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THE ROLES OF ELECTROHYDRODYNAMIC PHENOMENA IN THE MOTION OF DROPS AND BUBBLES

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ABSTRACT

The dynamics of small drops and bubbles are investigated with regard to the effects of an external electric field and an electrically charged layer situated on their interfaces. Specifically, stresses engendered due to the convection of surface charge alter both the motion and shape of single droplets and the bulk properties of suspensions.

INTRODUCTION

Motions produced inside and outside a neutrally buoyant drop immersed in a viscous fluid when an electric field is present are due to the interaction of induced charge with the field, an interaction which produces tangential shear stresses at the interface. A theory developed by G.I. Taylor(1) describes how the sense of the motion and the deformation depend on the various parameters when both fluids are poor conductors. He showed that, to leading order, the deformation and speed of circulation are proportional to $a\varepsilon E_0{}^2/\gamma$. Here a denotes the radius, ε the dielectric constant of the outer fluid, E_0 the field strength and γ the interfacial tension. This dimensionless group is, in essence, a comparision between the electrical stress tending to deform the drop and the restoring force of interfacial tension. Electrical and physical properties alone determine whether the deformed spheroid is oblate or prolate.

In Taylor's theory the distribution of induced charge, which depends on the electrical relaxation times for the two fluids, plays a central role. The distribution is antisymmetric with respect to the equitorial plane normal to the field. If the charge relaxation time, ε/σ, (σ denotes conductivity) of the inner fluid exceeds that of the outer fluid then flow is from the poles towards the equator. When the ratio of relaxation times is less than unity the charge distribution and flow are reversed. That theory, moreover, is in substantial agreement with experiments by Allan and Mason(2) and Torza, Cox and Mason,(3) who studied the deformation and burst of neutrally buoyant drops of various fluids.

Taylor's theory and its extensions to oscillatory electric fields by Torza, Cox and Mason(3) and Sozu(4) ignore, quite properly, the charge convection process which takes place at the interface. Bulk free charge is taken to be identically zero and the induced surface charge is convected by a motion which is $O(a\varepsilon E_0{}^2/\gamma)$. Thus, the alteration of stress due to convection of charge is $O(a\varepsilon E_0{}^2/\gamma)^2$ and therefore small.

If the drop undergoes translation, however, as is frequently the case if the densities are unmatched or the drop carries a charge, then charge convection induces electrical stresses which are $O(\eta U/\gamma \cdot a\varepsilon E_o^2/\gamma)$. These alter the translational speed and the shape. The purpose of this paper is to describe the influences of charge convection both because of its intrinsic interest and its relevance to drop breakup and coalescence.

It is readily seen that the shape alteration will differ from that found by Taylor since stresses resulting from charge convection due to streaming will be asymmetric. Thus, instead of a symmetrical deformation proportional to $P_2(\cos\theta)$ * the deformation will be represented in terms of $P_2(\cos\theta)$ and $P_3(\cos\theta)$. Experimental evidence for this sort of shape arising in the fashion proposed is sparse since all of the published work relates to neutrally buoyant drops. However, one prescient sequence of photographs by Torza, Cox and Mason does show the expected asymmetry [Figure 10, plate 7 of their paper]. It shows a drop flattened into an oblate spheroid, as would be expected from Taylor's theory. Then, perhaps due to the accumulation of charge, it begins to migrate and loses its symmetrical form. Although the amount of deformation is greater than that which could be rigorously modelled by a linearized theory the shape is clearly of the form expected from the consequences of charge convection.

Asymmetric deformation could also result from movement of the surface of a charged drop. Such a charge might be in the form of a monolayer or double-layer. Extant theories of the motion of drops with double-layers[5,6] allow for the convection of charge to some extent but the deformation is identically zero due to the extremely simple forms of the velocity and potential when charge relaxation is rapid. A more comprehensive theory is presented here which is applicable as well to cases where charge relaxation is slow enough for convection to be important.

The development proceeds along familiar lines with electrical effects described by the electrohydrodynamic simplifications of Maxwell's equations and motion inside and outside the globule described by solutions of the linearized Navier-Stokes equations. A key feature is the proper accounting for convection of surface charge. The system under study is depicted in Figure 1. A fluid sphere of radius a is immersed in another immiscible fluid. Both are Newtonian and incompressible with interfacial tension γ. Density and viscosity are denoted by ρ and ν, the shear viscosity by η. Carets are used to distinguish the variables pertaining to the globule. Three situations will be discussed:

(a) An uncharged globule in the presence of a uniform electric field, both fluids being ohmic conductors.

(b) A charged globule in a viscous non-conductor (the monolayer problem).

(c) A charged globule in a viscous conductor with a perfectly polarized interface (the double layer problem).

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* $P_n(\cos\theta)$ is a Legendre polynomial of order n, θ is measured from the rear stagnation point.

It will be assumed in the formal analysis that electrical stresses are small compared to interfacial tension, viz., $a\varepsilon E_o^2/\gamma<1$.

The remainder of the presentation is divided into sections dealing with the formal aspects of electric fields, forces and boundary conditions; fluid motion; then results for the uncharged drop, or drop with a mono-layer, and a drop with a double-layer. Before concluding, the effect of charge convection on the electrical conductivity of a suspension of fluid drops is discussed briefly.

