the Maxwell–Wagner approach

The permittivity of a suspension can be defined in the following way [8,9]. We consider two macroscopic spherical samples with the same radius  $R$ , one made of the suspension, and the other of some homogeneous material, Fig. 1. We now

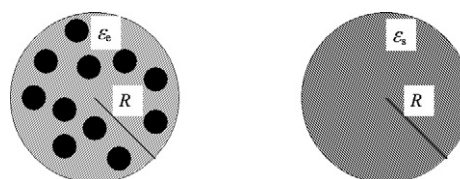


Fig. 1. Macroscopic spheres made of the suspension and of a homogeneous material, used in the Maxwell–Wagner approach.

\* Corresponding author.

E-mail address: [cgrosse@herrera.unt.edu.ar](mailto:cgrosse@herrera.unt.edu.ar) (C. Grosse).

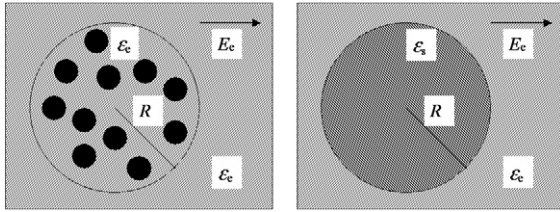


Fig. 2. Macroscopic heterogeneous and homogeneous spheres immersed in the unbound suspending medium in the presence of an initially uniform electric field.

choose the permittivity of this material in such a way that, when immersed in the same external medium and acted upon by the same external field, the total field outside the two spheres is also the same. The permittivity obtained in this way is defined as being the permittivity of the suspension:  $\varepsilon_s$ .

Since, by hypothesis, the dielectric behavior of the two spheres must always be the same, we are free to choose the configuration that leads to the result in the simplest possible way. We therefore consider that both spheres are immersed in the same medium in which the particles are suspended, which is characterized by a permittivity  $\varepsilon_e$ , Fig. 2.

A uniform electric field  $E_e$  directed along the  $x$  axis is now applied to both systems. The resulting potential outside the sphere (1) made of the suspension is determined by the sum of the contributions of its particles,

$$U_e^{(1)} = -E_e r \cos \theta + NV \frac{\langle p \rangle \cos \theta}{4\pi \varepsilon_0 r^2}, \quad (2)$$

where  $r$ ,  $\theta$  are spherical coordinates in a system with origin centered on the sphere and polar axis directed parallel to the applied field,  $N$  is the number of particles per unit volume,  $V = 4\pi R^3/3$  is the volume of the sample,

$$\langle p \rangle = \frac{1}{NV} \left| \sum_{i=1}^{NV} \mathbf{p}_i \right|$$

is the average value of the dipole moment of a particle (including the polarization charges in the surrounding medium), and  $\varepsilon_0$  is the absolute permittivity of free space. Actually, Eq. (2) is rigorously valid only far from the sphere, since the particles are not located at its center. However, this is not a limitation of this particular deduction, but a general result stating that it is impossible to express all the properties of a heterogeneous material by a homogeneous one.

The potential inside and outside the homogeneous sphere (2), solution of the Laplace equation, has the following form:

$$U_s^{(2)} = Ar \cos \theta, \\ U_e^{(2)} = -E_e r \cos \theta + B \frac{\cos \theta}{r^2}.$$

The coefficients  $A$  and  $B$  can be determined using the following boundary conditions on the surface of sphere (2):

1. Continuity of the electric potential:

$$U_s^{(2)}(R, \theta) = U_e^{(2)}(R, \theta).$$

2. Continuity of the radial component of the electric displacement:

$$-\varepsilon_s \frac{\partial U_s^{(2)}(r, \theta)}{\partial r} \Big|_R = -\varepsilon_e \frac{\partial U_e^{(2)}(r, \theta)}{\partial r} \Big|_R.$$

The resulting expressions for the potentials are

$$U_s^{(2)} = -\frac{3\varepsilon_e}{\varepsilon_s + 2\varepsilon_e} E_e r \cos \theta, \quad (3)$$

$$U_e^{(2)} = -E_e r \cos \theta + \frac{\varepsilon_s - \varepsilon_e}{\varepsilon_s + 2\varepsilon_e} R^3 E_e \frac{\cos \theta}{r^2}. \quad (4)$$

Equating the expressions for the potentials outside both spheres, Eqs. (2) and (4), leads to the following result for the permittivity of the suspension:

$$\varepsilon_s = \varepsilon_e \frac{1 + 2NV \frac{\langle p \rangle}{4\pi \varepsilon_0 R^3 E_e}}{1 - NV \frac{\langle p \rangle}{4\pi \varepsilon_0 R^3 E_e}}. \quad (5)$$

