Electrohydrodynamic interactions of drop pairs

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We study the pairwise interactions of drops in an applied uniform DC electric field within the framework of the leaky dielectric model. We develop three-dimensional numerical simulations using the boundary integral method and an analytical theory assuming small drop deformations. We apply the simulations and the theory to explore the electrohydrodynamic interactions between two identical drops with arbitrary orientation of the their line of centers relative to the applied field direction. Our results show complex dynamics depending on the conductivities and permittivities of the drops and suspending fluids, and the initial drop pair alignment with the applied electric field.

1. Introduction

The interaction of fluids and electric fields is at the heart of natural phenomena such as the disintegration of raindrops in thunderstorms and many practical applications such as electrosprays (Ganan-Calvo et al. 2018), microfluidics (Stone et al. 2005), and crude oil demulsification (Eow & Ghadiri 2002). Many of these processes involve drops and there has been growing interest in understanding the drop-drop interactions of in the presence of electric fields.

A drop placed in an electric field polarizes if its permittivity and/or conductivity are different than the suspending fluid. The polarization leads to a jump in the electric stresses across the drop interface. In the case of fluids that are perfect dielectrics, only the normal electric stress is discontinuous at the interface. If the electric pressure can be balanced by surface tension, the drop adopts a steady prolate ellipsoidal shape and the fluids are quiescent. The physical picture changes dramatically if the fluids are conducting materials. Finite conductivity, even if very low, enables the passage of electric current and electrical charge accumulates at the drop interface. The electric field acting on this induced surface charge creates tangential electric stress, which shears the fluids into motion. The complicated interplay between the electrostatic and viscous fluid stresses results in either oblate or prolate drop deformation in weak fields (Taylor 1966), and complex dynamics in strong fields, such as break-up (Torza et al. 1971; Sherwood 1988), streaming either from the drop poles (Taylor 1964; de la Mora 2007; Collins et al. 2013; 2008; Herrada et al. 2012; Sengupta et al. 2017), or equator (Brosseau & Vlahovska 2017; Waggoner et al. 2020), and electrorotation (Ha & Yang 2000; Salipante & Vlahovska 2010; 2013; Das & Saintillan 2017).

While the prototypical problem of an isolated drop in a uniform electric field has...
been extensively studied (see for a recent review (Vlahovska 2019)), investigations of the collective dynamics of many drops are scarce (Fernandez 2008a, b; Casas et al. 2019) and mainly focused on the near-contact interaction preceding electrocoalescence (Anand et al. 2019; Roy et al. 2019). The dynamics of drop approach and interactions at arbitrary separations have been considered mainly in the case of droplet pairs aligned with the electric field (Sozou 1975; Baygents et al. 1998; Lin et al. 2012; Mhatre et al. 2015; Zabarankin 2020), because the axial symmetry greatly simplifies the calculations. These studies revealed that in weakly conducting fluid systems, which can be modeled using the leaky dielectric model (Melcher & Taylor 1969), the hydrodynamic interactions due to the electric-shear-driven flow can play a significant role. For example, in the case of a drop with drop-medium ratio of conductivities $R$ and permittivities $S$ such that $R/S > 1$, the electrohydrodynamic flow generates repulsion which opposes the electrostatic attraction due to the drop dipoles and the drops move apart.

The general case of an electric field applied at an angle to the line joining the centers of the two drops is studied only to a limited extent experimentally (Mhatre et al. 2015) and via numerical simulations in two dimensions (Dong & Sau 2018). This configuration has been systematically analyzed only for a pair of non-deformable, ideally polarizable spheres (Saintillan 2008). In this case, the flow about the spheres has the same stresslet-quadrupole structure as the electrohydrodynamic flow about a drop with $R/S < 1$ even though the flow is due to induced charge electroosmosis, unlike the leaky-dielectric drops where Debye charge clouds are absent. The study showed that the pair dynamics are not simple attraction or repulsion; depending on the angle between the center-to-center line with the undisturbed electric field, the relative motion of the two spheres can be quite complex: they attract in the direction of the field and move towards each other, pair up, and then separate in the transverse direction. To the best of our knowledge, such dynamics in the case of drops has not been reported. Motivated by the observed intricate trajectories of ideally polarizable spheres and the potential similarities to the electrohydrodynamic interactions of drops with $R/S < 1$, we set up to investigate the effects of drop electric properties (conductivity ratio $R$ and permittivity ratio $S$) and deformability on the relative motion of a drop pair initially misaligned with the applied field.

2. Problem formulation

Let us consider two identical neutrally-buoyant and charge-free drops with radius $a$, viscosity $\eta_d$, conductivity $\sigma_d$, and permittivity $\varepsilon_d$ suspended in a fluid with viscosity $\eta_s$, conductivity $\sigma_s$, and permittivity $\varepsilon_s$. The mismatch in drop and suspending fluid properties is characterized by the conductivity, permittivity, and viscosity ratios

$$R = \frac{\sigma_d}{\sigma_s}, \quad S = \frac{\varepsilon_d}{\varepsilon_s}, \quad \lambda = \frac{\eta_d}{\eta_s}. \quad (2.1)$$

The distance between the drops’ centroids is $d$ and the angle between the drops’ line-of-centers with the applied field direction is $\Theta$. The unit separation vector between the drops is defined by the difference between the position vectors of the drops’ centers of mass $\mathbf{d} = (x_2^d - x_1^d)/d$. The unit vector normal to the drops line-of-centers and orthogonal to $\mathbf{d}$ is $\mathbf{t}$. The problem geometry is sketched in Figure 1.