ELECTRIC FIELDS, FORCES AND BOUNDARY CONDITIONS

Maxwell's equations in the form appropriate to electrohydrodynamic phenomena read

$$\nabla\times\underset{\sim}{E} = 0, \quad \nabla\cdot\underset{\sim}{D} = 4\pi q, \quad \text{and} \quad \frac{\partial}{\partial t} q + \nabla\cdot\underset{\sim}{J} = 0. \tag{1}$$

$\underset{\sim}{E}$, $\underset{\sim}{D}$, q, and $\underset{\sim}{J}$ stand for the electric field strength, dielectric displacement, bulk free charge density, and current, respectively. The constitutive relations are

$$\underset{\sim}{D} = \varepsilon\underset{\sim}{E}, \quad J = \sigma\underset{\sim}{E} + q\underset{\sim}{v}\ . \tag{2}$$

In the situation under investigation free charge is initially concentrated at the interface either as a mono- or a double-layer and remains there. It follows then that electrical phenomena can be described by means of potential functions which are

$$\hat{\phi}(r,\mu) = \sum_1^\infty c_n r^n P_n(\mu) \tag{3}$$

inside and

$$\phi(r,\mu) = \phi_d(r) - rP_1(\mu) + \sum_0^\infty D_n r^{-(n+1)} P_n(\mu) \tag{4}$$

outside. Here $\phi_d(r)$ denotes the double-layer potential in the absence of convection. Its precise form is unimportant here since we are dealing with thin layers and all that is required is the gradient at the interface. $\phi_d(r)$ is suppressed in the absence of a double-layer; when the external field is absent the term $-rP_1(\mu)$ is omitted.

The physical phenomena are determined by boundary conditions and they are set forth next.

A. Uncharged globule in the presence of an external field. Here both fluids are presumed to be ohmic conductors and at the interface the tangential components of the field are to be continuous,

$$E_t = \hat{E}_t \tag{5}$$

The other condition arises from the conservation of induced charge, Q, at the interface. Q is defined by the jump in εE_n, viz., $\langle \varepsilon E_n \rangle = Q$, where $\langle \varepsilon E_n \rangle$ stands for $\varepsilon E_n - \hat{\varepsilon}\hat{E}_n$. Balancing conduction to and from the interface against convection leads to the expression

$$\langle \sigma E_n \rangle + \nabla_s \cdot (Q\underset{\sim}{v}) = 0 \ . \tag{6}$$

$\nabla_s \cdot$ denotes the surface divergence, $\underset{\sim}{v}$ the velocity.

B. Charged globule in a viscous non-conductor (charge monolayer). Here the boundary conditions are the same as before although the absence of conductivity in the outer fluid, which serves to keep the globule charged, does simplify Equation 6 somewhat.

C. Charged globule in a viscous conductor with the interface perfectly polarized (charge double-layer). In this situation a thin double-layer approximation is employed wherein that part of the double-layer residing in the outer fluid is collected into a spherical sheath of charge. Charge is transported to and from this sheath by conduction and in it by convection; no charge crosses the interface. The balance expression reads

$$\sigma E_n + \nabla_s \cdot (Q\underset{\sim}{v}) = 0 \tag{7}$$

Processes which are ignored are tangential currents due to conduction, which are vanishingly small since the layer is thin, and radial charge convection, which vanishes since the radial velocity is zero at the interface. The net charge on the outer sheath is related to the gradient of the potential in the usual manner,[7] viz.,

$$\left(\frac{d\phi_d}{dr}\right)_{r=1} + D_o = \frac{4\pi Q_o}{\varepsilon E_o} \tag{8}$$

Here and elsewhere the potentials have been made dimensionless with the scale aE_o. The scale for length is a, Q_o is the average charge per unit area, and E_o is the (uniform) field strength far from the drop. Coefficients in Equation 3 are evaluated by requiring the tangential components of the field to be continuous.

FLUID MOTIONS

Since the fluids being considered are isothermal, incompressible and Newtonian and inertial effects neglected the well-known simplifications[8] of the equations of motion can be made. Solutions to the linearized equations can then be expressed in terms of stream functions for the motion inside the drop,

$$\left.\begin{aligned}\hat{\psi}(r,\mu) &= \frac{1}{2}\,\frac{\eta U}{\gamma}\,\frac{r^2-r^4}{1+\kappa}\,Q_1(\mu) + \sum_1^{\infty}[\hat{A}_n r^{n+3} + \hat{B}_n r^{n+1}]Q_n(\mu),\\ &\text{and outside,}\\ \psi(r,\mu) &= -\frac{1}{2}\,\frac{\eta U}{\gamma}\,(2r^2 - \frac{2+3\kappa}{1+\kappa}\,r + \frac{\kappa}{1+\kappa}\,r^{-1})Q_1(\mu)\\ &\quad + \sum_1^{\infty}[A_n r^{-(n-2)} + B_n r^{-n}]Q_n(\mu)\ .\end{aligned}\right\} \quad (9)$$