Writing the average dipole moment  $\langle p \rangle$  in terms of the dipolar coefficient,

$$\frac{\langle p \rangle}{4\pi \varepsilon_0} = da^3 E_e, \quad (6)$$

where  $a$  is the radius of a particle, transforms Eq. (5) into

$$\varepsilon_s = \varepsilon_e \frac{1 + 2\phi d}{1 - \phi d}, \quad (7)$$

where

$$\phi = NV a^3 / R^3$$

is the volume fraction occupied by the particles in the suspension. For low concentration values ( $\phi \ll 1$ ), Eq. (7) reduces to

$$\varepsilon_s = \varepsilon_e (1 + 3\phi d). \quad (8)$$

While this deduction gives the impression that the assumption  $\phi \ll 1$  is only made when going from Eq. (7) to Eq. (8), it is actually made earlier, when Eq. (6) is written, since in this expression the dipolar coefficient is related to the electric field in the suspending medium far from any particle. Expressing the dipolar coefficient in terms of the macroscopic electric field in the suspension, Eq. (3),

$$\frac{\langle p \rangle}{4\pi \varepsilon_0} = da^3 \frac{3\varepsilon_e}{\varepsilon_s + 2\varepsilon_e} E_e,$$

directly transforms Eq. (5) into

$$\varepsilon_s = \varepsilon_e (1 + 3\phi d)$$

without making any assumptions regarding the value of  $\phi$ .

### 3. Deduction based on the Dukhin–Shilov approach

In this deduction we consider two identical parallel plate condensers [10]. The first (lower index 1) is filled with the suspending medium  $\varepsilon_e$  and the second (lower index 2) with the suspension  $\varepsilon_s$ ; see Fig. 3. Both are connected in series so that, when a DC voltage is applied to the system, the free charge on

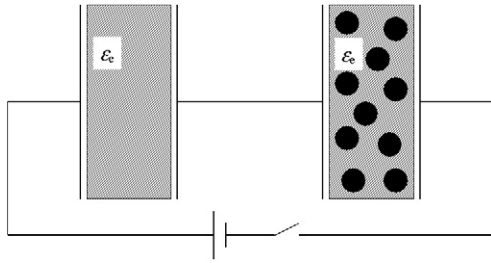


Fig. 3. Ideal parallel plane condensers filled with the suspending medium and with the suspension, used in the Dukhin–Shilov approach.

their plates has the same value. Therefore,

$$D_1 = \bar{D}_2, \tag{9}$$

where the upper segment has the meaning of a volume average. Using this expression together with

$$D_1 = \varepsilon_e E_1$$

and

$$\bar{D}_2 = \varepsilon_s \bar{E}_2$$

leads to the following expression for the permittivity of the suspension:

$$\varepsilon_e E_1 = \varepsilon_s \bar{E}_2. \tag{10}$$

Writing in this expression

$$\varepsilon_s = \varepsilon_e + \delta\varepsilon, \tag{11}$$

$$\bar{E}_2 = E_1 + \delta E \tag{12}$$

and neglecting the small addend containing the product  $\delta\varepsilon \delta E$  leads to

$$\delta\varepsilon = -\frac{\varepsilon_e \delta E}{E_1}. \tag{13}$$

The calculation of  $\delta E$  is performed by considering the electric potential at the two conducting plates of condenser 2, due to the presence of the particles in the suspension. The potential of a particle is

$$U_i(r, \theta) = \frac{p_{ix}}{4\pi\varepsilon_0} \frac{\cos\theta}{r^2}$$

or

$$U_i(x, y) = \frac{p_{ix}}{4\pi\varepsilon_0} \frac{x}{(x^2 + y^2)^{3/2}}.$$

In these expressions,  $p_{ix}$  is the component of the dipole moment of particle  $i$  in the direction of the applied field. The perpendicular component need not be considered because its contribution cancels out in the forthcoming calculation.