We adopt the leaky dielectric model (Melcher & Taylor 1969), which assumes creeping flow and charge-free bulk fluids acting as Ohmic conductors. Albeit an approximation of the actual electrokinetic problem (Saville 1997; Schnitzer & Yariv 2015; Ganan-Calvo et al. 2016; Mori & Young 2018; Ganan-Calvo et al. 2018), the leaky dielectric model has
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been successful in modeling many phenomena not only in poorly conducting fluids such as oils, but also aqueous electrolyte solutions such as in cell-mimicking vesicle systems (Vlahovska et al. 2009; Vlahovska 2019). The assumption of charge-free fluids decouples the electric and hydrodynamic fields in the bulk. Accordingly,

\[ \nabla \cdot T_{hd} = \eta \nabla^2 u - \nabla p = 0, \quad \nabla \cdot T_{el} = 0, \]

where \( T_{hd}^{ij} = -p\delta_{ij} + \eta(\partial_j u_i + \partial_i u_j) \) is the hydrodynamic stress and \( \delta_{ij} \) is the Kronecker delta function; \( u \) and \( p \) are the fluid velocity and pressure. The electric stress is given by the Maxwell stress tensor \( T_{el}^{ij} = \varepsilon (E_i E_j - E_k E_k \delta_{ij})/2 \). The coupling of the electric field and the fluid flow occurs at the drop interfaces \( D \), where the charges brought by conduction accumulate. The Gauss’ law dictates that the electric field \( E \) in the electroneutral bulk fluids is solenoidal, \( \nabla \cdot E = 0 \), however at the drop interface the electric displacement field, \( \varepsilon E \), is discontinuous and its jump corresponds to the surface charge density

\[ \varepsilon (E_n^s - SE_n^d) = q, \quad x \in D \]  

where \( E_n = E \cdot n \) and \( n \) is the outward pointing normal vector to the drop interface. The surface charge density adjusts to satisfy the current balance

\[ \frac{\partial q}{\partial t} + \nabla_s \cdot (uq) = \sigma_s (E_n^s - RE_n^d), \quad x \in D. \]

In this study, we neglect charge relaxation and convection, thereby reducing the charge conservation equation to continuity of the electrical current across the interface as originally proposed by Taylor (1966)

\[ E_n^s = RE_n^d. \]

The electric field acting on the induced surface charge gives rise to electric shear stress at the interface. The tangential stress balance yields

\[ (I - \text{nn}) \cdot (T^s - T^d) \cdot n + qE_t = 0, \quad x \in D, \]

where \( E_t = E - E_n \) is the tangential component of the electric field, which is continuous
across the interface, and \( I \) is the idemfactor. The normal stress balance is
\[
\mathbf{n} \cdot (\mathbf{T}^s - \mathbf{T}^d) + \frac{1}{2} \left( (E_n^s)^2 - S (E_n^d)^2 - (1 - S)E_t^2 \right) = \gamma \nabla_s \cdot \mathbf{n}, \quad \mathbf{x} \in \mathcal{D}, \tag{2.7}
\]
where \( \gamma \) is the interfacial tension.

Henceforth, all variables are nondimensionalized using the radius of the undeformed drops \( a \), the undisturbed field strength \( E_0 \), a characteristic applied stress \( \tau_c = \varepsilon_s E_0^2 \), and the properties of the suspending fluid. Accordingly, the time scale is \( t_c = \eta_c / \tau_c \) and the velocity scale is \( u_c = a \tau_c / \eta_c \). The ratio of the magnitude of the electric stresses and surface tension defines the electric capillary number
\[
Ca = \frac{\varepsilon_s E_0^2 a}{\gamma}. \tag{2.8}
\]
The simplification of the charge conservation equation Eq. (2.4) to Eq. (2.5) implies \( \varepsilon_s E_0^2 / (\eta_s \sigma_s) \ll 1 \). This condition is satisfied for the typical fluids used in experiments such as castor oil (conductivity is \( \sim 10^{-11} \text{ S/m} \), viscosity is \( \sim 1 \text{ Pa.s} \)) and low field strengths \( E_0 \sim 10^4 \text{ V/m} \). Furthermore, the viscous time scale for drops of typical size \( a \sim 1 \text{ mm} \) is much shorter than the electrohydrodynamic flow time scale \( \eta_c / (\varepsilon_s E_0^2) \), which justifies the use of the steady Stokes equation to describe the fluid flow Eq. (2.2).

3. Numerical method

We utilize the boundary integral method to solve for the flow and electric fields. Details of our three-dimensional formulation can be found in (Sorgentone et al. 2019). In brief, the electric field is computed following (Lac & Homsy 2007; Baygents et al. 1998):
\[
E^\infty - \sum_{j=1}^2 \int_{\mathcal{D}_j} \frac{\hat{x}}{4\pi r^3} (E^s - E^d) \cdot \mathbf{n} dS(y) = \begin{cases} 
E^s(x) & \text{if } x \text{ inside } \mathcal{D}, \\
\frac{1}{2} (E^d(x) + E^s(x)) & \text{if } x \in \mathcal{D}, \\
E^s(x) & \text{if } x \text{ outside } \mathcal{D},
\end{cases} \tag{3.1}
\]
where \( \hat{x} = x - y \) and \( r = |\hat{x}| \). The normal and tangential components of the electric field are calculated from the above equation
\[
E_n(x) = \frac{2R}{R + 1} E^\infty \cdot \mathbf{n} + \frac{R - 1}{R + 1} \sum_{j=1}^2 \mathbf{n}(x) \cdot \int_{\mathcal{D}_j} \frac{\hat{x}}{2\pi r^3} E_n(y) dS(y),
\tag{3.2}
\]
\[
E_t(x) = \frac{E^s + E^d}{2} - \frac{1 + R}{2R} E_n \mathbf{n}.
\]
For the flow field, we have developed the method for fluids of arbitrary viscosity, but for the sake of brevity here we list the equation in the case of equiviscous drops and suspending fluids. The velocity is given by
\[
2 \mathbf{u}(x) = -\sum_{j=1}^2 \left( \frac{1}{4\pi} \int_{\mathcal{D}_j} \left( \frac{\mathbf{f}(y)}{\epsilon} - \mathbf{f}^E(y) \right) \cdot \left( \frac{I}{r} + \frac{\hat{x} \hat{x}}{r^3} \right) dS(y) \right), \tag{3.3}
\]
where \( \mathbf{f} \) and \( \mathbf{f}^E \) are the interfacial stresses due to surface tension and electric field
\[
\mathbf{f} = \nabla \cdot \mathbf{n}, \quad \mathbf{f}^E = (E^s \cdot \mathbf{n}) E^s - \frac{1}{2} (E^s \cdot E^s) \mathbf{n} - S \left( (E^d \cdot \mathbf{n}) E^d - \frac{1}{2} (E^d \cdot E^d) \mathbf{n} \right). \tag{3.4}
\]
Drop velocity and centroid are computed from the volume averages
\[
U_j = \frac{1}{V} \int_{V_j} u dV = \frac{1}{V} \int_{D_j} \mathbf{n} \cdot (\mathbf{u} x) \, dS, \quad x^c_j = \frac{1}{V} \int_{V_j} x dV = \frac{1}{2V} \int_{D_j} \mathbf{n} \cdot (x \cdot x) \, dS. \quad (3.5)
\]

