Shape deformation of a vesicle under an axisymmetric non-uniform alternating electric field

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Abstract
We suggest that non-uniform electric fields that are commonly used to study vesicle dielectrophoresis can be employed in hitherto relatively unexplored areas of vesicle deformation (for electromechanical characterization) and electroporation. Conventionally, the tension generated in vesicles is commonly modeled to be entropic or enthalpic in origin. A comparison of the configuration of a vesicle in the enthalpic and entropic regimes as well as the cross over between the two regimes during vesicle deformation has eluded understanding. A lucid demonstration of this concept is provided by the study of vesicle deformation under axisymmetric quadrupole electric field and the shapes of the vesicles obtained using the entropic and the enthalpic approaches, show significant differences. A strong dependence of the final vesicle shapes on the ratio of electrical conductivities of the fluids inside and outside the vesicle as well as on the frequency of the applied quadrupole electric field is observed. A comparison with experimental data from the literature is also made. Moreover, an excess area dependent transition between the entropic and enthalpic regimes is observed. The method could be used to estimate electromechanical properties of the vesicle.

Keywords: vesicles, quadrupole field, entropic and enthalpic tension, deformation, electrohydrodynamics

Supplementary material for this article is available online
(Some figures may appear in colour only in the online journal)

1. Introduction
A vesicle under non-uniform, axisymmetric quadrupole electric field, not only exhibits dielectrophoresis, but can also deform. Although several experimental and theoretical papers have demonstrated vesicle deformation under AC [1–5], pulsed DC [6], DC fields [7, 8], these fields are mostly uniform. A uniform field leads to prolate and oblate spheroidal (dipolar) deformations, and these have been summarized into a phase diagram [3, 9, 10] using spherical as well as spheroidal harmonics. However, non-uniform fields have also shown promise in more efficient electroporation as compared to uniform fields [11]. Recently a systematic review about deformable particles in arbitrary shear and electric field was provided, although the specific case of quadrupolar electric field is not addressed in this work [12]. Interestingly, very few such experimental studies on deformation of a vesicle have been conducted [11, 13] in non-uniform fields whereas there is hardly any theoretical study reported. The experiments [11, 13] indicate fascinating shapes (prolate, oblate, pear, diamond, square) due to action of quadrupolar and higher order potentials and associated Maxwell stress. The resulting shape is clearly a balance of electric, hydrodynamic and membrane stress. Amongst the different membrane stresses, there is a good understanding of the bending stress as well as the non-uniform tension that arises on account of local membrane incompressibility. To describe the uniform tension, two approaches have been used.
The entropic approach, wherein, the tension arises due to the thermal undulations of the excess area. The assumption here is that under an external force, the excess area present in the thermal undulations is reduced, leading to a tension (hereafter called as the entropic tension). The membrane is then assumed to have enough excess area not to cause stretching at a molecular level and was employed to describe vesicle deformation under a uniform electric field [2, 14]. On the other hand, when a membrane is completely stretched, the uniform tension arises because the excess area of a vesicle cannot increase during the shape deformation process (we call this the enthalpic tension). This approach has been used to describe shape deformation for vesicles in shear flow [15], wherein shapes are described by the second Legendre mode, akin to deformation under a uniform electric field. On the other hand, the quadrupolar field provides two degrees of freedom for shape deformation, namely the second and fourth Legendre modes.

Seifert [16] addressed the issue of simultaneously including the entropic and enthalpic tensions for the case of a vesicle in shear flow and a semi-numerical approach is required due to coupling of various modes. In shear flows and in uniform electric field, the excess area in a vesicle is stored in the \( l = 2 \) Legendre modes. In quadrupole electric fields, the excess area could be stored in the \( l = 2, 4 \) modes, thereby making the study of distribution between these modes interesting.

These concepts form the basis for micropipette experiments which were initially proposed by Evans and Rawicz [17], wherein it was shown that for tensions lower than 0.5 mN m\(^{-1}\), the aspiration can be considered entropic and the area change is logarithmic in tension. On the other hand for higher values of tension, a membrane stretches proportional to the tension, and inverse to the area incompressibility modulus (typically of the order 100–200 mN m\(^{-1}\)).

