Dipole interaction of the Quincke rotating particles

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(Received 25 November 2011; published 27 February 2012)

We study the behavior of particles having a finite electric permittivity and conductivity in a weakly conducting fluid under the action of the external electric field. We consider the case when the strength of the external electric field is above the threshold, and particles rotate due to the Quincke effect. We determine the magnitude of the dipole interaction of the Quincke rotating particles and the shift of frequency of the Quincke rotation caused by the dipole interaction between the particles. It is demonstrated that depending on the mutual orientation of the vectors of angular velocities of particles, vector-directed along the straight line between the centers of the particles and the external electric field strength vector, particles can attract or repel each other. In contrast to the case of nonrotating particles when the magnitude of the dipole interaction increases with the increase of the strength of the external electric field, the magnitude of the dipole interaction of the Quincke rotating particles either does not change or decreases with the increase of the strength of the external electric field depending on the strength of the external electric field and electrodynamic parameters of the particles.

DOI: 10.1103/PhysRevE.85.026608 PACS number(s): 41.20.−q, 77.22.−d

I. INTRODUCTION

In spite of the large number of investigations concerned with a behavior of weakly conducting fluid-particle suspensions subjected to an external electric field, this system continues to attract the attention of experimentalists and theoreticians working in the field. The main two reasons are the importance of fluid-particle suspensions subjected to an external electric field in various fields of physics and biophysics [1,2] and the variety of physical effects which are characteristic for these systems. Systematic investigation of these systems based on the unified approach started in the study by Melcher and Taylor [3] and was developed further in numerous investigations (see, e.g. [3–6]).

Special emphasis in the above mentioned theoretical and experimental studies was given to the case when particle electric conductivity σ_p and permittivity ε_p satisfy a condition

$$\varepsilon_p/\sigma_p > \varepsilon_{\mathrm{out}}/\sigma_{\mathrm{out}}, \tag{1}$$

where $\varepsilon_{\mathrm{out}}$ and σ_{out} are permittivity and electric conductivity of a host fluid, respectively. Condition (1) corresponds to the case when a characteristic relaxation time of the free charge inside the particle $\tau_p = \varepsilon_0\varepsilon_p/\sigma_p$ is larger than a characteristic relaxation time of the free charge in the host fluid, $\tau_{\mathrm{out}} = \varepsilon_0\varepsilon_{\mathrm{out}}/\sigma_{\mathrm{out}}$. Under these conditions and in the presence of a sufficiently strong external electric field particles rotate around their axes of symmetry. This effect of rotation (the Quincke effect) is associated with the dependence of the direction of torque acting at the particle in the external electric field by the relation between the characteristic times, τ_p and τ_{out}. Changing the inequality (1) to the opposite results in the reversal of the direction of the dipole moment of the induced free charge at the surface of the particle. In a case of the spherical particle the dipole moment, which is caused by the "instantaneous" polarization of the particle, is always directed along the direction of the external electric field and does not result in torque acting at the particle. However, the torque caused by a free charge can change sign and corresponds to the stable or unstable particle equilibrium. In the latter case particles can rotate with a constant angular velocity even in a stagnant host fluid. Herewith the electric charge which is carried away by a convective electric current caused by particle rotation is compensated by the conductivity current. Consequently, the effective distribution of electric charges at the particle surface does not change and corresponds to such distribution whereby the direction of the dipole moment of the particle does not coincide with the direction of the external electric field. Clearly particle rotation can be caused also by other forces. Therefore the inequality (1) determines the condition when the torque which is required for particle rotation is less than in the case without the external electric field. The nature of this effect is manifested in full capacity when the external rotating electric field is applied [6]. In this case, due to the finite relaxation time of the dipole moment, the direction of the dipole moment does not coincide with the direction of the external electric field, and therefore the torque is applied at the particle independent of whether the condition (1) is satisfied or not. The difference is that when the condition (1) is not satisfied particles rotate in the direction of rotation of the external electric field with the lower angular velocity. When angular velocity of rotation of the external electric field vanishes the angular velocity of particle rotation also vanishes. If condition (1) is satisfied the balance between a convective and a conductive electric current is altered so that stationary rotation occurs when particles rotate against the direction of rotation of the external electric field. Therewith when a velocity of rotation of the external electric field vanishes but the amplitude of the field exceeds some critical value, particles continue to rotate. It was suggested to call particles which meet the condition (1) negative electroviscosity particles (NEV particles) [6] since in the main rotation regime in the presence of the external rotating electric field these particles rotate in the opposite direction.

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Studies of the effect of Quincke rotation on the behavior of the system can be tentatively classified into two groups: (i) investigations of the dynamics of a single solid [7,8] or liquid [9] particle (body) and (ii) investigations of the changes of the properties of emulsions due to Quincke effect, e.g., modification of viscosity [10] or electric conductivity [11,12]. Although quite a few studies on the Quincke rotation were published in the past, some facets of this phenomenon remained unexplored. One of these facets is associated with interaction between the dipole moments of rotating particles. In the present study we consider a case when both particles are NEV particles. In Sec. II of this study we present a detailed derivation of the electrodynamic equations governing the behavior of particles in a dipole approximation. We derived equations which determine the strengths of the electric fields acting at the particles during their stationary rotation. In this section we also present general formulas which determine the force of particle interaction for an arbitrary orientation of the straight line connecting the centers of spherical particles with respect to the direction of the external electric field. In Sec. III using the derived formulas we investigate the dependence of the shift of the frequency of the Quincke rotation on the mutual orientation of particles with respect to the direction of the external electric field and directions of particle angular velocity vectors. In Sec. IV we investigate the dependence of the force of interaction of rotating particles vs mutual orientation of particles with respect to the direction of the external electric field and the directions of particle angular velocity vectors.

II. MATHEMATICAL MODEL

Let us consider a system of two spherical particles with parameters $\varepsilon_i,\sigma_i,a_i$, where a_i is a radius of the ith particle; ε_i,σ_i are the permittivity and the electric conductivity of the ith particle and $i = 1,2$. Assume that these particles are embedded in a host fluid with the permittivity ε_{out} and the electric conductivity σ_{out}, and the electric field $\vec{E} = E_0\vec{e}$ is applied to the whole system ($\vec{e}$ is a unit vector, see Fig. 1).The volumetric charge of the host fluid and the total charge of the particles are equal to zero. Consequently, there are no fields with the monopole symmetry in the system. Rotation of the particle subjected to the external electric field in the overdamped regime is governed by the following equation (see [13], Chap. II, Sec. 20):

$$\vec{\omega} = \frac{\vec{M}_e}{6\pi\eta V}, \tag{2}$$

where $\vec{\omega}$ is angular velocity of the particle, η is dynamic viscosity of the host fluid, V is particle volume, and $\vec{M}_e$ is torque acting at the particle due to the presence of the external electric field. In Eq. (2) the ordinal number of the particle, i, is dropped out and the hydrodynamic interaction between the particles is neglected. It is assumed that particles are located far away from the electrodes so that their interaction with the electrodes can be neglected.