The average value of the potential of a particle, calculated over the area of a circle of radius  $R$  on a plate located a distance  $x_0$  from the particle, as shown in Fig. 4, is

$$\bar{U}_i(x_0) = \frac{1}{\pi R^2} \int_0^R U_i(x_0, y) 2\pi y dy$$

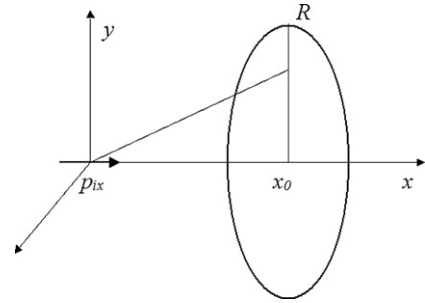


Fig. 4. Coordinate system used for the calculation of the average potential on one of the electrodes due to the dipole moment of a suspended particle.

$$\begin{aligned} &= \frac{p_{ix}}{4\pi\varepsilon_0} \frac{2x_0}{R^2} \int_0^R \frac{y dy}{(x_0^2 + y^2)^{3/2}} \\ &= \frac{p_{ix}}{4\pi\varepsilon_0} \frac{2x_0}{R^2} \left[ \frac{1}{|x_0|} - \frac{1}{\sqrt{x_0^2 + R^2}} \right], \end{aligned}$$

which, for  $R \gg x_0$ , becomes

$$\bar{U}_i(x_0) \rightarrow \frac{p_{ix}}{4\pi\varepsilon_0} \frac{2}{R^2}.$$

Clearly, the average potential calculated over the other plate has the same value but with a minus sign, so that the potential difference is two times larger. Moreover, since this value does not depend on the position of the particle in the condenser, the potential difference due to all the particles is a simple sum of their individual contributions. If the condenser has circular plates of radius  $R$  and spacing  $L$ , and contains  $N$  particles per unit volume, the potential difference  $\delta U$  due to all the particles is

$$\delta U = 2 \sum_{i=1}^{\pi R^2 LN} \left( \frac{p_{ix}}{4\pi\varepsilon_0} \frac{2}{R^2} \right) = 4\pi LN \frac{\langle p \rangle}{4\pi\varepsilon_0},$$

where

$$\langle p \rangle = \frac{1}{\pi R^2 LN} \sum_{i=1}^{\pi R^2 LN} p_{ix} = \frac{1}{\pi R^2 LN} \left| \sum_{i=1}^{\pi R^2 LN} p_i \right|,$$

since the sum of all the dipole moments has the direction of the applied field. Therefore

$$\delta E = -\frac{\delta U}{L} = -4\pi N \frac{\langle p \rangle}{4\pi\varepsilon_0}. \tag{14}$$

Combining this result with Eqs. (11) and (13) leads to the expression for the permittivity of the suspension:

$$\varepsilon_s = \varepsilon_e \left( 1 + 4\pi N \frac{\langle p \rangle}{4\pi\varepsilon_0 E_1} \right). \tag{15}$$

Relating the average value of the particle dipole moment to the dipolar coefficient,

$$\frac{\langle p \rangle}{4\pi\varepsilon_0} = da^3 E_1, \tag{16}$$

leads to the final result,

$$\varepsilon_s = \varepsilon_e (1 + 3\phi d), \tag{17}$$

where

$$\phi = N4\pi a^3/3 \quad (18)$$

is the volume fraction of particles in the suspension.

Note that in the deduction of Eq. (17), the assumption that the particle concentration is low was made twice: first, in writing Eq. (13) the addend containing  $\delta\varepsilon \delta E$  was neglected and, second, in writing Eq. (16) the dipole coefficient was expressed in terms of the field  $E_1$  rather than the macroscopic field in the suspension  $\bar{E}_2$ .

Avoiding the first of these assumptions, Eq. (13) becomes

$$\delta\varepsilon = -\frac{\varepsilon_e \delta E}{E_1 + \delta E} \quad (19)$$

so that Eq. (15) for the permittivity of the suspension transforms into

$$\varepsilon_s = \varepsilon_e \left( 1 + \frac{4\pi N \frac{\langle p \rangle}{4\pi\varepsilon_0 E_1}}{1 - 4\pi N \frac{\langle p \rangle}{4\pi\varepsilon_0 E_1}} \right). \quad (20)$$

Expressing the dipole coefficient in terms of  $\bar{E}_2$ , the macroscopic field in the suspension (condenser 2), rather than  $E_1$ , the macroscopic field in the suspending medium (condenser 1),

$$\frac{\langle p \rangle}{4\pi\varepsilon_0} = da^3 \bar{E}_2,$$

leads finally to

$$\varepsilon_s = \varepsilon_e(1 + 3\phi d)$$

without making any assumptions regarding the value of  $\phi$ .