To solve the system of equations Eq. (3.2) and Eq. (3.3) we utilize the boundary integral method presented in Sorgentone et al. (2019). In the current study, however, we modify the time-stepper scheme to the adaptive fourth order Runge-Kutta introduced in Kennedy & Carpenter (2003). All variables are expanded in spherical harmonics which provides an accurate representation even for relatively low expansion order. In this respect, to make sure that all the geometrical quantities of interest (e.g. mean curvature) are computed with high accuracy as well, we adopt an adaptive upsampling procedure introduced by Rahimian et al. (2015) which is based on the decay of the mean curvature spectrum and seems to work very well for this kind of simulation. When the drops are well separated from each other, the regular quadrature based on the trapezoidal rule in the longitudinal direction and on the Gauss-Legendre quadrature rule in the non-periodic direction works well. As they get closer, regular quadrature on a finer grid can still be used. Here, the density is interpolated to the finer (upsampled) grid, where the nearly singular kernel is better resolved. But at some point, a special quadrature method is needed since the quadrature error grows exponentially as we approach the surface and it is not possible to resolve the problem by grid refinement, i.e. upsampling. In Sorgentone & Tornberg (2018) a numerical procedure based on interpolation first introduced by Ying et al. (2006) is discussed and optimized to handle these complicated situations. The idea introduced in Ying et al. (2006) for the nearly singular integration was to find the point \( x_s \) on the surface that is closest to the target point \( x_0 \). Then, continuing along a line that passes through \( x_s \) and \( x_0 \), the integral is evaluated at a number of points \( x_1, \ldots, x_n \) further away from the surface. This can be done by regular quadrature on the standard grid or on the upsampled grid, depending on how far the target point is from the surface. The value of the integral on the surface (at \( x_s \)) needs to be computed by a specialized quadrature rule for singular integrals. At this point a 1D Lagrangian interpolation is used to compute the value at \( x_0 \) by interpolating the values at \( x_s \) and \( x_i, i = 1,\ldots,N. \) In Sorgentone & Tornberg (2018) it has been shown how to optimize this procedure, implementing a cell list algorithm to hierarchically find the closest point on the surface and using the spherical harmonic expansion to interpolate the onsurface integral value previously obtained on the whole surface (at the grid points only) by the special quadrature for singular integrals introduced in Veerapaneni et al. (2011) Rahimian et al. (2015). The accuracy of the method depends on the numerical parameters involved: the maximum distance before we need to upsample the grid for the regular quadrature (that of course will depend on the grid resolution), the upsampling rate used in the intermediate region, the number of points used for interpolation for target points in the nearest region, the distance and the distribution of these points (Sorgentone & Tornberg (2018)). We also use the spectral reparametrization technique presented in the same paper, designed to keep the representation optimal even under strong deformations. In our work, in order to be able to run long simulations and well resolve the close interactions, we set the spherical harmonic expansion order to \( p = 9 \), and for the nearly singular quadrature, we set the upsampling rate in the intermediate region to 4 and the number of interpolating points to 8. The viscosity contrast is \( \lambda = 1. \) Unless otherwise explicitly stated, the electric capillary number is \( Ca = 0.1. \)

Our numerical method was validated against the simulation results of Baygents et al. (1998) and an analytical theory developed by us and presented in the next Section. Figure 2 shows the results for the drop steady velocity as a function of the drop centroid...
Figure 2. Comparison between our fully three-dimensional simulations and the axisymmetric simulations of a drop pair aligned with the field by Baygents et al. (1998). (a) Radial component of the steady drop velocity as a function of separation for \( \text{Ca} = 0.1 \) (red dots) and 1 (blue dots) for a drop with \( R = 5, S = 4 \). Black dots is the data from Figure 9 in Baygents et al. (1998) with \( \text{Ca} = 1 \). Solid line is the theoretical \( U_2 \cdot \hat{d} \) given by Eq. (4.7). The drop velocity at large separations shows the \( 1/d^2 \) behavior of a stresslet flow. (b) Steady velocity of a perfectly dielectric drop \( R = S = 5 \) corresponding to Figure 4a in Baygents et al. (1998). The black dots are from our fully 3D code, the solid line is the DEP velocity, Eq. (4.7), showing \( 1/d^4 \) dependence.