These expressions are in dimensionless form with U denoting the streaming speed far from the object, $\kappa = \hat{\eta}/\eta$ and

$$Q_n(\mu) = \int_{-1}^{\mu} P_n(\alpha)\,d\alpha\ . \quad (10)$$

The set of coefficients denoted as $\hat{A}_n$, $\hat{B}_n$, A_n, and B_n are evaluated from boundary conditions applied at the interface. These are: (i) continuity of the various components of velocity and (ii) continuity of the tangential components of the stress. The former reveals that

$$\hat{A}_n = -\hat{B}_n = A_n = -B_n \quad ; \quad n \geq 1\ . \quad (11)$$

Continuity of the stress is expressed as

$$\{r\,\frac{\partial}{\partial r}\,\frac{v_\theta}{r} + \frac{1}{r}\,\frac{\partial}{\partial\theta}\,v_r\} + T_t^{(e)} = \kappa\{r\,\frac{\partial}{\partial r}\,\frac{\hat{v}_\theta}{r} + \frac{1}{r}\,\frac{\partial}{\partial\theta}\,\hat{v}_r\} + \hat{T}_t^{(e)} \quad (12)$$

with the electrical stresses, $T_t^{(e)}$, evaluated from Maxwells stress tensor,(9)

$$T_t^{(e)} = \frac{a\varepsilon E_o^{\,2}}{4\pi\gamma}\,E_r E_\theta\Big|_{r=1} \quad (13)$$

in dimensionless form. The balance of normal stresses fixes the shape.

BEHAVIOR OF AN UNCHARGED GLOBULE

Explicit analytical solutions are obtained from simultaneous solution of the equations resulting from enforcing the boundary conditions. The calculations are straightforward and so tedious details are omitted. Complete expressions can be derived from those given in Reference 10.

Continuity of electrical stress and velocity enable one to reduce the problem to the evaluation of two sets of coefficients, B_n and C_n, say. Then an expansion scheme with $a\varepsilon E_o^2/\gamma \stackrel{d}{=} \delta$ treated as a small parameter is employed. From expressions of the form

$$\left.\begin{aligned} B_n &= B_n^{(0)} + B_n^{(1)} + \ldots \quad n = 1, 2, \ldots \\ C_n &= C_n^{(0)} + C_n^{(1)} + \ldots \quad n = 1, 2, \ldots \end{aligned}\right\} \tag{14}$$

we find

$$B_1^{(0)} = B_3^{(0)} = B_4^{(0)} = C_2^{(0)} = C_3^{(0)} = 0 \tag{15}$$

The orders of the coefficients that survive are:

$C_1^{(0)}:0(1)$; $C_2^{(1)}:0(\eta U/\gamma)$; $B_2^{(0)}, C_1^{(1)}, C_2^{(2)}, C_3^{(1)}:0(\delta)$; and

$B_1^{(1)}, B_3^{(1)}:0(\delta\eta U/\gamma)$. The other coefficients are of an even smaller order and therefore neglected. The formulas for $C_1^{(0)}$ and $B_2^{(0)}$ correspond to those given by Taylor.(1) $C_1^{(1)}$, $C_2^{(2)}$ and $C_3^{(1)}$ are associated with convection of induced charge by the electrically induced field and $C_2^{(1)}$ from the streaming. The velocity field consists of terms representing flow due to uniforming streaming and electrical stress arising from the induced charged whose distribution is altered, in turn, by the streaming.

For the settling velocity we find

$$\frac{U}{U_{st}} = \frac{3}{\frac{2+3\kappa}{1+\kappa} + f(R,S,\kappa)\frac{(\varepsilon E_o)^2}{\eta\sigma}} \tag{16}$$

when the direction of the uniform electric field is opposite to the gravitational field. Here $U_{st} = 2a^2 g(1-\hat{\rho}/\rho)/9\nu$ and

$$f(R,S,\kappa) \stackrel{.}{=} \frac{6}{5}\,\frac{1}{(1+\kappa)^2}\,\frac{R^2}{(3+2R)(2+R)^2}\,[3(1+R^{-1}) - \frac{\hat{\tau}_r}{\tau_r}][1 - \frac{\hat{\tau}_r}{\tau_r}] \; .$$

R stands for $\hat{\sigma}/\sigma$ and τ_r for the ratio of an electrical relaxation time, ε/σ, to the time scale for fluid motion, $a\eta/\gamma$, based on the outer fluid. Note that $\hat{\tau}_r/\tau_r = 1/RS$.

It is easy to show that the electric field can either increase or decrease the rate of translation of the globule, depending upon the electrical properties of the fluids under discussion. The particular condition under which the speed will increase is $1 < \hat{\tau}_r/\tau_r < 3(1+R^{-1})$. If this restriction is not met the motion of the droplet will be retarded. Figure 2 illustrates the magnitude of the effect for typical values of the parameters.

For an explanation of these results we examined the manner in which the streaming motion alters the induced charge and the tangential stresses (see Figure 3). In the case of a neutrally buoyant drop both the polarization and direction of fluid circulation are determined by the ratio of electrical relaxation times in the droplet and medium in the manner depicted. When the ratio of electrical relaxation times is unity the drop remains unpolarized and the electrical shearing stresses vanish.

When the electrical relaxation time of the droplet exceeds that of the surrounding fluid, the streaming motion alters the distribution as shown in Figure 3. In a manner analogous to that for the neutrally buoyant case, interaction of the altered charge distribution with the tangential component of the electric field results in the shear stress distribution indicated. These shearing stresses induce motions which enhance the streaming motion of the droplet. Compression of the negative charge toward the rear of the droplet results in electrical shearing stresses which retard motion. The settling speed of the droplet will be altered, then, depending upon the relative magnitudes of these two opposing phenomena.