In the framework of the piecewise continuous model whereby parameters of the system ε and σ change only at the interface between different media, the electric field in the volume occupied by a particle can be considered as a superposition of the continuous field due to the external sources and the field produced by the particle. The electric field produced by the particle has a finite jump at the particle surface due to the total surface charge. The formula for the total surface charge density reads (see, e.g., [14,15])

$$\gamma_T = \varepsilon_0\vec{n}\cdot[\vec{E}], \tag{3}$$

where $[E] = E_+ - E_-$, E_+ and E_- are the values of the function A at the external and internal surfaces, respectively, and $\vec{n}$ is the external unit normal vector at the particle surface.

The force and torque acting at the particle are determined by the following relations (for details see [16]):

$$\vec{F} = \int \gamma_T\vec{E}_c dA, \quad \vec{M}_e = \int \gamma_T\vec{r}(A)\times\vec{E}_c dA, \tag{4}$$

where $\vec{E}_c$ is the continuous-at-the-surface component of the electric field which will be determined further, symbols $\vec{r}(A)$ and dA denote that vector $\vec{r}$ is taken at the particle surface, and integration is performed over the surface of the particle.

The system of equations which determine the electrodynamic part of the problem reads (for details see [3])

$$\vec{\nabla}\cdot\vec{D} = \rho_{ex}, \tag{5}$$

$$\frac{\partial\rho_{ex}}{\partial t} + \vec{\nabla}\cdot\vec{j} = 0, \tag{6}$$

$$\begin{aligned} &\vec{D} = \varepsilon_0\varepsilon\vec{E}, \quad \vec{j} = \vec{j}_\sigma + \vec{j}_c, \quad \vec{j}_\sigma = \sigma\vec{E}, \\ &\vec{j}_c = \rho_{ex}\vec{v}_c, \quad \vec{E} = -\vec{\nabla}\Phi, \end{aligned} \tag{7}$$

where ρ_{ex} is the density of the free charge which is formed during the relaxation process that is governed by Eq. (6), ε and σ are the local values of permittivity and conductivity, $\vec{j}_c$ is the density of the convective current caused by the macroscopic motion of charges, $\vec{v}_c$ is the velocity of a macroscopic motion of electric charge, $\vec{v}_c(\vec{r}) = \dot{\vec{c}} + \vec{\omega}\times(\vec{r}-\vec{c})$, $\vec{c}$ is the location of the center of the spherical particle, $\dot{\vec{c}}$ is the velocity of translational motion of the particle, and $\vec{\omega}$ is the angular velocity.

Hereafter we consider an electrically neutral medium where free charges can be formed at the boundaries between different media. Consequently, the surface density of the free charge γ and the density of the free charge ρ_{ex} are related:

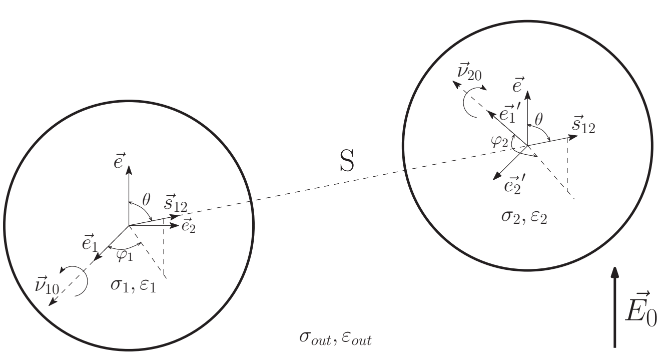

FIG. 1. Schematic view of the system of weakly conducting spherical particles subjected to the homogeneous external electric field.

$\int \rho_{ex} dV = \int \gamma dA$ or $\rho_{ex} = \gamma\delta(w)|\vec{\nabla} w|$, where $\delta(w)$ is the Dirac's delta function, $w = \Psi(x,y,z,t)$, and $w = 0$ is an equation of the particle surface. Equations (4) and (6) must be supplemented with the boundary conditions:

$$[\vec{n}\cdot\vec{D}] = \gamma, \quad [\vec{n}\cdot\vec{j}_\sigma] = -\frac{\partial\gamma}{\partial t} - \vec{v}_c\cdot\vec{\nabla}\gamma. \tag{8}$$

Equations (7) imply that for determining electric fields and currents it is sufficient to determine the electric potential, Φ. Due to the linearity of the problem the electric potential in the medium with the piecewise constant properties, which includes particles embedded in a host fluid, can be found by partitioning the space into the domains occupied by the particles and the external space. Hereafter we consider the problem in a dipole approximation whereby the electric potential in the vicinity of the ith particle can be written as follows:

$$\Phi_i(\vec{r}) = -\vec{E}_{ci}\cdot\vec{r}_i + \theta_i\vec{d}_i\cdot\vec{r}_i + (1-\theta_i)\,a_i^3\frac{\vec{d}_i\cdot\vec{r}_i}{|\vec{r}_i|^3}. \tag{9}$$

Here $\vec{E}_{ci}$ is a continuous component of the electric field which is the sum of the external electric field $\vec{E}_0$ and the field produced by other particles in the vicinity of the ith particle, $\vec{r}_i = \vec{r} - \vec{c}_i$, $\vec{d}_i$ is the dipole moment per unit volume of the particle, $\theta_i = 1$ inside the ith particle, and $\theta_i = 0$ outside the ith particle. Therefore the expression for the electric field $\vec{E}_{ci}$ can be written as follows:

$$\vec{E}_{ci} = \vec{E}_0 + \vec{E}_j(c_i), \quad \vec{E}_j(\vec{c}_i) = \vec{E}_j(\vec{r})|_{\vec{r}=\vec{c}_i},$$
$$\vec{E}_j(\vec{r}) = \frac{a_j^3}{|\vec{r}-\vec{c}_j|^3}\vec{d}_j\cdot\hat{T}_j(\vec{r}), \tag{10}$$

where matrix $\hat{T}_j(\vec{r}) = 3\vec{s}_j\vec{s}_j - \hat{e}$, $\vec{s}_j = (\vec{r}-\vec{c}_j)/|\vec{r}-\vec{c}_j|$, and $\hat{e}$ is a unit tensor. For a system of two particles, $i = 1,2$ and $j = 3 - i$.