Figure 3. Comparison between the simulations (black) and the analytical theory (red) for a drop pair with \( R = 1, S = 3 \), initial separation 3.5, and initial angle between the line of centers and the applied field direction 50°. The trajectory was computed from the relative drop velocity \( \mathbf{U} = 2U_2 \hat{d} \) Eq. (4.7). Time evolution of the (a) separation, (b) angle between the line of centers and applied field direction, (c) radial component of the relative velocity \( \mathbf{U} \cdot \hat{d} \), and (d) tangential component of the relative velocity \( \mathbf{U} \cdot \hat{t} \).
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separation for drops aligned with the field. Figure 3 illustrates the more general case of drops initially misaligned with the field. The simulations agree very well with the theory and show complex dynamics such as drops line-of-centers rotating away from the applied field direction and interaction switching from attraction to repulsion. These dynamics will be explored in more detail in Section 5.

4. Theory: Far-field interactions

To gain more physical insight, it is instructive to analyze the interaction of two widely separated spherical drops. In this case, the drops can be approximated by point-dipoles. The disturbance field \( \mathbf{E}_1 \) of the drop dipole \( \mathbf{P}_1 \) induces a dielectrophoretic (DEP) force on the dipole \( \mathbf{P}_2 \) located at \( x^c_2 = d \hat{d} \), given by

\[
\mathbf{F}(d) = \mathbf{P}_1 \mathbf{P}_2 \cdot \nabla \left( \frac{1}{r^3} - \frac{3 \mathbf{x} \mathbf{x}}{r^5} \right) \bigg|_{r=d}, \quad \mathbf{P}_1 = \mathbf{P}_2 = \frac{R-1}{R+2} \mathbf{E}^\infty
\]

The drop velocity under the action of this force can be estimated from Stokes law,

\[
\mathbf{U}_{\text{dep}}^2 = -\frac{\mathbf{F}}{\zeta} = -\frac{\mathbf{F}}{2\pi \rho (3 \lambda + 2)/(\lambda + 1)}. \quad (4.1)
\]

It is attractive if \( \Theta < \Theta_c = \arccos \left( \frac{1}{\sqrt{3}} \right) \approx 54.7^\circ \), e.g., when the drops are lined up with the field, and repulsive if the line of centers of the two drops is perpendicular to the field.

The electrohydrodynamic (EHD) flow about an isolated, spherical drop in an applied uniform electric field is a combination of a stresslet and a quadrupole (see Appendix for the flow evolution upon application of the electric field). At steady state,

\[
\mathbf{u} = \frac{9}{10} \frac{S - R}{(2 + R)(\lambda + 1)} \mathbf{E}^\infty \mathbf{E}^\infty : \left[ \left( \frac{1}{r^3} - \frac{3 \mathbf{x} \mathbf{x}}{r^5} \right) \mathbf{x} + \frac{1}{3} \nabla \left( \frac{1}{r^3} - \frac{3 \mathbf{x} \mathbf{x}}{r^5} \right) \right]. \quad (4.3)
\]

At the surface of the drop,

\[
\mathbf{u}(r=1) = \beta_T \sin(2\Theta) \hat{\Theta}, \quad \beta_T = \frac{9}{10} \frac{R - S}{(1 + \lambda) (R + 2)^2}. \quad (4.4)
\]

If \( R/S < 1 \), the surface flow is from pole to equator, i.e, fluid is drawn in at the poles and pushed away from the drop at the equator. The flow direction is reversed for \( R/S > 1 \).

A second drop moves in response to the electrohydrodynamic flow Eq. (4.3). The drop translational velocity is found from Faxen’s law [Kim & Karrila 1991]

\[
\mathbf{U}_{\text{ehd}}^2 = \frac{1 + \lambda}{2(3 \lambda + 2)} \nabla^2 \mathbf{u}|_{r=d}. \quad (4.5)
\]

Inserting Eq. (4.3) in the above equation leads to

\[
\mathbf{U}_{\text{ehd}}^2 = \beta_T \left( \frac{1}{d^2} - \frac{2}{d^4} \left( \frac{1 + 3 \lambda}{2 + 3 \lambda} \right) \right) (-1 + 3 \cos^2 \theta) \hat{d} - \frac{2 \beta_T}{d^4} \left( \frac{1 + 3 \lambda}{2 + 3 \lambda} \right) \sin(2\Theta) \hat{t} + O(d^{-5}). \quad (4.6)
\]

Combining the electrohydrodynamic and the dielectrophoretic velocities yields

\[
\mathbf{U}_2 = \frac{\beta_T}{d^2} (-1 + 3 \cos^2 \theta) \hat{d} - \Phi(R, S, \lambda) \frac{2}{d^4} \left( (-1 + 3 \cos^2 \theta) \hat{d} + \sin(2\Theta) \hat{t} \right), \quad (4.7)
\]
Figure 4. (a) Phase diagram of drop deformations and alignment with the field for different viscosity ratios. The solid line corresponds to $\Phi(\lambda, R, S) = 0$ given by Eq. (4.8). In the shaded regions, the line of centers of the two drops rotates away from the applied field direction $\Phi < 0$. The dashed line corresponds to the Taylor discriminating function Eq. (A 3); in the parameter space above it, drop deformation is oblate. (b) Critical separation above which EHD dominates the interactions for $\lambda = 1$ and $S = 0.1$ (blue), $S = 1$ (black) and $S = 10$ (red).

where

$$\Phi = \frac{1 + 3\lambda}{2 + 3\lambda} \left( \beta_T + 3\beta_D \frac{1 + \lambda}{1 + 3\lambda} \right). \quad (4.8)$$

The discriminant $\Phi$ quantifies the drop pair alignment with the field and the interplay of EHD and DEP interactions in drop attraction or repulsion. The line of centers between two drops with $\Phi > 0$ rotates towards a parallel orientation with respect to the applied electric field, since $\dot{\Theta} = U_2 \cdot \hat{z} \sim -\Phi$. However, in the case of $\Phi < 0$ (which occurs only for $R/S < 1$ drops), the line of centers between the drops rotates towards a perpendicular orientation with respect to the applied electric field. Figure 4a summarizes the regimes of alignment and deformation.