A similar analysis for case (b) shows that the interaction of the altered charge distribution with the tangential component of the electric field always tends to retard the motion of the droplet when $\hat{\tau}_r/\tau_r < 1$. Motion is further retarded due to compression of positive charge toward the rear of the globule.

Deformation of the globule is due to electrical effects since uniform streaming *per se* causes no deformation if inertial effects are absent.[11] The deformation from the spherical form is represented as

$$\zeta(\mu) = \sum_{2}^{\infty} \beta_n P_n(\mu) \tag{17}$$

so that the center of mass is fixed and the globule is incompressible. The surviving coefficients, to $O(\delta)$, are β_2 and β_3. Normal stresses which give rise to β_2 are due to electrical phenomena present in the absence of streaming as found by Taylor while deformation due to charge convection is described by β_3.

Figure 4 depicts the manner in which a falling fluid sphere deforms when subject to a uniform electric field. The lack of fore to aft symmetry of the droplet can be understood in terms of the normal stresses engendered by the lack of symmetry of the charge distribution with respect to the equitorial plane of the droplet. This may be contrasted with the oblate spheroid which develops when charge convection is not taken into account (Figure 5).

The shape shown in Figure 4 is quite similar to the form depicted by Torza, Cox and Mason to which earlier reference was made. Calculations made using the parameters given in their paper are qualitatively the same although a direct comparison is not possible due to the lack of information on the rate of translation.

BEHAVIOR OF A CHARGED GLOBULE IN A VISCOUS NON-CONDUCTOR

Results for this situation, obtained in a fashion similar to that employed earlier, show that deformation tends to be prolate since the conductivity ratio is effectively infinite. Charge convection alters the symmetry, however. The translational velocity is altered by convection of both the induced charge and the net surface charge, viz.,

$$U = -\frac{\dfrac{2aQ_oE_o}{\eta}}{\dfrac{2+3\kappa}{1+\kappa} + \dfrac{9}{5}\,\dfrac{(\varepsilon E_o)^2}{(1+\kappa)^2 4\pi\ \eta\hat{\sigma}} + \dfrac{2}{3}\,\dfrac{1}{(1+\kappa)^2}\,\dfrac{4\pi Q_o{}^2}{\eta\hat{\sigma}}} \tag{18}$$

for a charged, neutrally buoyant drop in a viscous dielectric. It is worth noting here that convection of charge always produces shearing stresses which retard motion. This is consistent with the behavior identified with the uncharged globule where it was shown that if the ratio of electrical relaxation times is less than unity then motion is impeded.

BEHAVIOR OF A CHARGED GLOBULE WITH A PERFECTLY POLARIZED INTERFACE

A typical shape is shown as Figure 6. The asymmetry due to charge convection is evident and, in contrast to the situation shown in Figure 4, the front part of the drop is elongated due to the choice of physical properties. Nevertheless it should be noted that the deformation is $0(\tau_r)$ and when the relaxation is rapid as it would be with, say, a mercury drop in an ionic solution, the deformation will be quite small.

The translational velocity is

$$U = \frac{aQ_oE_o/\eta}{2+3\kappa+g} \tag{19}$$

where

$$g = \tau_r\,\frac{a\varepsilon E_o{}^2}{4\pi\gamma}\,\{(\frac{4\pi Q_o}{\varepsilon E_o})^2 + \frac{3}{10(1+\kappa)S^2}\,[\tfrac{1}{4}(8+10\kappa) - \tfrac{3S}{5}(7+8\kappa)]\}\ .$$

This shows, again, how charge convection impedes the rate of translation. If internal electric stresses are ignored by taking $\hat{\varepsilon} = 0$ $(S = \infty)$ then Equation (19) reduces the classical result due to Levich.(5,6)

CHARGE CONVECTION AND THE CONDUCTIVITY OF SUSPENSIONS

Processes of the sort just studied in connection with the behavior of single drops ought to manifest themselves in their effects on the properties of suspensions and drops. Two of the more obvious properties are electrical conductivity and viscosity. Indeed, just as it is possible to alter the properties of suspensions of solid, orientatible particles using external fields,[12] it will likewise be possible to alter matters in suspensions of fluid particles by, for example, controlling charge convection. Here we focus attention on the electrical conductivity of an otherwise motionless suspension of fluid particles.

The potentials inside and outside a single drop exposed to a uniform field are

$$\hat{\phi}(r,\mu) = -\frac{3}{2+R}\, rP_1(\mu) + C_1^{(1)} rP_1(\mu) + C_3^{(1)} r^3 P_3(\mu)$$

and

$$\phi(r,\mu) + \left[-r + \frac{R-1}{R+2}\, r^{-2}\right] P_1(\mu) + C_1^{(1)} r^{-2} P_1(\mu) + C_3^{(1)} r^{-4} P_3(\mu) \quad . \qquad (20)$$