Equations (5)–(10) yield the following set of equations for determining the free charge, γ_i:

$$\vec{d}_i\cdot\vec{n}(3+\kappa_{\varepsilon i}) = \kappa_{\varepsilon i}\vec{n}\cdot\vec{E}_{ci} + \gamma_i/\varepsilon_0\varepsilon_{\text{out}}, \tag{11}$$
$$\vec{d}_i\cdot\vec{n}(3+\kappa_{\sigma i}) = \kappa_{\sigma i}\vec{n}\cdot\vec{E}_{ci} - (\dot{\gamma}_i + \vec{v}_c\cdot\vec{\nabla}\gamma_i)/\sigma_{\text{out}}, \tag{12}$$

where $\kappa_{\varepsilon i} = \varepsilon_i/\varepsilon_{\text{out}} - 1$ and $\kappa_{\sigma i} = \sigma_i/\sigma_{\text{out}} - 1$. For further analysis it is convenient to introduce the dipole moment:

$$\vec{P} = \int \gamma_T(\vec{r}-\vec{c})\,dA, \tag{13}$$

where integration is performed over the particle's surface. Equations (3)–(9) imply the following expression for the total surface charge, γ_T:

$$\gamma_T = 3\varepsilon_0\vec{d}\cdot\vec{n}.$$

Derivation of Eqs. (11) and (12) from Eq. (8) using Eq. (9) and development of the latter expression for the total surface charge through the vector of the dipole moment $\vec{d}$ are outlined in Appendix. The latter formula for the total surface charge and Eq. (13) yield formulas for the torque and forces acting at the particle. Keeping only the first terms in the expansion of the electric field in the vicinity of $\vec{r} = \vec{c}_i$ which yield nonzero contribution to the torque and the force, we obtain the following formulas instead of Eqs. (4):

$$\vec{M}_i = \vec{P}_i\times\vec{E}_{ci}, \quad \vec{F}_{ij} = (\vec{P}_i\cdot\vec{\nabla})\vec{E}_j|_{\vec{r}=\vec{c}_i}. \tag{14}$$

Formulas (14) determine the torque and the force acting at the particle due to the external electric field and the electric field produced by polarization of the neighboring particle. Definitions of the surface density of the total external charge and of the dipole moment [Eqs. (3) and (13)] and formula (9) for the potential yield the following expression for the dipole moment:

$$\vec{P}_i = 3\varepsilon_0\vec{d}_i V_i.$$

The force applied by jth particle at the ith particle is a function of three vectors: $\vec{d}_i$, $\vec{d}_j$, and $\vec{s}_{ij} = (\vec{c}_i - \vec{c}_j)/|\vec{c}_i - \vec{c}_j|$. Using these vectors allows us to rewrite Eqs. (14) as follows:

$$\vec{F}_{ij} = \frac{9\varepsilon_0 V_i a_j^3}{|\vec{c}_i-\vec{c}_j|^4}[\vec{d}_i(\vec{d}_j\cdot\vec{s}_{ij}) + \vec{d}_j(\vec{d}_i\cdot\vec{s}_{ij}) + \vec{s}_{ij}(\vec{d}_i\cdot\vec{d}_j)$$
$$- 5(\vec{d}_i\cdot\vec{s}_{ij})(\vec{d}_j\cdot\vec{s}_{ij})\vec{s}_{ij}], \tag{15}$$

$$\vec{M}_i = 3\varepsilon_0 V_i\vec{d}_i\times\vec{E}_{ci}. \tag{16}$$

Equation (15) implies that $\vec{F}_{ij} = -\vec{F}_{ji}$.

It is convenient to rewrite Eqs. (11) and (12) introducing the dipole moments, $\vec{d}_\varepsilon$ and $\vec{d}_\sigma$, such that $\vec{d}_i = \vec{d}_\varepsilon + \vec{d}_\sigma$:

$$\vec{d}_{\varepsilon i} = \frac{\kappa_{\varepsilon i}\vec{E}_{ci}}{3+\kappa_{\varepsilon i}}, \quad \vec{d}_{\sigma i}\cdot\vec{n} = \frac{\gamma_i}{\varepsilon_0\varepsilon_{\text{out}}(3+\kappa_{\varepsilon i})}. \tag{17}$$

Using Eqs. (11), (12), and (17) and taking into account that $\dot{\gamma}_i + \dot{\vec{c}}_i\cdot\vec{\nabla}\gamma_i = 0$ we arrive at the following equation:

$$\frac{\partial\vec{d}_{\sigma i}}{\partial t} - \vec{\omega}_i\times\vec{d}_{\sigma i} + \frac{\vec{d}_{\sigma i}}{\tau_i} = \frac{(\kappa_{\sigma i}-\kappa_{\varepsilon i})\vec{E}_{ci}}{\tau_i(1+\kappa_{\sigma i}/3)(\kappa_{\varepsilon i}+3)}, \tag{18}$$

where $\tau_i = \tau_0(3+\kappa_{\varepsilon i})/(3+\kappa_{\sigma i})$ and $\tau_0 = \varepsilon_0\varepsilon_{\text{out}}/\sigma_{\text{out}}$ and $\vec{\omega}$ is the vector of particle angular velocity.