The relative radial motion of the two drops at a given separation depends on $\Phi$ and $\beta_T$, where $\beta_T \hat{z} \ddot{z}$ is the strength of the far-field EHD stresslet flow

$$u_r(x) = \beta_T \left( -1 + 3\cos^2 \Theta \right) \frac{x}{r^2}. \quad (4.9)$$

There is a critical separation $d_c$ corresponding to $U_2(d_c) \cdot \hat{d} = 0$, which yields $d_c^2 = 2\Phi/\beta_T$. For $\Phi > 0$ and $R/S < 1$ ($\beta_T < 0$), $d_c$ does not exist and EHD and DEP interactions are cooperative and act in the same direction (note that system with $\Phi < 0$ and $R/S > 1$ cannot exist). For $\Phi > 0$ and $R/S > 1$ or $\Phi < 0$ and $R/S < 1$, there is competition between EHD and DEP, with the quadrupolar DEP winning out closer to the drops and the EHD taking over via the stresslet flow in the far-field. The fact that depending on separation drops may attract or repel in the case the antagonistic EHD and DEP interactions has been discussed by [Baygents et al., 1998; Zabarankin, 2020]. Note that for drops with $R/S < 1$, EHD effectively dominates DEP at all separations since $d_c$ is smaller than the minimum separation of spherical drops, 2. Figure 4b illustrates the dependence of the critical separation $d_c$ for three typical cases. If $S = 10$, $d_c$ is always less than the minimum separation between two spheres, 2, and accordingly the interactions are dominated by
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Figure 5. Radial component $U \cdot \hat{d}$ of the relative velocity of the two drops $U = U_2 - U_1$ at $t = 0$ as a function of the angle made between the applied field and the line of centers of the two spheres $E^\infty \cdot \hat{d} = \cos \Theta$. Initial separation is $d = 4$. (a) $R/S < 1$: $R = 0.1, S = 1$ (black), $R = 1, S = 10$ (blue). At $\Theta = 0$, both electrostatic (DEP) and electrohydrodynamic (EHD) interactions are attractive. At $\Theta = \pi/2$, both DEP and EHD are repulsive. (b) $R/S > 1$: $R = 1, S = 0.1$ (red), $R = 100, S = 1$ (green). Solid line corresponds to the velocity computed from the transient theory Eq. (A.7), while the dashed line corresponds to the steady drop velocity Eq. (4.7). Points are the numerical simulations.

5. Results and discussion

An isolated charge-neutral drop in a uniform DC electric field experiences no net force. However, a drop pair can move due to their mutual electrostatic (due to the induced surface charge) and hydrodynamic (due to the flow driven by the surface electric shear) interactions. While the theory in Section 4 describes the steady drop velocities, our simulations consider transient deformations of the drops. Here, we explore the pair dynamics at different initial configurations using the simulations and a transient small deformation theory. The transient theory is derived similarly to the steady velocity in the previous section and is summarized in the Appendix.

5.1. Initial drop interactions

The initial interaction of two drops at different misalignment with the applied field is illustrated in Figure 5 and Figure 6 by the dependence on $\Theta$ of the initial relative velocity $U = U_2 - U_1$ of the two drops. Figure 5 shows that the radial component of the velocity changes sign. In the case $R/S < 1$, the center-to-center electrostatic (DEP) and electrohydrodynamic (EHD) interactions work in the same direction. When the drop pair is aligned with the field ($\Theta = 0$), the drops attract. As $\Theta$ increases, the attraction decreases and changes to repulsion around $\Theta \approx \Theta_c$. The repulsion is strongest in the configuration with $\Theta = \pi/2$, i.e., line of drop centers perpendicular to the applied field. The critical angle at which the total interaction changes sign is close to the far-field result $\Theta_c = 54.7^\circ$ and its dependence on the separation between the drops is shown in Figure 6b. The tangential component of $U \cdot \hat{t} = -2\Phi \sin (2\Theta)$. Accordingly, it is maximal at $\Theta = \pi/4$ as confirmed by Figure 6b.
Figure 6. (a) Tangential component $\mathbf{U} \cdot \hat{\mathbf{t}}$ of the relative velocity of the two drops $\mathbf{U} = \mathbf{U}_2 - \mathbf{U}_1$ at $t = 0$ as a function of the angle made between the applied field and the line of centers of the two drops $\mathbf{E}^\infty \cdot \hat{\mathbf{d}} = \cos \Theta$. Initial separation is $d = 4$. $R = 1$, $S = 10$ (blue), $R = 0.1$, $S = 1$ (black), $R = 1$, $S = 0.1$ (red), $R = 100$, $S = 1$ (green). Solid line corresponds to the velocity computed from the transient theory Eq. (A 7), while the dashed line corresponds to the steady drop velocity Eq. (4.7). Points are the numerical simulations. (b) Critical value of the angle $\Theta$ for which the initial radial velocity between two spheres is zero, plotted as a function of the separation distance between the drops. This critical angle separates configurations for which drops attract ($\mathbf{U} \cdot \hat{\mathbf{d}} < 0$) and repel ($\mathbf{U} \cdot \hat{\mathbf{d}} > 0$). Points are the numerical simulations and the lines are added to guide the eye.