Motivated by these issues we ask the following questions:

1. What is the vesicle deformation in pure quadrupolar field (as well as a mix of uniform and quadrupole fields), and can the prolate, oblate, pear, diamond and square shapes, seen in experiments be explained?
2. When is the deformation dominated by entropy and enthalpy?

2. Mathematical formulation

2.1. Model description

The system, considered in this work, consists of a spherical vesicle of radius \( R_o \), surrounded by a non-conducting bilayer membrane of thickness \( \delta_m \) that has an electrical conductivity \( \sigma_{m} \) and a finite permittivity \( \varepsilon_{m} \). This bilayer membrane which separates the inner fluid from the suspending medium is characterized by an interfacial tension \( \gamma_{m,0} \) and bending rigidity \( \kappa_b \). The dilatational viscosity of the membrane is assumed to be, \( \mu_{m} = 0 \). The Newtonian fluid enclosed within a vesicle has permittivity \( \varepsilon_{in} \), conductivity \( \sigma_{in} \), and viscosity \( \mu_{in} \). The suspending Newtonian medium has permittivity \( \varepsilon_{ex} \), conductivity \( \sigma_{ex} \), and viscosity \( \mu_{ex} \). Gravity effects are neglected on account of their small size (\( R_o = 5 \)–10 \( \mu \)m). We define the ratios of fluid physical properties as \( \sigma_{j} = \sigma_{m}/\sigma_{ex}, \varepsilon_{j} = \varepsilon_{m}/\varepsilon_{ex}, \mu_{t} = \mu_{m}/\mu_{ex} \). Note that subscript ‘in’ and ‘ex’ represent quantities associated with the inner and the outer fluid, respectively.

To generate a non-uniform electric field, axisymmetric quadrupole electrodes (symmetric about the Z-axis) are used in this work (figure 1). The geometric center of the axisymmetric electrode setup is the region of minimum electric field, whereas the electric field is maximum at the electrode edges. A spherical coordinate system \((r, \theta, \Phi)\) is assumed such that the origin of the coordinate system is at the geometric center of the electrode system. A time-periodic, non-uniform, axisymmetric, AC electric field is externally applied to a vesicle placed at the center of the electrode system. The applied electric potential is expressed as a sum of uniform and quadrupole electric potentials as \( \phi_{\infty} = (-E_o r P_1(cot \theta) - \Lambda_o \varepsilon_0^2 P_2(cot \theta)) \cos \omega t \), where \( E_o \) and \( \Lambda_o \) are the intensities of the uniform and the quadrupole electric fields, respectively. Here \( P_1(cot \theta) \) and \( P_2(cot \theta) \) denote Legendre polynomials of first and second degree, respectively. The electric field generated due to this applied electric potential can be expressed as \( E_\infty = -\nabla \phi_\infty \). In this work, most of the equations are expressed in their dimensional form (with no over-bar) but the results are primarily presented in non-dimensional form (represented with an over-bar) using appropriate dimensionless parameters.

2.2. Governing equations and boundary conditions

The inner and outer fluids are assumed to be leaky dielectrics. The charge convection [10] is ignored since the focus of the work is on understanding steady state, and time independent deformation of vesicles, wherein the equilibrium state is characterized by absence of flow. Moreover, the convection term scales as \( t_{m}/t_{H} \), where \( t_{ex} \sim \varepsilon_{ex}/\sigma_{ex} \) and \( t_{H} \sim \mu_{ex}/\varepsilon_{ex}E_{o}^{2} \) and is typically small for moderately conducting fluids with voltages below electroporation voltages of 1 kV cm\(^{-1}\) (see appendix). The solution of Laplace equation \( \nabla^2 \phi = 0 \) (where \( j = ex, in \)) in spherical coordinate system results in the electric potential outside \( \phi_{ex} \) and inside \( \phi_{in} \) the vesicle. The normal and tangential electric fields in the outer and inner
region are obtained from the electric potentials using the definitions $E_{ex,in} = -\frac{1}{\varepsilon_0} \frac{\partial \phi_{ex,in}}{\partial n}$ and $E_0 = -\frac{1}{\varepsilon_0} \frac{\partial \phi_0}{\partial n}$.