Consider a stationary regime. In this case angular velocity $\vec{\omega}$ is aligned with the torque Eq. (16), $\vec{\omega}$ is normal to the dipole moment of the particle and the direction of the angular velocity does not change. Solution of Eq. (18) in this case reads

$$\vec{d}_{\sigma i} = \frac{\beta_i}{(1+\nu_i^2)(\kappa_{\varepsilon i}+3)}(\vec{E}_{ci} + \vec{\nu}_i\times\vec{E}_{ci}),$$
$$\beta_i = \frac{\kappa_{\sigma i}-\kappa_{\varepsilon i}}{1+\kappa_{\sigma i}/3}, \quad \vec{\nu}_i = \vec{\omega}_i\tau_i. \tag{19}$$

Taking into account the definitions of $\vec{d}_\varepsilon$ and $\vec{d}_\sigma$ we arrive at the following equation for $\vec{d}_i = \vec{d}_{\varepsilon i} + \vec{d}_{\sigma i}$:

$$\vec{d}_i = \frac{1}{\kappa_{\varepsilon i}+3}\left[\left(\kappa_{\varepsilon i} + \frac{\beta_i}{1+\nu_i^2}\right)\vec{E}_{ci} + \beta_i\frac{\vec{\nu}_i\times\vec{E}_{ci}}{1+\nu_i^2}\right]. \tag{20}$$

In order to simplify Eq. (20) let us introduce the following notations:

$$\xi_i = \frac{1}{\kappa_{\varepsilon i}+3}\left(\kappa_{\varepsilon i} + \frac{\beta_i}{1+\nu_i^2}\right),$$

and the antisymmetric matrix

$$\hat{\Omega}_{k\ell} = -\hat{\Omega}_{\ell k} = \frac{\beta_i}{(\kappa_{\varepsilon i}+3)\left(1+\nu_i^2\right)}\varepsilon_{km\ell}\,(\vec{\nu}_i)_m\,,$$

where $\varepsilon_{km\ell}$ is the Levi-Civita symbol.

Using these notations Eq. (20) can be written as follows:

$$\vec{d}_i = (\xi_i\hat{e} + \hat{\Omega}_i)\cdot\vec{E}_{ci}\,.$$

Using the definition of the field $\vec{E}_{ci}$ [Eq. (10)] we obtain a relation between the fields $\vec{E}_{c1}$ and $\vec{E}_{c2}$:

$$\vec{E}_{c1} = \vec{E}_0 + \lambda_2\hat{T}(\xi_2\hat{e}+\hat{\Omega}_2)\vec{E}_{c2},\quad \lambda_2 = \frac{a_2^3}{S^3},$$
$$\hat{T} = \hat{T}_j(\vec{c}_i),\quad S = |\vec{c}_1-\vec{c}_2|. \tag{21}$$

Using a similar relation which expresses the strength of the electric field $\vec{E}_{c2}$ through the field strength $\vec{E}_{c1}$, we can exclude $\vec{E}_{c2}$ and arrive at the following equation:

$$(\hat{e}-\hat{R}_2\cdot\hat{R}_1)\vec{E}_{c1} = \hat{R}_2\cdot\vec{E}_0+\vec{E}_0, \tag{22}$$

where $\hat{R}_i = \lambda_i\hat{T}\cdot\hat{L}_i$ and $\hat{L}_i = \xi_i\hat{e}+\hat{\Omega}_i$. If the inverse matrix to the matrix $\hat{e}-\hat{R}_2\cdot\hat{R}_1$ exists, then Eq. (22) determines the electric field $\vec{E}_{c1}$ in the leading dipole approximation. The particular case of Eq. (22), when the particles do not rotate and are located at the straight line which is aligned in the direction of the external electric field, was considered in Ref. [16]. The interchange of indices in Eq. (22) yields an equation for determining the electric field $\vec{E}_{c2}$.

Taking into account Eqs. (19) and (20) we can rewrite formulas (2) and (14) for the torques $\vec{M}_i$ as follows:

$$1+\nu_i^2 = \frac{E_{ci}^2}{E_{*i}^2},\quad E_{*i}^2 = \frac{2\eta(\kappa_{\sigma i}+3)^2}{3\varepsilon_0\tau_0(\kappa_{\varepsilon i}-\kappa_{\sigma i})}. \tag{23}$$

Equations (23) for $i=1,2$ together with Eq. (20) for the electric fields $\vec{E}_{c1}$ and $\vec{E}_{c2}$ provide a complete set of equations for determining the vectors of the particle angular velocities. It must be noted that although according to Eqs. (22) and (23) the fields $\vec{E}_{c1}$ and $\vec{E}_{c2}$ appear in the equations separately, the angular velocity of one dipole depends on the angular velocity of the other dipole due to the structure of the matrix $\hat{R}_1$. Therefore, the electric fields $\vec{E}_{c1}$ and $\vec{E}_{c2}$ are interdependent. Equation (21) implies that for particle rotation the conditions given by Eq. (1) must be satisfied. If this condition is not met then the particle cannot rotate with a constant angular velocity, even when the other particle experiences the Quincke rotation. The latter assertion is associated with the fact that the dipole moments of the rotating particles remain constant, and, consequently, the magnitudes of the electric fields produced by the particles remain constant. In a case of a single particle, $\vec{E}_{ci}=\vec{E}_0$, and Eq. (23) determines the dependence of the angular velocity of the Quincke rotation of a single particle vs the amplitude of the external electric field $\vec{E}_{ci}=\vec{E}_0$. Note that the magnitude of the electric field scale, $\vec{E}_*$, is determined by the value of the parameter $\eta/(\varepsilon_0\tau_0)$. For $\tau_0\sim 1s$ and $\eta\sim 1\,\mathrm{kg/(m\cdot s)}$, $E_*\sim 3\times10^6$ V/m.

III. SHIFT OF THE ANGULAR VELOCITY OF INTERACTING PARTICLES

Let us determine the shift of the angular velocity of the particles caused by their dipole interaction. We consider a problem in the first nonvanishing approximation with respect to the parameters $\lambda_1 = a_1^3/S^3$ and $\lambda_2 = a_2^3/S^3$. In this case the magnitude of the electric fields $\vec{E}_{ci}$ is given by the following relation:

$$\vec{E}_{ci} = \hat{R}_{3-i}\cdot\vec{E}_0+\vec{E}_0. \tag{24}$$

Substituting Eq. (24) in Eq. (23) and keeping only the first nonvanishing terms with respect to the parameter λ_{3-i} we arrive at the system of equations which determine the frequencies of the particle rotation:

$$1+\nu_i^2 = \frac{E_0^2}{E_{*i}^2}[1+2\lambda_{3-i}\xi_{3-i}(3\mu^2-1)$$
$$+6\lambda_{3-i}\mu(\vec{V}_{3-i}\cdot\vec{\nu}_{3-i})],\quad i=1,2, \tag{25}$$