Here

$$C_1^{(1)} = \frac{54}{25}\,\frac{1}{1+\kappa}\,\frac{R^2}{(2+R)^4}\left[1 - \frac{\hat{\tau}_r}{\tau_r}\right]^2 \frac{(\varepsilon E_o)^2}{4\pi\eta\sigma}$$

and

$$C_3^{(1)} = \frac{216}{25}\,\frac{1}{1+\kappa}\,\frac{R^2}{(4+3R)(2+R)}\left[1 - \frac{\hat{\tau}_r}{\tau_r}\right]^2 \frac{(\varepsilon E_o)^2}{4\pi\eta\sigma} \quad .$$

From Equation 20 we find that charge convection always acts so as to decrease the potential drop across a single particle, leading us to expect that the effective conductivity of a dilute suspension will be below that given in Maxwell's theory (see Reference 12). This turns out to be the case and, using an adaption of Batchelor's formalism[12] so as to account for charge convection, the effective electrical conductivity σ^* is found to be

$$\frac{\sigma^*}{\sigma} = 1 + \left[3\,\frac{R-1}{R+2} - (2R+1)C_1^{(1)}\right] c \quad . \qquad (21)$$

Here c denotes the volume fraction of fluid particles. Since $C_1^{(1)}$, which is always positive, depends on the field strength the conductivity is field dependent. Several other situations have been investigated[10] and results will be reported shortly.

CONCLUDING REMARKS

Attention was focused on two of the ways whereby the electrohydrodynamic effects of charge convection alter the behavior of single fluid drops, specifically their shape and rate of translation. In addition it was shown how the bulk conductivity of a suspension of drops can be altered by the same process. The principal limitations on the results arise from the restriction to small deformations, on the one hand, and the simplified models of interfacial behavior on the other.

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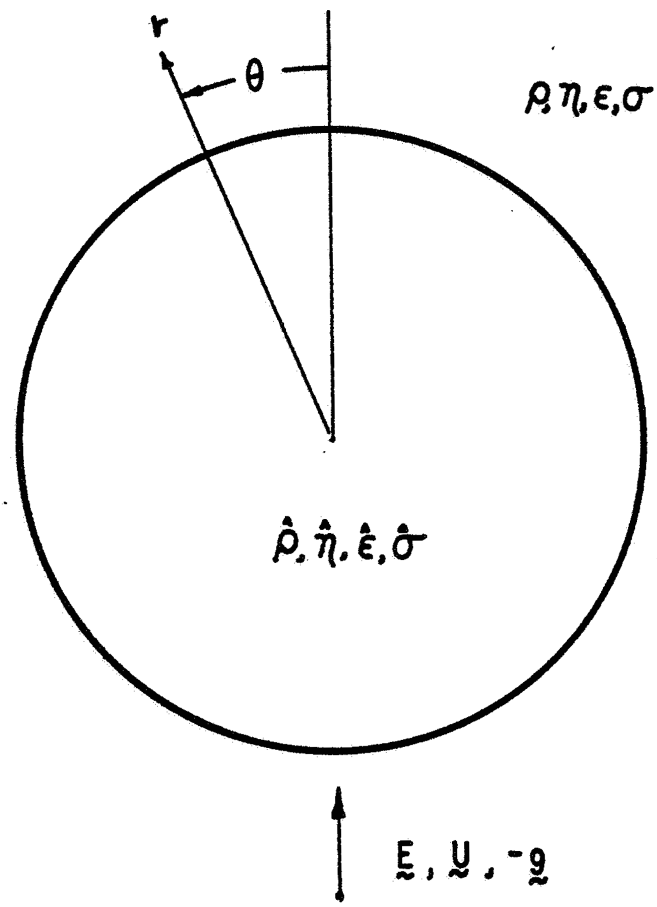
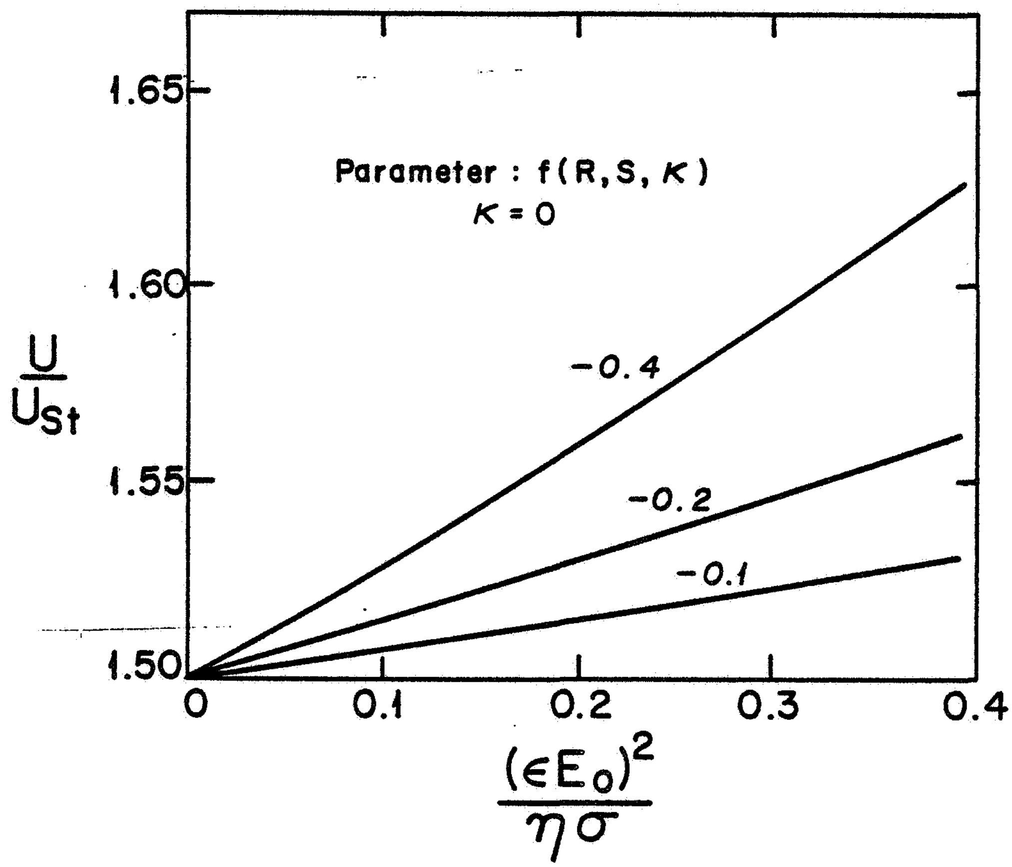