#### 4. Deduction based on the Landau–Lifchitz formalism

We consider a heterogeneous dielectric system made of particles immersed in a homogeneous medium with permittivity  $\varepsilon_e$  and calculate the macroscopic permittivity of the whole system,  $\varepsilon_s$ , using as definition for this magnitude the seemingly most general expression given in Section 9 of Ref. [11]:

$$\bar{\mathbf{D}} = \varepsilon_s \bar{\mathbf{E}}. \quad (21)$$

In this expression

$$\bar{\mathbf{D}} = \frac{1}{V} \iiint_V \mathbf{D}(\mathbf{r}) dV, \quad \bar{\mathbf{E}} = \frac{1}{V} \iiint_V \mathbf{E}(\mathbf{r}) dV$$

are volume averages of the electric displacement and of the electric field,  $\mathbf{D}(\mathbf{r})$  and  $\mathbf{E}(\mathbf{r})$  are the local values of these fields, and  $\mathbf{r}$  is the radius vector of a generic point in the system. The integration is performed over the whole volume  $V$  of the system that is assumed to be large as compared to the characteristic size of the particles.

We consider that the system is macroscopically isotropic and that the electric field is applied in the direction of the  $x$  axis. Under these conditions, the vectors  $\bar{\mathbf{D}}$  and  $\bar{\mathbf{E}}$  will also be oriented along this same axis. The permittivity of the suspension thus becomes

$$\varepsilon_s = \frac{\bar{D}_x}{\bar{E}_x} = \frac{\frac{1}{V} \iiint_V D_x(\mathbf{r}) dV}{\frac{1}{V} \iiint_V E_x(\mathbf{r}) dV}. \quad (22)$$

Writing the  $x$  components of the two field vectors as  $D_x = \mathbf{D} \cdot \text{grad } x$  and  $E_x = \mathbf{E} \cdot \text{grad } x$ , and using the identity

$$\text{div}(\varphi \mathbf{A}) = \varphi \text{div } \mathbf{A} + \mathbf{A} \cdot \text{grad } \varphi,$$

leads to

$$D_x = \text{div}(x \mathbf{D}) - x \text{div } \mathbf{D}, \quad E_x = \text{div}(x \mathbf{E}) - x \text{div } \mathbf{E}. \quad (23)$$

We now use the Maxwell equations relating the electric displacement and electric field to the volume densities of free (ion) charge  $\rho_f$ , bound (polarization) charge  $\rho_b$ , and total charge

$$\rho = \rho_f + \rho_b:$$

$$\text{div } \mathbf{D} = \rho_f, \quad \text{div } \mathbf{E} = \frac{1}{\varepsilon_0} \rho. \quad (24)$$

Combining Eqs. (23) and (24), and taking into account that the considered problem is of the “dielectric inside a dielectric” type, so that there are no free charges ( $\rho_f = 0$ ), leads to

$$D_x = \text{div}(x \mathbf{D}), \quad E_x = \text{div}(x \mathbf{E}) - \frac{x}{\varepsilon_0} \rho. \quad (25)$$

We now substitute Eq. (25) for  $D_x(\mathbf{r})$  and  $E_x(\mathbf{r})$  in the integrals appearing in Eq. (22) and use the identity

$$\iiint_V \text{div } \mathbf{A} dV = \iint_S \mathbf{A} \cdot d\mathbf{S}.$$

We consider that the volume  $V$  corresponds to the whole sample so that, at all points of its surface  $S$ , the permittivity is that of the dispersion medium  $\varepsilon_e$  and, therefore,

$$\mathbf{D} = \varepsilon_e \mathbf{E}. \quad (26)$$

We thus obtain

$$\begin{aligned} \iiint_V D_x(\mathbf{r}) dV &= \iint_S (x \mathbf{D} \cdot d\mathbf{S}) = \varepsilon_e \iint_S (x \mathbf{E} \cdot d\mathbf{S}), \\ \iiint_V E_x(\mathbf{r}) dV &= \iint_S (x \mathbf{E} \cdot d\mathbf{S}) - \frac{1}{\varepsilon_0} \iiint_V \rho x dV. \end{aligned} \quad (27)$$