where $\mu=\vec{s}\cdot\vec{e}$, $\vec{e}$ is a unit vector in the direction of the external electric field $\vec{E}_0$, $\vec{V}_{3-i}=\vec{e}_p[\xi_{3-i}-\kappa_{\varepsilon 3-i}/(\kappa_{\varepsilon 3-i}+3)]$, $\vec{e}_p = \vec{e}\times\vec{s}$, and the vector $\vec{s}$ is either the vector $\vec{s}_{ij}$ or the vector $\vec{s}_{ji}=-\vec{s}_{ij}$ since only even powers of $\vec{s}_{ij}$ appear in Eq. (24). Since the system of equations (25) is obtained in the first nonvanishing approximation with respect to the parameter λ_{3-i}, in the formulas for the rotation frequencies one can use the value of ν_{3-i} obtained in the zeroth approximation with respect to the parameter λ_{3-i}:

$$\nu_{3-i} = \nu_{3-i0} = \sqrt{\frac{E_0^2}{E_{*3-i}^2}-1}. \tag{26}$$

The vector of the rotation frequency $\vec{\nu}_{3-i}$ is perpendicular to the direction of the external electric field $\vec{e}$. In many systems used for experimental investigation of Quincke rotation (see, e.g., [8,10,12]) the value of the parameter κ_ε, which is determined by the difference between the real parts of permittivities of the particle and the host medium, is much less than the magnitude of the parameter κ_σ, which is determined by the difference between electric conductivities of the particle and the host medium. In view of the latter remark hereafter we neglect κ_ε in order to considerably simplify derivations, assume that $\kappa_\varepsilon\ll\kappa_\sigma,\kappa_\varepsilon\ll 3$, and set $\kappa_\varepsilon=0$.

Using Eqs. (25) we arrive at the following formula for the difference $\nu_i^2-\nu_{i0}^2$, where $1+\nu_{i0}^2 = E_0^2/E_{*i}^2$:

$$\nu_i^2-\nu_{i0}^2 = \Delta_0\left(\mu^2-\frac{1}{3}+\mu\chi\nu_{3-i0}\right),$$
$$\Delta_0 = 6\lambda_{3-i}\frac{E_0^2\kappa_{\sigma i}\,(\kappa_{\sigma 3-i}+3)}{E_{*i}^2\,(\kappa_{\sigma i}+3)^2}, \tag{27}$$
$$\chi = \vec{s}\cdot(\vec{e}_{\nu 3-i}\times\vec{e})\,,$$

where vector $\vec{e}_{\nu 3-i}$ is a unit vector in the direction of the rotation frequency vector, $\vec{\nu}_{3-i0}$.

Equation (27) implies that the signs of the frequency shift are opposite in the cases when $\mu=\pm1$ and $\mu=0$. Since the Quincke rotation of the ith particles occurs only when $\kappa_{\sigma i}<0$, in the case when the straight line connecting the particle centers is normal to the external electric field, the frequency shift is positive and in the limit $\kappa_{\sigma i}\approx\kappa_{\sigma 3-i}\approx-1$,

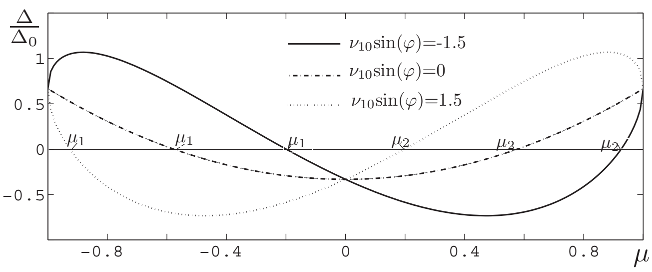

FIG. 2. Dependence of the shift of rotation frequency of the Quincke rotating particle vs the magnitude and orientation of angular velocity of the neighboring particle.

this shift is of the order of $\nu_i^2 - \nu_{i0}^2 \approx a_{3-i}^3 E_0^2/(S^3 E_*^2)$. When the straight line connecting the centers of particles having the same parameters is parallel to the external electric field, the frequency shift is negative, $\nu_i^2 - \nu_{i0}^2 \approx -3a_{3-i}^3 E_0^2/(S^3 E_*^2)$. For the arbitrary values of the parameter μ and given electric conductivities of particles and of the host fluid, the frequency shift of the ith particle is determined by scalar products, $\vec{s}\cdot\vec{e}$ and $\vec{s}\cdot\vec{e}_{\nu 3-i}$, i.e., by the projections of the unit vector $\vec{s}$ on the direction vector of the external electric field $\vec{e}$ and on the direction vector of the angular velocity $\vec{e}_{\nu 3-i}$. For the analysis of the behavior of the frequency shift $\Delta = \nu_i^2 - \nu_{i0}^2$ which is determined by Eq. (27) it is convenient to expand the unit vector $\vec{s}$ into the three unit vectors, $\vec{e}$, $\vec{e}_{\nu 3-i}$, $\vec{e}_{\nu 3-i}\times\vec{e}$:

$$\vec{s} = \mu\vec{e} + \sqrt{1-\mu^2}(\cos\varphi_{3-i}\vec{e}_{\nu 3-i} - \sin\varphi_{3-i}\vec{e}_{\nu 3-i}\times\vec{e}), \quad (28)$$

where φ_{3-i} is the azimuthal angle of the vector $\vec{s}$ in the counterclockwise direction. The schematic view of the system is shown in Fig. 1. The parameter μ in the above formulas is related with the angle θ in this figure, $\mu = \cos\theta$. Let us set $i = 2$ so that Eq. (27) determines the dependence of the shift of the rotation frequency of the second particle vs the direction and the magnitude of the unperturbed angular velocity of the first particle, ν_{10}. Expansion (28) allows us to rewrite Eq. (27) as follows:

$$\Delta = \Delta_0 U(\mu,\varphi),$$
$$U(\mu,\varphi) = \mu^2 - \tfrac{1}{3} + \sin(\varphi)\nu_{10}\mu\sqrt{1-\mu^2}.$$

Equation $U(\mu,\varphi) = 0$ has two roots, $\mu_1(\varphi)$ and $\mu_2(\varphi)$, where $\mu_1(\varphi) < \mu_2(\varphi)$. In the domain $k_1 = \nu_{10}\sin(\varphi) > 0$, $\mu_1 = -\sqrt{b+\sqrt{b^2-c}}$, $\mu_2 = \sqrt{b-\sqrt{b^2-c}}$, where $b = (2+3k_1^2)/6(1+k_1^2)$ and $c = 1/9(1+k_1^2)$. In the domain $k_1 < 0$, $\mu_1 = -\sqrt{b-\sqrt{b^2-c}}$, $\mu_2 = \sqrt{b+\sqrt{b^2-c}}$. The behavior of the function $U(\mu,\varphi)$ is showed in Fig. 2. Inspection of this figure shows that $U > 0$ in the range $\mu < \mu_1$ and $\mu > \mu_2$. When $\kappa_\varepsilon = 0$, the condition (1) implies that $\kappa_{\sigma i} < 0$. Since $\kappa_{\sigma i} > -1$, the range of the angles where $U > 0$ corresponds to the negative shift of the oscillation frequency, $\Delta < 0$.