Using the Maxwell's stress tensor $\mathbf{T} = \varepsilon_0 (\mathbf{E} \cdot \mathbf{E})$ and $\mathbf{I}$ is the identity tensor, the normal ($\mathbf{e}_n \cdot \mathbf{T} \cdot \mathbf{e}_n$) and tangential ($\mathbf{e}_t \cdot \mathbf{T} \cdot \mathbf{e}_t$) electric stresses acting at the underformed vesicle surface, for example, can be estimated. Here $\mathbf{e}_n$ and $\mathbf{e}_t$ represents the normal and tangent unit vectors to an undeformed sphere.

The velocity and pressure fields corresponding to each fluid region (inner or outer) are given by the Stokes equation and the continuity equation ($\nabla \cdot \mathbf{u}_{in,ex} = \mu_{in,ex} \nabla^2 \mathbf{u}_{in,ex}$, $\nabla \cdot \mathbf{u}_{in,ex} = 0$). Here, the inertial effects are ignored, thereby addressing small Reynolds number conditions.

The surface of a deformed vesicle is described by

$$\mathbf{r}(\theta) = \mathbf{r}_0 + \delta \mathbf{r}_1 \phi(\theta)$$

where $r_0$ is the radial position of a slightly deformed vesicle surface from its center, $s_1, s_2, s_3, s_4$ are deformation amplitudes associated with respective Legendre modes, $s_3$ deformation mode deforms a vesicle into a prolate/oblate ellipsoids, the $s_3$ mode makes the shapes non-axisymmetric about the Z-axis, while the $s_4$ mode imparts higher order shapes (square, diamond, pear etc), the area excess over a sphere of the same volume is described by $\Delta$.

\[
\alpha = \frac{1}{3} \left( 1 - \left( \frac{2}{3} \mathbf{r}_1^2 + \frac{5}{3} \mathbf{r}_2^2 + \frac{7}{3} \mathbf{r}_3^2 + \frac{9}{3} \mathbf{r}_4^2 \right) \right) R_0^2.
\] (2)

The stress due electric field and hydrodynamic membrane flow are balanced by the resistance offered by the membrane on account of shape deformation. These include bending ($\tau^b$), uniform ($\tau^u$) and nonuniform tension ($\tau^t_{mn}$) components of the membrane stresses.

The dimensional tangential stress boundary condition is given by

\[
[[\tau^H_n]] + [[\tau^E_n]] + \mathbf{t} \cdot \nabla \mathbf{y}^n = 0
\] (4)

where the tangential hydrodynamic and electric stresses are obtained as $\mathbf{t} \cdot \nabla \mathbf{y}$ and $\tau^t_{mn} = \mathbf{t} \cdot \nabla \mathbf{y}^m$ and where $[[\cdot]]$ represents the difference in properties of outer and inner fluid across the interface. The non-dimensional normal stress balance condition is given by

\[
[[\tau^H_n]] + [[\tau^E_n]] - \kappa_{b} \left( \nabla^2 C - \frac{C}{2} \frac{C^2}{K} - 4K \right) = 0.
\] (5)

Where the normal hydrodynamic and electric stresses are obtained as $\mathbf{n} \cdot \mathbf{t}$ and the curvature is given by $C = \nabla \cdot \mathbf{n}$. $K$ here is the Gaussian curvature, $\nabla^2$ is the surface Laplace Beltrami operator. The last two terms represent the membrane uniform tension $\tau^u$ while the bending stress is given by $\kappa_{b}$. The uniform tension $\tau^u$ is estimated depending upon whether the system is in enthalpic or entropic regime, a point elaborated later in the manuscript. The non-uniform tension is calculated using the condition of surface-area conservation of lipids that yields, $\nabla \cdot \mathbf{v} + C \mathbf{v} \cdot \mathbf{n} = 0$, where $\nabla = (I - m\nabla)$ and $\mathbf{v}$ is the velocity vector.