Using the general definition of the dipole moment of a system, the last integral in Eq. (27) represents the sum of the dipole moments (including the polarization charges of the surrounding medium) of all the particles present in the sample (or rather their components along the direction of the applied field):

$$\iiint_V \rho x dV = \sum_{i=1}^{NV} p_{ix}. \quad (28)$$

Equating the surface integrals on the right-hand side of the expressions (27),

$$\iiint_V D_x(\mathbf{r}) dV = \varepsilon_e \iiint_V E_x(\mathbf{r}) dV + \frac{\varepsilon_e}{\varepsilon_0} \sum_{i=1}^{NV} p_{ix}, \quad (29)$$

and combining with Eq. (22) leads, finally, to the general expression for the permittivity of the suspension:

$$\varepsilon_s = \varepsilon_e + \frac{\varepsilon_e}{\varepsilon_0} \frac{\frac{1}{V} \sum_{i=1}^{NV} p_{ix}}{\bar{E}_x}. \quad (30)$$

This result, deduced using the most general expression (21), is valid for any value of the particle concentration and for any particle shape, provided that their distribution and orientation is isotropic so that the macroscopic system is also isotropic. This requirement is the sole restriction on this deduction.

In the particular case of a suspension of monodispersed spherical particles,

$$\sum_{i=1}^{NV} p_{xi} = NV \langle p \rangle.$$

Expressing the average dipole moment in terms of the dipolar coefficient and the average electric field in the suspension,

$$\langle p \rangle = 4\pi \varepsilon_0 da^3 \bar{E}_x,$$

and using Eq. (18) leads to the final result,

$$\varepsilon_s = \varepsilon_e (1 + 3\phi d).$$

## 5. Generalization to conducting systems

We shall now generalize the considered deductions to conducting systems. In this most general case, the dielectric properties can be characterized by the complex permittivity, defined as

$$\varepsilon^*(\omega) = \varepsilon(\omega) - j \frac{\sigma(\omega)}{\omega}, \quad (31)$$

where the asterisk (\*) denotes a complex quantity,  $j = \sqrt{-1}$ , while  $\varepsilon(\omega)$  and  $\sigma(\omega)$  are the absolute permittivity and the conductivity (real functions of the frequency).

### 5.1. Generalization of the deduction based on the Maxwell–Wagner approach

In this case, the generalization is straightforward. The considered macroscopic spheres are the same as before, except that they are now characterized by the complex permittivities  $\varepsilon_e^*(\omega)$  and  $\varepsilon_s^*(\omega)$ , and an AC electric field  $E_e^*(\omega)$  is used. All the equations and arguments hold unaltered, except for the substitution of all the frequency independent quantities by their complex frequency dependent analogs.

### 5.2. Generalization of the deduction based on the Dukhin–Shilov approach

Again, the considered systems are the same as before, except that the two condensers are now characterized by the complex permittivities  $\varepsilon_e^*(\omega)$  and  $\varepsilon_s^*(\omega)$  and an AC voltage is used. Since the two condensers are connected in series, the current  $I^*(\omega)$  and the total voltage  $U^*(\omega)$  across the system are related by

$$U^* = I^* \left( \frac{1}{j\omega C_1^*} + \frac{1}{j\omega C_2^*} \right),$$

where

$$C_{1,2}^* = \frac{\varepsilon_{e,s}^* \pi R^2}{L}.$$

Therefore, the voltages across the individual condensers are

$$U_{1,2}^* = \frac{I^*}{j\omega C_{1,2}^*} = U^* \frac{C_{2,1}^*}{C_1^* + C_2^*}$$

so that the electric fields inside the condensers are

$$E_1^* = -\frac{U_1^*}{L} = -\frac{U^*}{L} \frac{\varepsilon_s^*}{\varepsilon_e^* + \varepsilon_s^*} \quad \text{and}$$

$$\bar{E}_2^* = -\frac{U_2^*}{L} = -\frac{U^*}{L} \frac{\varepsilon_e^*}{\varepsilon_e^* + \varepsilon_s^*}.$$

These expressions show that the electric displacements

$$D_1^* = \varepsilon_e^* E_1^* = -\frac{U^*}{L} \frac{\varepsilon_e^* \varepsilon_s^*}{\varepsilon_e^* + \varepsilon_s^*} \quad \text{and}$$

$$\bar{D}_2^* = \varepsilon_s^* \bar{E}_2^* = -\frac{U^*}{L} \frac{\varepsilon_s^* \varepsilon_e^*}{\varepsilon_e^* + \varepsilon_s^*}$$

are equal to one another, leading to the complex generalization of Eq. (9).

From here on, all the following equations and arguments hold unaltered, except for the replacement of all the frequency-independent quantities by their complex frequency-dependent analogs.