The case $R/S > 1$ is more complicated because the electrostatic (DEP) and electrohydrodynamic (EHD) interactions are antagonistic. As discussed in Section 4, the EHD interactions dominate only beyond a crossover separation $d_c = \sqrt{2\Phi/\beta_T}$. Based on the stresslet approximation, EHD interactions are predicted to change from repulsive to attractive as $\Theta$ increases, while the DEP follows the opposite trend. Figure 5b shows that indeed the interaction at $t = 0$ is dominated by the EHD contribution. The solution of the transient electrohydrodynamic problem, which accounts for transient deformation effects on the fluid flow, however highlights that the relative velocity can reverse sign before deformation reaches nearly steady state on a typical time scale $\sim C a$ (see Appendix). The steady state theory predicts that for the $R = 100$, $S = 1$ drops, $d_c = 23$ and drop interactions at steady state are dominated by DEP; hence the steady state theory (dashed line) follows the opposite trend to the $t = 0$ results (Figure 5b).

The question arises what happens after the initial attraction or repulsion? How do drop deformation, hydrodynamic and electrostatic interactions affect the drop trajectory? Here we show that their interplay gives rise to intricate trajectories.

5.2. Drop pair trajectories: evolution of separation and alignment with the field
Figures 7 and 8 illustrate the evolution of the center-to-center distance, the radial component of the relative velocity, and the deformation parameter of drop 1 in the case of drops initially placed in the two extreme configurations, aligned and perpendicular to the field, $\Theta = 0$ and $\Theta = \pi/2$, respectively. The simulations are compared to the steady theory, where the separation is computed from the radial velocity, $\dot{d} = \mathbf{U} \cdot \hat{\mathbf{d}}$ and the deformation parameter is for an isolated drop. The tangential component of the relative velocity, $\mathbf{U} \cdot \hat{\mathbf{t}}$, is zero during the interaction and accordingly the drop pair orientation with the field remains unchanged, i.e., the angle between the line of centers remains as the initial configuration. In both cases, $R/S < 1$ and $R/S > 1$, the drops attract in the $\Theta = 0$ configuration, and repel if aligned perpendicularly with the applied field, $\Theta = \pi/2$. However, the interaction in the $R/S < 1$ case is controlled by EHD, while in the $R/S > 1$
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Figure 7. Electrohydrodynamics of a pair of deforming drops with $R = 0.1$ and $S = 1$ with initial separation $d = 4$ and inclination $\Theta = 0$ (top) and $\Theta = \pi/2$ (bottom). Evolution of the center-to-center distance (first column), the radial component of the relative velocity (second column), and the deformation parameter (third column). Black line corresponds to the numerical simulations. Red line corresponds to the radial velocity and separation predicted by Eq. (4.7) and the deformation of an isolated drop Eq. (5.2).

Figure 8. Electrohydrodynamics of a pair of deforming drops with $R = 100$ and $S = 1$ with initial separation $d = 4$ and inclination $\Theta = 0$ (top) and $\Theta = \pi/2$ (bottom). Evolution of the center-to-center distance (first column), the radial component of the relative velocity (second column), and the deformation parameter (third column). Black line corresponds to the numerical simulations. Red line corresponds to the radial velocity and separation predicted by Eq. (4.7) and the deformation of an isolated drop Eq. (5.2). The insets show the sign reversal of the radial velocity.
case – by DEP, since the critical distance \( d_c \) in the considered system \( R = 100, S = 1 \) is about 23, much larger than the initial separation. The radial velocity, \( U \cdot \hat{d} \), varies in time and in the case \( R/S < 1 \) (Figure 7) does not change sign (it remains either negative, indicating attraction, or positive, indicating repulsion). In the \( \Theta = 0 \) case, drops attract and the distance between the drop decreases; if \( \Theta = \pi/2 \), the drops repel and the separation increases. In the case \( R/S > 1 \) (Figure 8), the radial velocity reverses sign on a short time scale \( \sim Ca \). If \( \Theta = 0 \), drops attract after a short transient repulsion and separation decreases in time. The opposite occurs in the \( \Theta = \pi/2 \) configuration. The theoretical trajectory computed from the steady state velocity and the simulations are in good agreement since drop shape remains close to a sphere and drop translation is slow compared to the deformation time scale.

The deformation parameter is defined as \( D_T = \frac{a_{||} - a_{\perp}}{a_{||} + a_{\perp}} \), where \( a_{||} \) and \( a_{\perp} \) are the drop lengths in directions parallel and perpendicular to the applied field. For an isolated drop, in weak fields \( (Ca \ll 1) \) the equilibrium shape is given by

\[
D_T = \frac{9Ca}{16(2 + R)^2} \left[ R^2 + 1 - 2S + 3(R - S) \frac{2 + 3\lambda}{5(1 + \lambda)} \right].
\]

(5.1)

Upon application of the field, the drop approaches the steady state monotonically (Esmeeeli & Sharifi 2011)

\[
D(t) = D_T \left( 1 - e^{-t/t_r} \right) \quad \text{where} \quad t_r = \frac{\eta_0 \alpha}{\gamma} \left( \frac{(3 + 2\lambda)(16 + 19\lambda)}{40(1 + \lambda)} \right).
\]

(5.2)

Figures 7 and 8 show that upon application of the field the drops deform into an oblate or prolate ellipsoid depending on the Taylor discriminating function. The deformation parameter increases monotonically, similarly to the isolated drop case, and approaches a nearly steady value, which is close to that for an isolated drop given by Eq. (5.1). Due to the axial symmetry, the deformation parameters of both drops are identical. The difference between the two drop and the isolated drop results is greater in the \( \Theta = 0 \) case because as the drops are moving closer their interaction is getting stronger; in the \( \Theta = \pi/2 \) configuration, the drops move away from each other and become more isolated.