2.3. Description in the axisymmetric case: leading order solution

A systematic regular perturbation analysis can be conducted for this problem. This is discussed later in the article and details presented in the calculations presented in this work consider the electrostatics and hydrodynamics estimated over an undeformed sphere, while the membrane forces are obtained over a deformed sphere, correct upto leading order in the shape deformation coefficients, all within the axisymmetric assumption. These assumptions are consistent with more formal regular perturbation analysis to the leading order.

2.3.1. Electrodynamics. The solution of the Laplace equation for electric potential is

\[
\phi_{ex} = \phi_{\infty} + \frac{A_1}{r^2} P_1(\cos \theta) + \frac{A_2}{r^2} P_2(\cos \theta)
\] (6)

\[
\phi_{in} = B_1 r P_1(\cos \theta) + B_2 r^2 P_2(\cos \theta)
\] (7)

where the complex coefficients $A_1, A_2, B_1, B_2$, are obtained by solving the following electrostatic boundary conditions at the membrane interface ($r = R_m$) and using the orthogonality of Legendre polynomials [2, 14].

\[
\phi_{in} - \phi_{ex} = V_{m1} P_1(\cos \theta) - V_{m2} P_2(\cos \theta) = 0
\] (8)

\[
(\sigma_{in} + i\omega \varepsilon_{in}) \frac{d\phi_{in}}{dr} - (\sigma_{ex} + i\omega \varepsilon_{ex}) \frac{d\phi_{ex}}{dr} = 0
\] (9)

\[
(\sigma_{ex} + i\omega \varepsilon_{ex}) \frac{d\phi_{ex}}{dr} + C_{m} i\omega (V_{m1} P_1(\cos \theta) + V_{m2} P_2(\cos \theta)) + G_{m}(V_{m1} P_1(\cos \theta) + V_{m2} P_2(\cos \theta)) = 0.
\] (10)

Here $V_{m1}$ and $V_{m2}$ are the transmembrane potentials across the membrane associated with the $P_1$ and $P_2$ Legendre modes, respectively. $C_{m}$ and $G_{m}$, which can be modeled as $C_{m} = \varepsilon_{m}/\delta_{m}$ and $G_{m} = \sigma_{m}/\delta_{m}$, are the membrane capacitance and conductance respectively.

The non-oscillatory part of the Maxwell stresses can be further expressed as [14, 18, 19].
\[
\tau_{r_{\text{ex,in}}}^{E} = \frac{\varepsilon \varepsilon_0}{4} (E_{r_{\text{ex,in}}} E_{r_{\text{ex,in}}}^* - E_{\theta_{\text{ex,in}}} E_{\theta_{\text{ex,in}}}^*) \quad (11)
\]

\[
\tau_{\theta_{\text{ex,in}}}^{E} = \frac{\varepsilon \varepsilon_0}{4} (E_{\theta_{\text{ex,in}}} E_{\theta_{\text{ex,in}}}^* + E_{r_{\text{ex,in}}} E_{r_{\text{ex,in}}}^*) \quad (12)
\]

where * represents complex conjugate of the respective physical quantity and \( E_r \) and \( E_{\theta} \) are the radial and tangential electric fields, respectively. The net normal and tangential electric stresses on the vesicle are \( \tau_r^{E} = \tau_{r_{\text{ex}}}^{E} - \tau_{r_{\text{in}}}^{E} \), \( \tau_{\theta}^{E} = \tau_{\theta_{\text{ex}}}^{E} - \tau_{\theta_{\text{in}}}^{E} \), respectively.

The expressions for the coefficients associated with the electric potential are provided in the online supplementary material (stacks.iop.org/JPhysCM/31/035101/mtmedia).