IV. DIPOLE INTERACTION OF QUINCKE ROTATING PARTICLES

In this section we analyze the behavior of the force of the dipole interaction between two particles in the whole range of the external electric field. Hereafter we assume that for two particles, $i = 1,2$, the following condition is satisfied: $E_{*1} < E_{*2}$. We consider the behavior of the force of the dipole interaction in three ranges of the external electric field: (i) $E_0 < E_{*1} < E_{*2}$; (ii) $E_{*1} < E_0 < E_{*2}$; (iii) $E_{*1} < E_{*2} < E_0$. In the first range the external field does not cause particle rotation. In the second range only particle 1 participates in the Quincke rotation while in the third range both particles rotate. As we will see from further analysis in each of these domains there is a particular qualitatively different dependence of the interaction forces vs the amplitude of the external electric field and parameters of the problem. Clearly, at the boundaries between these domains these dependencies coincide.

Calculation of the interaction force is based upon Eq. (15). In the first nonvanishing approximation with respect to the parameters λ_i the dipole moment of the particle is the dipole moment that is induced by the external electric field, $\vec{E}_0$. The same approximation has been used earlier in Sec. III.

Under these conditions let us consider the magnitude of dipole interaction in the range $E_0 < E_{*1} < E_{*2}$. Setting $\nu_i = 0$ in Eq. (20) we find that $\vec{d}_i = \vec{E}_0\kappa_{\sigma i}/(3+\kappa_{\sigma i})$. Substituting the latter relation in Eq. (15) yields

$$\vec{F} = F_0[(1-5\mu^2)\vec{s}_{12} + 2\mu\vec{e}], \quad (29)$$

where $\mu = \vec{e}\cdot\vec{s}_{12}$ and the amplitude of the force F_0, which is independent of the characteristic directions in the problem, is determined by the following formula:

$$F_0 = \frac{12\pi\varepsilon_0 a_1^3 a_2^3 \alpha E_0^2}{S^4}, \quad \alpha = \frac{\kappa_{\sigma 1}\kappa_{\sigma 2}}{(\kappa_{\sigma 1}+3)(\kappa_{\sigma 2}+3)}. \quad (30)$$

Equation (29) determines the force acting at particle 1 by particle 2. The force applied by particle 1 at particle 2 is determined by Eq. (29) where $\vec{s}_{12}$ is replaced by $\vec{s}_{21} = (\vec{c}_2 - \vec{c}_1)/S$. The formula for the component of the force along the line connecting the centers of the particles reads

$$F_s = \vec{F}\cdot\vec{s}_{12} = F_0(1-3\mu^2). \quad (31)$$

This component changes its sign at $\mu^2 = 1/3$. When $\mu^2 > 1/3$ and $\alpha > 0$ particles attract each other. In the case when the line connecting the centers of the particles is parallel to the direction of the external electric field, $\mu = \pm 1$, Eqs. (29) and (30) recover formula (23) in Ref. [16]. Equation (29) determines the force of particle interaction in the stationary regime when as the result of recharging of the system the surface density of the free charges at the boundaries between different media attains its saturation. In order to determine the force of particle interaction at the initial moment when the surface density of the free charges at the interfaces is small, it is sufficient to replace $\kappa_{\sigma i}$ in Eq. (30) by $\kappa_{\varepsilon i}$. Therefore, without rotation the particle interaction force for small time scales is determined by the instantaneous polarizability of particles while at large time scales it is determined by their conductivities.

The situation is different in the case with the Quincke rotation. In the stationary regime, which is attained at large time scales, Eq. (20) implies that the dipole moment of the particle depends not only on its conductivity but also on the polarizability. Hereafter, in the analysis of particle interaction when at least one of the particles rotates, we neglect the contribution of polarizability to the interaction and assume that

$\kappa_\varepsilon = 0$. The conditions for the validity of this approximation can be easily obtained from Eq. (20) and are not presented here.

In the range of the external electric field $E_{*1} < E_0 < E_{*2}$, Eq. (20) yields:

$$\vec{d}_1 = \frac{\kappa_{\sigma 1}(\vec{E}_0 + \vec{\nu}_{10} \times \vec{E}_0)}{(\kappa_{\sigma 1} + 3)\left(1 + \nu_{10}^2\right)}, \quad \vec{d}_2 = \frac{\kappa_{\sigma 2}}{3 + \kappa_{\sigma 2}}\vec{E}_0,$$
$$\vec{\nu}_{10} = \nu_{10}\vec{e}_1, \tag{32}$$

where the absolute value of the rotation frequency ν_{10} is determined by Eq. (26) and the unit vector $\vec{e}_1$ is perpendicular to the direction of the external electric field $\vec{E}_0$.

Equations (15) and (32) yield the expression for the force of interaction between the particles:

$$\vec{F} = F_0\{[1 - 5\mu(\mu - \nu_{10}\sin\varphi_1\sqrt{1-\mu^2})]\vec{s}_{12} + (2\mu - \nu_{10}\sin\varphi_1\sqrt{1-\mu^2})\vec{e} - \mu\nu_{10}\vec{e}_2\}, \tag{33}$$

where $F_0 = 12\pi\varepsilon_0 a_1^3 a_2^3 \alpha E_{*1}^2/S^4$, $\vec{e}_2 = \vec{e} \times \vec{e}_1$, and the azimuthal angle, φ_1, is the angle between the vector $\vec{e}$ and the reference vector $\vec{e}_1$ in the counterclockwise direction (see Fig. 1). It must be noted that in the range of the amplitude of the external electric field $E_{*1} < E_0 < E_{*2}$, the coefficient F_0 does not depend on the amplitude of the external field. The component of the force acting at the particle 1 in the direction of the straight line connecting particle centers is given by the following formula:

$$F_s = \vec{F} \cdot \vec{s}_{12} = F_0(1 - 3\mu^2 + 3\nu_{10}\sin\varphi_1\mu\sqrt{1-\mu^2}). \tag{34}$$

Equation (34) implies that if the straight line connecting particle centers is perpendicular to the external field ($\mu = 0$) or parallel to the external field ($\mu = \pm 1$), the particle interaction force in this range of the external field amplitudes remains independent of the amplitude. The formula for the force Eq. (34) can be written as follows: $F_s = 3F_0U(\mu,\varphi)$, where function $U(\mu,\varphi)$ also determines the frequency shift Δ/Δ_0 (see the above analysis) and is shown in Fig. 2.