The effect of the initial misalignment of the drop pair and the applied field direction is illustrated in Figure 9 with the three-dimensional trajectory of drops in the two canonical cases \( R/S < 1 \) and \( R/S > 1 \). While in most cases drops display monotonic separation or attraction, Figure 9 highlights some more intriguing dynamics: repulsion followed by attraction with centerline rotating towards the applied field direction (a) and (d), attraction followed by repulsion with centerline rotating towards the applied field direction (c), and attraction followed by repulsion with centerline rotating away from the applied field direction (b). The drops remain in the plane defined by the initial separation vector and the applied field direction, in this case the \( xz \) plane. The transient pairing dynamics are clearly seen in the trajectories in the \( xz \) plane. These dynamics are illustrated in more detail in Figure 10. In the systems \( R = 0.1 \) and \( S = 1 \) (\( \Phi > 0 \)), \( R = 1 \) and \( S = 10 \) (\( \Phi < 0 \)), \( R = 1 \) and \( S = 0.1 \) (\( \Phi > 0 \)), the electrohydrodynamic interactions are dominant; in particular, for a sphere, \( R = 1 \) completely switches off the DEP interaction. In the \( R = 1 \) and \( S = 10 \) (\( \Phi < 0 \)) case (Figure 10), the initial drop centerline angle is below \( \Theta_c \) and the EHD interaction is attractive. The drops initially attract along the direction of the electric field, but the rotation of the centerline away from the field axis increases the tilt angle above \( \Theta_c \) leading to repulsion and separation in direction perpendicular to the field. The angle between the separation vector and the applied field continuously increases and around 65° the interaction changes from attractive to repulsive. At this point the drops attain minimum separation, and after that the drops move away from each other with
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Figure 9. Trajectories of two identical drops with (a) \( R=0.1, S=1 \), (b) \( R=1, S=10 \), (c) \( R=1, S=0.1 \) and (d) \( R=100, S=1 \). Initially the drops are in the \( xz \) plane, the separation in all cases is \( d=4 \) and the angle with the applied field direction is (a) \( \Theta = 60^\circ \), (b) \( \Theta = 45^\circ \), (c) \( \Theta = 65^\circ \), and (d) \( \Theta = 80^\circ \). Bottom: trajectories in the \( xz \) planes.

velocity that overshoots. At long times the drop pair approaches a nearly perpendicular orientation relative to the field direction, where the repulsive DEP and EHD interactions push the drops apart. These “kiss-and-run” dynamics is similar to the one observed with ideally polarizable spheres (Saintillan 2008) and has implications to electrocoalescence since the switching from attraction to repulsion prevents drops from reaching proximity sufficient to initiate merger. In all other cases, for which \( \Phi > 0 \) drops move to align with the field. Figure 10a and 10c illustrate repulsion/attraction and attraction/repulsion dynamics. In both cases, the drop is released at an initial angle above the critical, but the \( R = 0.1 \) and \( S = 1 \) EHD stresslet flow is repulsive, while the \( R = 1 \) and \( S = 0.1 \) EHD flow is attractive. Since \( \Phi > 0 \), the drop centerline rotates towards the field direction bringing the drops into the range of angles where the electrohydrodynamic flow causes the drop interaction to reverse sign.

DEP interactions become very important for large conductivity ratios \( R \gg 1 \). As \( R \) increases the EHD flow weakens (see Eq. 4.3), while the DEP force plateaus as the dipole strength \( (R-1)/(R+2) \) approaches 1 (see Eq. 4.2). As a result the crossover separation beyond which the EHD flow becomes important increases. The \( R = 100, S = 1 \) case (Figure 10d) illustrates dynamics in this DEP dominated regime. Choosing an initial angle larger than \( \Theta_c \) causes the drops to repel, but \( \Phi > 0 \) drives rotation toward the field direction and crossing over into the angle range for attraction.

6. Conclusions and outlook

The three-dimensional interactions of a drop pair in an applied electric field are studied using numerical simulations and a small-deformation theory based on the the leaky dielectric model. We present results for the case of a uniform electric field and arbitrary angle between the drops’ line-of-centers and the applied field direction, where the non-axisymmetric geometry necessitates three-dimensional simulations.

The pair dynamics depend on the interplay between the electrohydrodynamic (EHD) and dielectrophoretic (DEP) interactions, which are cooperative in the case of \( R/S < 1 \), and antagonistic for \( R/S > 1 \). DEP interaction favors drop-pair alignment with the field
and is attractive for small angles and repulsive otherwise. The critical angle where center-to-center motion changes sign can be estimated from the point-dipole approximation, \( \Theta_c = \arccos \left( \frac{1}{\sqrt{3}} \right) \approx 54.7^\circ \). The EHD interaction depends on the sign of the induced free-charge dipole \( \sim (R - S) \). If \( R/S < 1 \), the pole to equator flow pulls the drops together when aligned parallel to the applied field direction and pushes them apart when the center-to-center line is perpendicular to the field; this scenario reverses for \( R/S > 1 \).

The critical angle which separates attraction from repulsion can be estimated from the stresslet approximation of the EHD flow and is the same as the dielectrophoretic force. Hence, to leading order in separation and drop deformation, both the DEP and EHD
change sign at $\Theta_c$. Unlike DEP, the EHD interaction can cause the drops’ line-of-centers to rotate toward or away from the applied field direction. The theory highlights the importance of the function $\Phi(\lambda, R, S)$, given by Eq. (4.8), which discriminates between the drop pair moving to align with the field or in a direction transverse to the field.