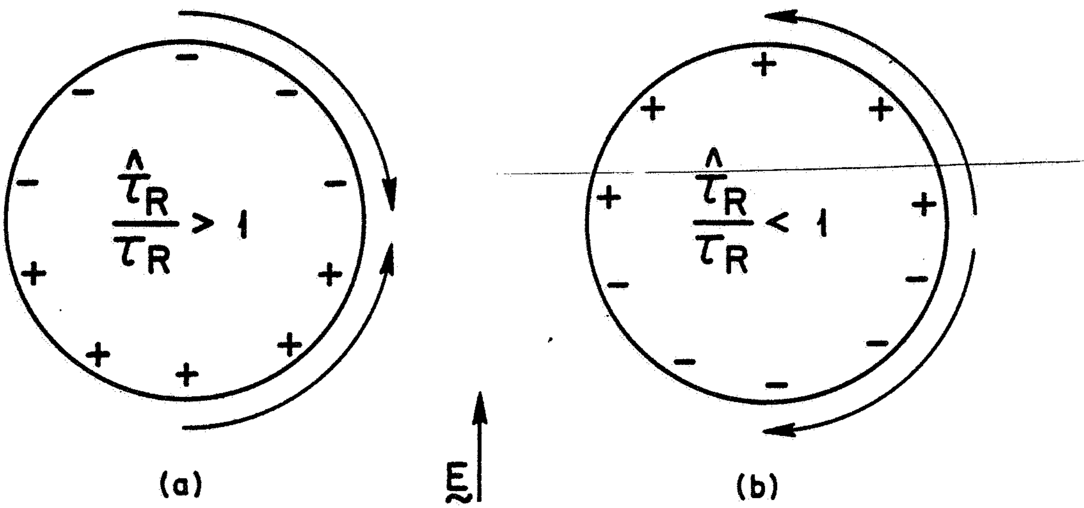
Definition Sketch

FIG. 1

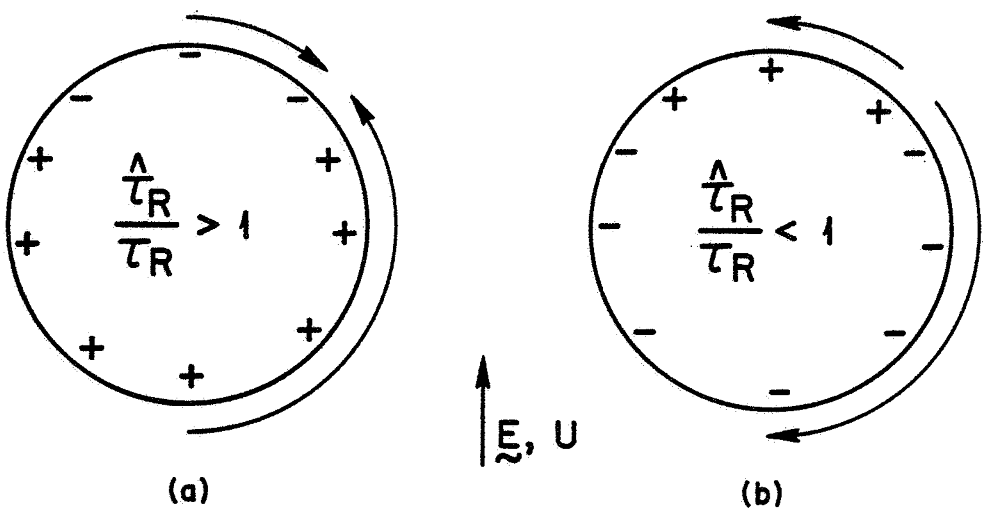


Translation of a Droplet in an External Field

FIG.2



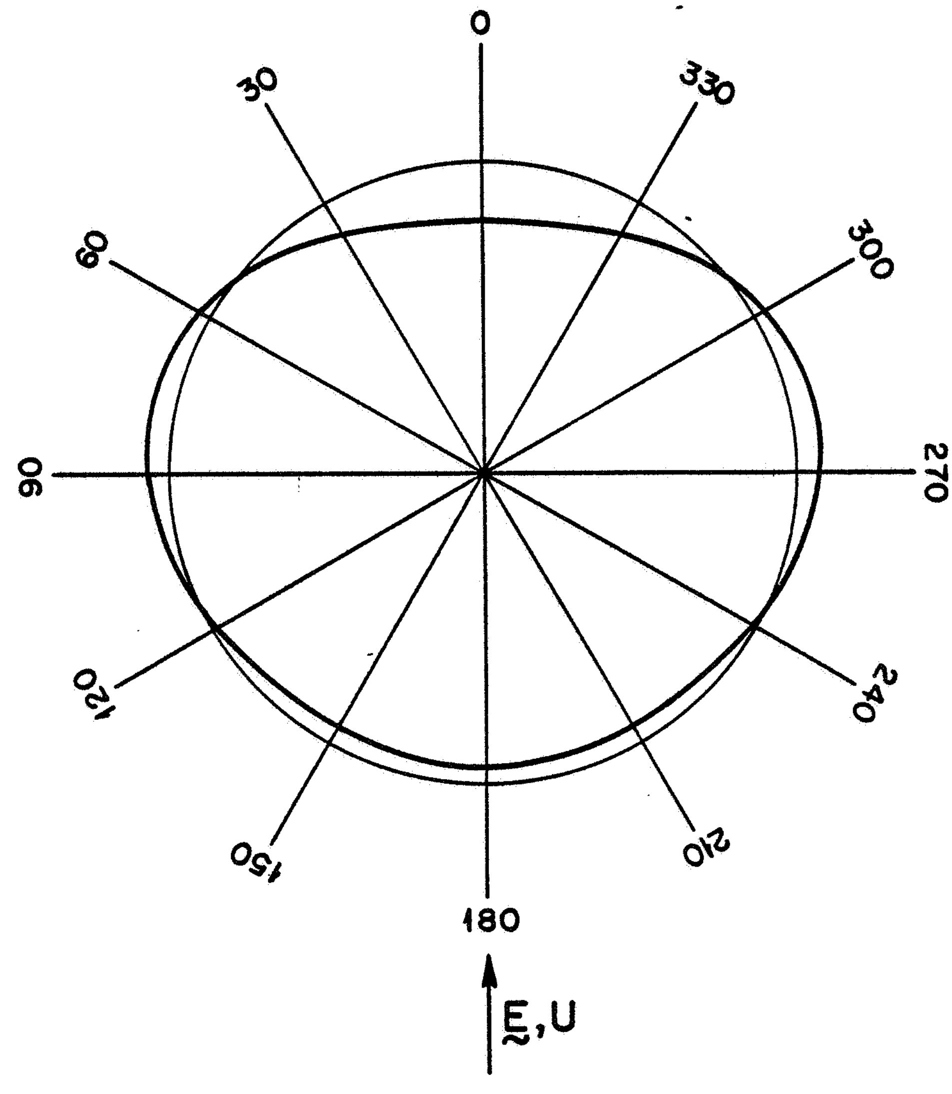
NEUTRALLY BUOYANT CASE



DROPLET SETTLING DUE TO GRAVITY

FIG. 3

Qualitative Picture of Effect of External Electric Field on Settling Velocity