### 5.3. Generalization of the deduction based on the Landau–Lifchitz formalism

Unlike the preceding cases, this generalization is not immediate, since in the deduction of Eq. (25) it was specifically assumed that the considered system is non-conducting. However, when the system is conducting, the field-induced polarization of the double layer leads to the appearance of a free charge density near the surfaces of the suspended particles (at a distance of the order of the Debye screening length). This charge density  $\rho_f \neq 0$ , together with Eq. (24), shows that  $\text{div } \mathbf{D} \neq 0$  so that, in the complex formulation, there is no direct transition from Eq. (23) to Eq. (25). Therefore, it is not possible to simply replace the permittivities of the components by their complex counterparts.

In order to surmount this problem, we consider the expression for the full complex electric displacement  $\mathbf{D}^*$  as a sum of two terms: the first one,  $\mathbf{D}_b^*$ , reflecting the contribution due to dielectric polarization (displacement of *bound* charges) and the second one,  $\mathbf{J}_f^*$ , related to the ionic current density (movement of *free* charges):

$$\mathbf{D}^*(\omega, \mathbf{r}) = \mathbf{D}_b^*(\omega, \mathbf{r}) - j \frac{\mathbf{J}_f^*(\omega, \mathbf{r})}{\omega}. \quad (32)$$

We now consider the continuity equation for the free charge density,

$$\text{div } \mathbf{J}_f^* = -\frac{\partial \rho_f^*}{\partial t} = -j\omega \rho_f^*,$$

and the Poisson equation,

$$\text{div } \mathbf{D}_b^* = \rho_f^*.$$

Combining these expressions with the divergence of the complex electric displacement, Eq. (32), leads to the result

$$\operatorname{div} \mathbf{D}^* = \operatorname{div} \mathbf{D}_b^* - j \frac{\operatorname{div} \mathbf{J}_f^*}{\omega} = \rho_f^* - j \frac{-j\omega\rho_f^*}{\omega} = 0.$$

Therefore, despite the presence of the field-induced free charge density  $\rho_f^*$ , the divergence of the complex electric displacement, which includes the contribution of the ion current density, is equal to zero. This provides the path for the transition from the complex formulation of Eq. (23) to that of Eq. (25), which becomes

$$D_x^* = \operatorname{div}(x\mathbf{D}^*), \quad E_x^* = \operatorname{div}(xE^*) - \frac{1}{\varepsilon_0}\rho^*x.$$

From here on, all the arguments and equations leading from Eq. (21) to Eq. (30) are readily generalized, so that the complex analog of that equation becomes

$$\varepsilon_s^* = \varepsilon_e^* + \frac{\varepsilon_e^*}{\varepsilon_0} \frac{1}{V} \sum_{i=1}^{NV} P_{ix}^* \bar{E}_x. \quad (33)$$

It should be noted that, in general, the ionic current density  $\mathbf{J}_f^*(\omega, \mathbf{r})$  in Eq. (32) is not necessarily caused solely by the electric field. For example, in the charged liquid within the diffuse part of the electric double layer, the local electric current density may be also related to the hydrodynamic flow (convective electric current) and to ion diffusion (diffusion current). Therefore, the generalization of Eq. (26) is not valid at every point of the electrolyte solution inside the suspension. However, in the presented deduction, this equation is only used over the macroscopic surface surrounding the whole system where the electrolyte solution is electroneutral so that the generalization of Eq. (26) is valid. Therefore, the validity of Eq. (33) is not restricted to the case when this current density is only due to electromigration (as is the case in the Maxwell–Wagner model). On the contrary, it may include other fields, such as the diffusion current, which is responsible for the low-frequency or  $\alpha$  dispersion in colloidal suspensions.

## 6. Conclusion

We have shown that the fundamental expression (1) for the complex permittivity of a suspension is valid for any value of the volume fraction  $\phi$  of suspended particles. However, this assertion requires a generalization of the definition of the dipolar coefficient,

$$\frac{\langle p^* \rangle}{4\pi\varepsilon_0} = d^* a^3 E^*,$$

where  $\langle p^* \rangle$  is the ensemble average of the dipole moments of the particles in the suspension and  $E^*$  is the macroscopic electric field.

Because of this generalization, the present work does not solve the more than a hundred year old mixture formula problem [8,9,12–14]: the relationship between the dielectric properties of a suspension and either the dielectric properties of its components (particles and electrolyte solution) or the field-induced dipole moment of a single suspended particle.

However, the obtained result is far from trivial. It makes it possible, for example, to calculate the complex permittivity of a concentrated suspension using numerical results for the dipole moment of the suspended particles, or, inverting the problem, to determine the average dipole moment of the suspended particles in a concentrated suspension from experimental data on its dielectric properties.

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