Our study finds that if the drop-pair angle with the field initially is close to the critical angle for reversal of the interaction sign, the drops do not experience monotonic attraction or repulsion; instead their trajectories follow three scenarios: motion in the direction of the field accompanied by either attraction followed by separation or vice versa (repulsion followed by attraction), and attraction followed by separation in a direction transverse to the field. The dynamics of drops with $R/S < 1$ and $\Phi < 0$ is similar to ideally-polarizable spheres [Saintillan 2008] due to the similarities of the flow pattern (despite different flow origins): the drops attract and move in the direction of the field and then separate in the transverse direction. Hence, coalescence will be prevented in such cases. Drops with $R/S > 1$ tend to align with the field but the sign of the interaction depends on drop separation. DEP dominates when drops are close, while EHD controls the far field interaction.

The comparison of the analytical theory and the simulations shows that the theory performs quite well in a wide range of drop separations and angles with the applied field direction for $Ca < 1$, and thus can serve as an efficient means to estimate drop pair dynamics and trajectories in an applied electric field. However, the simulations are indispensable in modeling the near-contact motions of the drops and drop dynamics in stronger fields. Our three-dimensional boundary integral method is also capable of simulating the dynamics of dissimilar drops (different size, viscosities, $R$ and $S$), and many drops, which we plan to explore in the future. Charge convection can also be included in order to study symmetry-breaking three-dimensional instabilities such as the Quincke electrorotation [Salipante & Vlahovska 2010, 2013, Vlahovska 2016].

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Appendix A. Transient migration velocity

Let us consider drop dynamics upon the application of an uniform electric field in the limit of small deformations $Ca \ll 1$. At leading order in $Ca$, the shape is described by $r_s = 1 + f_2(t) (-1 + 3 \cos^2 \theta)$, and the velocity field outside the drop at distance $r$ from the drop center and an angle $\theta$ with the applied field direction is given by [Vlahovska 2016]

\[ u = \left( \frac{\alpha + \beta}{r^2} - \frac{\beta}{r^4} \right) (-1 + 3 \cos^2 \theta) \hat{r} - \frac{\beta}{r^4} \sin(2\theta) \hat{\theta}, \]

where

\[ \alpha = \frac{15(\lambda + 1)}{(3 + 2\lambda)(16 + 19\lambda)} \left( F_T(R, S, \lambda) - Ca^{-1} \frac{8}{3} f_2(t) \right) \]

\[ \beta = \frac{12(2 + 3\lambda)}{(3 + 2\lambda)(16 + 19\lambda)} \left( B_T(R, S, \lambda) - Ca^{-1} f_2(t) \right). \]
where $F_T$ is the Taylor discriminating function

$$F_T(R, S, \lambda) = \frac{1}{(2 + R)^2} \left( R^2 + 1 - 2S + 3(R - S) \frac{2 + 3\lambda}{5(\lambda + 1)} \right),$$  \hspace{1cm} (A3)

and

$$B_T(R, S, \lambda) = \frac{9 \left( \lambda \left( 3R^2 + 13R - 19S + 3 \right) + 2 \left( R^2 + 6R - 8S + 1 \right) \right)}{2(3\lambda + 2)(R + 2)^2}. \hspace{1cm} (A4)$$

The shape evolution equation is obtained from the kinematic condition $\dot{r}_s = u_r(r = 1)$

$$\dot{f}_2 = \frac{15(\lambda + 1)}{(3 + 2\lambda)(16 + 19\lambda)} \left( F_T(R, S, \lambda) - Ca^{-\frac{8}{3}} f_2(t) \right),$$  \hspace{1cm} (A5)

Note that the Taylor deformation parameter is related to $f_2$, $D_2 = \frac{3}{2} f_2$, which leads to Eq. (5.2) describing the transient shape of an isolated drop.

If a second drop is present at location $x_2 = d\hat{d}$, its migration velocity due to the electrohydrodynamic flow of the first drop can be obtained using Faxen’s law (Kim & Karrila 1991)

$$U_{ehd}^{2, r} = \left( 1 + \frac{\lambda}{2(3\lambda + 2)} \nabla^2 \right) u(r = d). \hspace{1cm} (A6)$$

Inserting Eq. (A1) into the above equation yields

$$U_{ehd}^{2, r} = \left( \frac{\alpha + 3\beta}{r^2} - \frac{1}{r^4} \left( \beta + \frac{3\lambda}{2 + 3\lambda} (\alpha + \beta) \right) (-1 + 3\cos^2 \theta) \right) \left( -1 + 3\cos^2 \theta \right),$$  \hspace{1cm} (A7)

At steady state $f_2 = \frac{3}{8} CaF_T$, $\alpha = 0$ and $\beta$ reduces to the Taylor’s result

$$\beta_T = \frac{9(R - S)}{10(1 + \lambda) (2 + R)^2}, \hspace{1cm} (A8)$$

which leads to the steady EHD contribution to the migration velocity Eq. (4.7)

$$U_{ehd}^{2, r} = \left( \frac{\beta_T}{r^2} - \frac{1}{r^4} \left( \beta_T + \frac{3\lambda}{2 + 3\lambda} \beta_T \right) \right) (-1 + 3\cos^2 \theta),$$  \hspace{1cm} (A9)

$$U_{ehd}^{2, \theta} = -\frac{1}{r^4} \left( \frac{3\lambda}{2 + 3\lambda} \beta_T \right) \sin(2\theta).$$

Figure 11 compares trajectories computed from the transient and steady translation velocity. Decreasing $Ca$ shortens the transient period and the trajectory approaches the steady result. However, a long transient results in an offset.

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Figure 11. Comparison between the transient and steady migration theories with the numerical simulations. Centroid distance (a) and relative radial velocity (b) as a function of time for a pair of identical drops with $R = 1$, $S = 10$, aligned with the field ($\Theta = 0$) and initial separation $d = 4$, $Ca = 0.1$. The red line corresponds to the trajectory computed using the steady state velocity Eq. (4.7). The transient solution using Eq. (A 7) with $Ca = 0.05$ is given in black and $Ca = 0.1$ in blue.

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