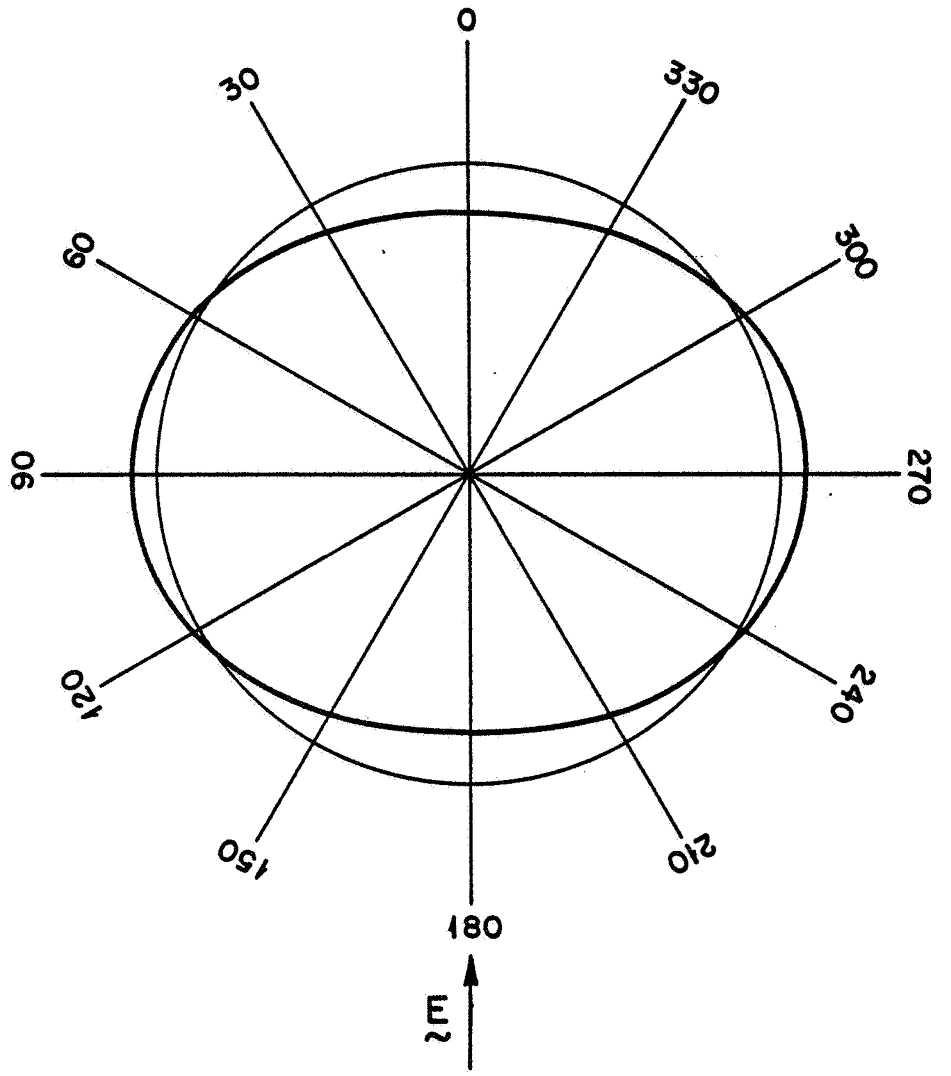


Shape of Droplet (with charge convection)

$K = 0.1$ $S = 0.25$ $R = 1.0$ $\tau_R = 1.0$ $\delta = 0.2$

$$\frac{\rho \nu U}{\gamma} = 0.4 \quad \frac{Q_0}{\epsilon E_0} = 0.0$$

FIG. 4

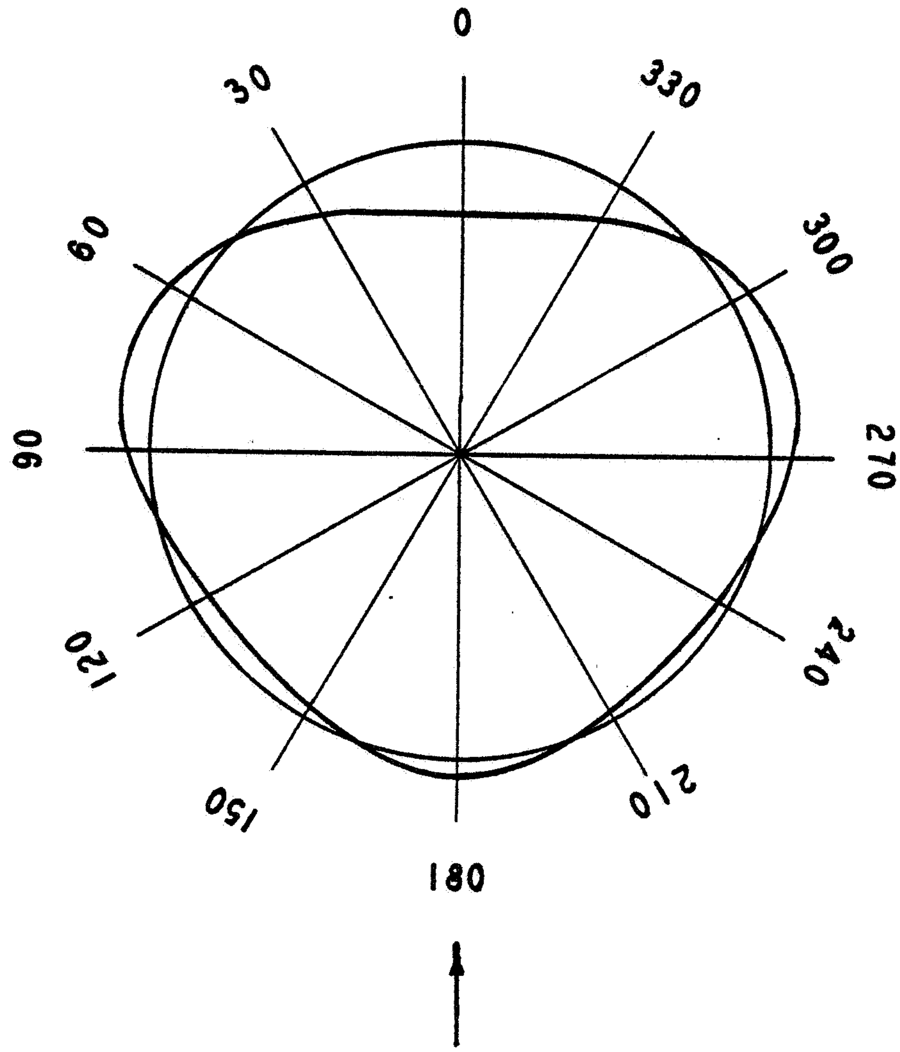


Shape of Droplet (no charge convection)

$K = 0.1 \quad S = 0.25 \quad R = 1.0 \quad \tau_R = 1.0 \quad \delta = 0.2$

$$\frac{\rho \nu U}{\gamma} = 0.4 \quad \frac{Q_0}{\epsilon E_0} = 0.0$$

FIG. 5



Shape of Droplet (with charge convection)

$K = 0.1$ $S = 0.5$ $R = 0.0$ $\tau_R = 1.0$ $\delta = 0.2$

$\frac{\rho \nu U}{\gamma} = 0.5$ $\frac{Q_0}{\epsilon E_0} = 1.0$

FIG. 6