### 2.3.2. Hydrodynamics.

These equations are solved assuming axisymmetry and by adopting a stream function approach and the resulting pressure and velocity fields are used to estimate hydrodynamic stress (coefficients provided in the supplementary material). The stream functions for the outer and inner regions are of the generalized form

\[
\psi_{\text{ex}} = \sum (C_l^e + C_d^e) G_2 + \left( C_l^e + C_d^e \right) G_3 + \left( C_l^e + C_d^e \right) G_4 + \left( C_l^e + C_d^e \right) G_5, \quad (13)
\]

\[
\psi_{\text{in}} = \sum (C_l^i + C_d^i) G_2 + \left( C_l^i + C_d^i \right) G_3 + \left( C_l^i + C_d^i \right) G_4 + \left( C_l^i + C_d^i \right) G_5, \quad (14)
\]

where \( G_1 - G_5 \) are the Gegenbauer’s function of first kind.

\[
G_2 = \frac{1}{2} (1 - \cos^2 \theta), \quad (15)
\]

\[
G_3 = \frac{1}{2} (1 - \cos^2 \theta) \cos \theta, \quad (16)
\]

\[
G_4 = \frac{1}{8} (1 - \cos^2 \theta) (5 \cos^2 \theta - 1), \quad (17)
\]

\[
G_5 = \frac{1}{8} (1 - \cos^2 \theta) (7 \cos^2 \theta - 3) \cos \theta. \quad (18)
\]

The velocity fields can be expressed in terms of stream functions \( \psi \) as \( v_r_{\text{ex,in}} = -\frac{1}{\sin \theta} \frac{\partial \psi_{\text{ex,in}}}{\partial \theta}, \) \( v_\theta_{\text{ex,in}} = -\frac{1}{\sin \theta} \frac{\partial \psi_{\text{ex,in}}}{\partial r}, \) where \( v_r \) and \( v_\theta \) are the normal and tangential velocity components, respectively.

The pressure is governed by the solution of the Laplace equation, \( \nabla^2 P = 0. \) For the outer and inner regions it is considered to be of the form

\[
p_{\text{ex}} = \sum \frac{C_l^e}{r^2} P_l (\cos \theta) + \sum \frac{C_d^e}{r^2} P_d (\cos \theta) + \frac{C_l^e}{r} P_l (\cos \theta) + \frac{C_d^e}{r} P_d (\cos \theta),
\]

\[
p_{\text{in}} = p_0 + \sum \frac{C_l^i}{r^2} P_l (\cos \theta) + \sum \frac{C_d^i}{r^2} P_d (\cos \theta) + \sum \frac{C_l^i}{r} P_l (\cos \theta) + \sum \frac{C_d^i}{r} P_d (\cos \theta),
\]

where \( C_l - C_{12} \) and \( C_d - C_{12} \) are unknown coefficients (determined in the supplementary material).

The normal and tangential hydrodynamic stresses at the outer and inner surfaces of the membrane are given by

\[
\tau_{r_{\text{ex,in}}}^H = -p_{\text{ex,in}} + 2 \mu_{\text{ex,in}} \frac{\partial v_r_{\text{ex,in}}}{\partial r}, \quad (21)
\]

\[
\tau_{\theta_{\text{ex,in}}}^H = \mu_{\text{ex,in}} \left( \frac{1}{r} \frac{\partial v_r_{\text{ex,in}}}{\partial \theta} - \frac{v_\theta_{\text{ex,in}}}{r} + \frac{\partial v_\theta_{\text{ex,in}}}{\partial r} \right). \quad (22)
\]