In order to determine the particle interaction force $\vec{F}$ in the range of the external electric field amplitude $E_{*1} < E_{*2} < E_0$ let us introduce the unit vectors $\vec{e}_1'$ and $\vec{e}_2'$. The unit vector $\vec{e}_1'$ is directed along the angular velocity vector of particle 2, $\vec{\nu}_{20}$, while the unit vector $\vec{e}_2' = \vec{e} \times \vec{e}_1'$. The unit vectors $\vec{e}_1'$ and $\vec{e}_2'$ are located in the plane that is parallel to the plane spanned by the vectors $\vec{e}_1$ and $\vec{e}_2$. Vectors $\vec{e}_1'$ and $\vec{e}_2'$ are related with the vectors $\vec{e}_1$ and $\vec{e}_2$ through rotation by the angle φ_0, where $\tan\varphi_0 = (\vec{e}_1' \times \vec{e}_1)\cdot\vec{e}/(\vec{e}_1' \cdot \vec{e}_1)$.

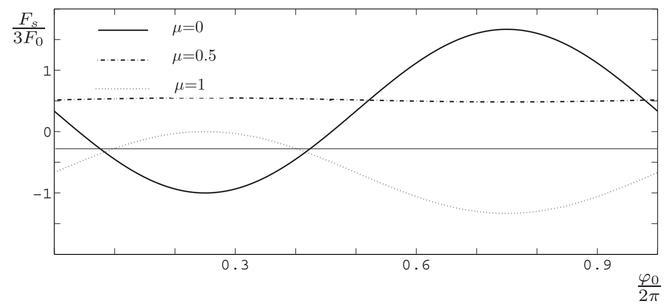

FIG. 4. Dependence of the normalized component of the interaction force of the Quincke rotating particles in the direction of the straight line connecting particle centers, $F_s/3F_0$, vs $\varphi_0 = \varphi_2 - \varphi_1$ for $\varphi_2 = \pi/2$.

The dipole moments $\vec{d}_1$ and $\vec{d}_2$ are determined by the first formula in Eqs. (32) with the corresponding indices. Using the introduced notations the formula for the particle interaction force $\vec{F}$ can be written as follows:

$$\vec{F} = F_0(f_s\vec{s}_{12} + f_2\vec{e}_2 + f_2'\vec{e}_2' + f\vec{e}), \tag{35}$$

where

$$F_0 = \frac{12\pi\varepsilon_0 a_1^3 a_2^3 \alpha E_{*1}^2 E_{*2}^2}{S^4 E_0^2}, \tag{36}$$

$$f_s = 1 + \cos\varphi_0\nu_{10}\nu_{20} - 5[\mu^2 - \mu\sqrt{1-\mu^2}(\nu_{10}\sin\varphi_1 + \nu_{20}\sin\varphi_2) + (1-\mu^2)\nu_{10}\nu_{20}\sin\varphi_1\sin\varphi_2], \tag{37}$$

$$f_2 = -\nu_{10}(\mu - \sqrt{1-\mu^2}\nu_{20}\sin\varphi_2),$$
$$f_2' = -\nu_{20}(\mu - \sqrt{1-\mu^2}\nu_{10}\sin\varphi_1), \tag{38}$$
$$f = 2\mu - \sqrt{1-\mu^2}(\nu_{10}\sin\varphi_1 + \nu_{20}\sin\varphi_2).$$

In Eqs. (37) and (38) $\varphi_2 = \varphi_0 + \varphi_1$. The component of the interaction force between the particles in the direction of the straight line connecting particle centers is given by the following formula:

$$F_s = \vec{F}\cdot\vec{s}_{12} = 3F_0[(k_1k_2 - 1)\mu^2 + (k_1 + k_2)\mu\sqrt{1-\mu^2} + (1 + k_0)/3 - k_1k_2], \tag{39}$$

where $k_0 = \nu_{10}\nu_{20}\cos\varphi_0, k_1 = \nu_{10}\sin\varphi_1, k_2 = \nu_{20}\sin\varphi_2$.

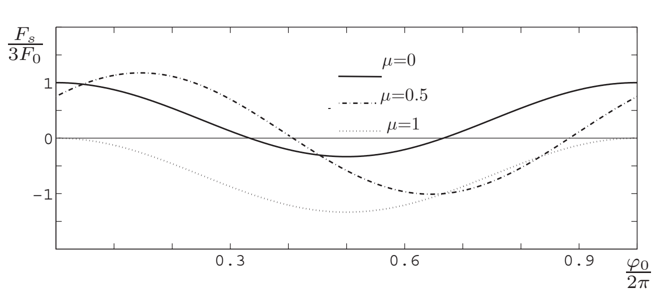

FIG. 3. Dependence of the normalized component of the interaction force of the Quincke rotating particles in the direction of the straight line connecting particle centers, $F_s/3F_0$, vs $\varphi_0 = \varphi_2 - \varphi_1$ for $\varphi_2 = 0$.

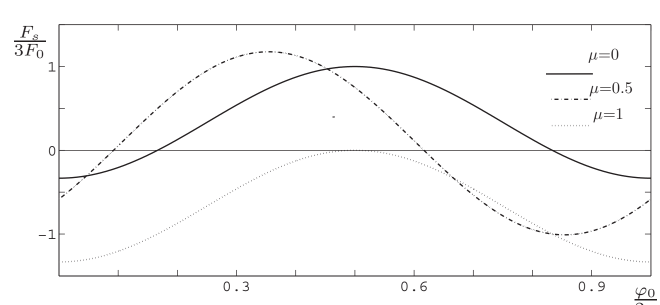

FIG. 5. Dependence of the normalized component of the interaction force of the Quincke rotating particles in the direction of the straight line connecting particle centers, $F_s/3F_0$, vs $\varphi_0 = \varphi_2 - \varphi_1$ for $\varphi_2 = \pi$.

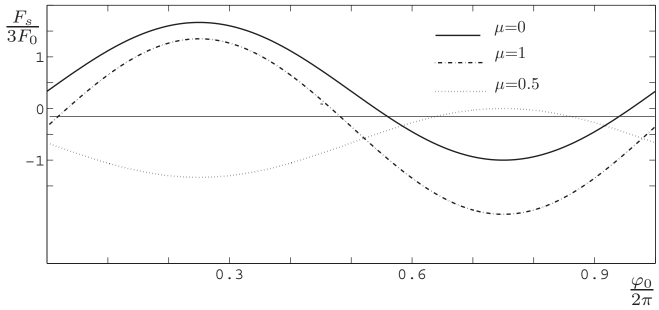

FIG. 6. Dependence of the normalized component of the interaction force of the Quincke rotating particles in the direction of the straight line connecting particle centers, $F_s/3F_0$, vs $\varphi_0 = \varphi_2 - \varphi_1$ for $\varphi_2 = 3\pi/2$.

Formula (39) implies that the interaction force in the direction of the straight line connecting particle centers can be written as $F_s = 3F_0 U_1(\mu,\varphi_1,\varphi_2)$, where the function $U_1(\mu,\varphi_1,\varphi_2)$ determines the angular distribution of the attraction and repulsion forces. Apart from the parameter k_1 which determines the behavior of the function $U(\mu,\varphi)$, $U_1(\mu,\varphi_1,\varphi_2)$ depends also upon the parameters k_0 and k_2. When $\nu_{20} \to 0$, $U_1(\mu,\varphi_1,\varphi_2) \to U(\mu,\varphi)$ and F_0, which is determined by Eq. (36), tends to F_0 which is given by Eq. (33). Equation (36) also implies that in the range of external field amplitude $E_{*1} < E_{*2} < E_0$, the magnitude of F_0 decreases when the amplitude of the external field grows. Although the algebraic structure of the function $U_1(\mu,\varphi_1,\varphi_2)$ is similar to the structure of function $U(\mu,\varphi)$, a large number of independent parameters results in the variety of behaviors of this function and complicates their analyses. In Figs. 3–6 we showed the behavior of the function $U_1(\mu,\varphi_1,\varphi_2)$ vs the difference of the azimuthal angles, $\varphi_0 = \varphi_2 - \varphi_1$, for $\varphi_2 = 0,\pi/2,\pi,3\pi/2$. Inspection of these figures reveals that the dependence of the particle interaction force vs the difference of the azimuthal angles, φ_0, depends also by the angle between the straight line connecting particle centers and the direction of the external electric field. Particle interaction force depends also upon the azimuthal angles φ_1 and φ_2, which determine the orientation of the straight line connecting particle centers with respect to the particle angular velocity vectors. It must be noted that although Eqs. (37) are invariant with respect to the substitution $k_1 \leftrightarrow k_2$, Eqs. (37) are not invariant with respect to the substitution $\varphi_1 \leftrightarrow \varphi_2$ since $\nu_{10} \neq \nu_{20}$.

V. CONCLUSIONS

We considered interaction of the Quincke rotating NEV particles when their parameters satisfy the condition (1), in the whole range of the external electric fields. It was demonstrated that depending on the mutual orientation of the particle angular velocity vectors, the direction of the external electric field and the direction of the straight line connecting particle centers, the magnitude and the sign of the particle interaction force change. We showed that in contrast to the case of nonrotating particles where the amplitude of the force, F_0, grows with the amplitude of the external field Eq. (30), in the case of the Quincke rotating particles, depending on the range of the external electric field strength, the force factor F_0 remains constant Eq. (33) or decreases when the amplitude of the external electric field grows Eq. (36). We investigated also the shift of particle rotation frequency as a function of the angular velocity vector of the rotating neighboring particle. Depending on the geometry of the problem which is determined by four vectors—(i) the vector of the direction of the external electric field, (ii) the vector of the direction of the straight line connecting the particle centers, (iii) the angular velocity vectors—the angular velocity shift can be either positive or negative.

APPENDIX: DERIVATION OF EQS. (11), (12), AND EXPRESSION FOR THE TOTAL SURFACE CHARGE

Equation (9) implies the following formula for the potential in the vicinity outside the ith sphere:

$$\Phi_{\mathrm{out},i}(\vec{r}) = -\vec{E}_{ci}\cdot\vec{r}_i + a_i^3\frac{\vec{d}_i\cdot\vec{r}_i}{|\vec{r}_i|^3}. \tag{A1}$$

Potential inside the ith sphere reads

$$\Phi_{\mathrm{ins},i}(\vec{r}) = -\vec{E}_{ci}\cdot\vec{r}_i + \vec{d}_i\cdot\vec{r}_i. \tag{A2}$$

For $\vec{r} = \vec{c}_i + a_i\vec{n}$ or $|\vec{r}_i| = |\vec{r} - \vec{c}_i| = a_i$, $\Phi_{\mathrm{out},i} = \Phi_{\mathrm{ins},i}$ and

$$\begin{aligned}\vec{\nabla}\Phi_{\mathrm{ins},i} &= -\vec{E}_{ci} + \vec{d}_i,\\ \vec{\nabla}\Phi_{\mathrm{out},i} &= -\vec{E}_{ci} + a_i^3\frac{\vec{d}_i}{|\vec{r}_i|^3} - 3a_i^3\frac{(\vec{d}_i\cdot\vec{r}_i)\vec{r}_i}{|\vec{r}_i|^5}.\end{aligned} \tag{A3}$$

Taking into account that $\vec{n} = (\vec{r} - \vec{c}_i)/a_i$, Eqs. (8), (A3), and the definition of induction [the first formula in Eqs. (7)] yield:

$$2\varepsilon_{\mathrm{out}}\vec{d}_i\cdot\vec{n} + \varepsilon_i\vec{d}_i\cdot\vec{n} + (\varepsilon_{\mathrm{out}} - \varepsilon_i)\vec{E}_{ci} = \frac{\gamma}{\varepsilon_0}. \tag{A4}$$

Introducing $\kappa_{\varepsilon i} = \varepsilon_i/\varepsilon_{\mathrm{out}} - 1$ yields Eq. (11). Equation (12) is derived similarly by replacing $\kappa_{\varepsilon i} \to \kappa_{\sigma i} = \sigma_i/\sigma_{\mathrm{out}} - 1$ and $\gamma/\varepsilon_0 \to -(\gamma_i + \vec{v}_c\cdot\vec{\nabla}\gamma_i)$.

Equation (A3) and the formula for the total surface charge density Eq. (3) imply that $\gamma_T = 3\varepsilon_0\vec{d}\cdot\vec{n}$.

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