### 2.3.3. Membrane mechanics.

The curvature is expressed in spherical harmonics. The nonlinear curvature and Gaussian curvature terms cancel out to leading order. The stress due membrane bending \( (\tau_b) \), due to both uniform \( (\tau_b^e) \) and non-uniform tensions \( (\tau_{b_{\text{un}}}^e, \tau_{b_{\text{un}}}^i) \) as well as due to normal and tangential interfacial membrane stresses \( (\tau_n^e, \tau_n^i) \) are given by \( [2, \quad 14] \)

\[
\tau_b = \frac{8 \mu_0}{R_0} \left( \sum_{l=2}^{4} \frac{4}{l(l+1)} (l+1) - 2) s_l P_l (\cos \theta) \right), \quad (23)
\]

\[
\tau_n^e = \gamma_u \left( \frac{2}{r^2} \sum_{l=2}^{4} \frac{4}{l(l+1)} (l+1) - 2) s_l P_l (\cos \theta) \right), \quad (24)
\]

\[
\tau_n^i = \gamma_u \sum_{l=1}^{4} \gamma_{ml} P_l (\cos \theta), \quad (25)
\]

\[
\tau_{\theta_{\text{un}}}^e = \frac{1}{R_0} \sum_{l=2}^{4} \frac{4}{l(l+1)} (l+1) - 2) s_l P_l (\cos \theta) \right), \quad (26)
\]

\[
\tau_{\theta_{\text{un}}}^i = \frac{1}{R_0} \sum_{l=2}^{4} \frac{4}{l(l+1)} (l+1) - 2) s_l P_l (\cos \theta) \right), \quad (27)
\]

\[
\tau_{\theta}^e = \frac{2b q_{\mu_{\text{ex}}}}{R_0} \frac{\partial}{\partial \theta} \left( \frac{1}{\sin \theta} \frac{\partial (v_\theta \sin \theta)}{\partial \theta} \right), \quad (28)
\]

The balance of membrane and fluid stresses and continuity of their velocity fields across the membrane interface are given by

\[
\tau_{r_{\text{ex,in}}}^H = -p_{\text{ex,in}} + 2 \mu_{\text{ex,in}} \frac{\partial v_r_{\text{ex,in}}}{\partial r}, \quad (21)
\]

\[
\tau_{\theta_{\text{ex,in}}}^H = \mu_{\text{ex,in}} \left( \frac{1}{r} \frac{\partial v_r_{\text{ex,in}}}{\partial \theta} - \frac{v_\theta_{\text{ex,in}}}{r} + \frac{\partial v_\theta_{\text{ex,in}}}{\partial r} \right). \quad (22)
\]
by expanding the following boundary conditions, using Taylor series, at the undeformed membrane surface \( r = R_o \).

(b1) Normal stress balance: \([\tau^I_{rr} + \tau^M_{rr}] = 0\).

(b2) Tangential velocity continuity: \( v_{in} = v_{ex} \).

(b3) Membrane incompressibility condition:

\[
\frac{1}{R_o} \frac{\partial (v_{ex} \sin \theta)}{\partial \theta} + \frac{2}{R_o} v_{ex} \sin \theta = 0.
\]

(b4) Tangential stress balance: \([\tau^I_{r\theta} + \tau^M_{r\theta}] = 0\).

(b5) Kinematic condition:

\[
v_{r\text{ in}} = v_{r\text{ ex}} = \sum_{i=1}^{4} \frac{dY}{dt} P_i (\cos \theta).
\]

The above boundary conditions (b1)–(b5) are solved using the orthogonality condition for Legendre polynomials. The boundary conditions are integrated at each order of the Legendre polynomials in order to get the unknown constants associated with the velocity and pressure fields for both the inner and the outer fluids (supplementary material).

3. Entropic and enthalpic tension approach for uniform tension

In the present work, the electric field induced shape deformations are estimated by two different ways of describing the uniform tension in the membrane. These are based on the two regimes for tension as discussed by Evan and Rawicz [17]. In the first case, when the induced tension is low, the tension is estimated by using the entropic theory. At low tension a membrane is in a highly fluctuating state where in the excess area of a vesicle is present in various deformation modes. Therefore an applied stress leads to a change in area that is described by the amplitudes \( s_2, s_3, s_4 \) of modes \( P_2, P_3, P_4 \) (these are too long and not provided here) due to straightening of the fluctuations (wiggles), resulting in membrane tension that is given by

\[
\gamma_{at}^{\text{ent}} = \gamma_{init, 0} \left( \frac{s_{2,3,4} \gamma_{ex}}{s_{2,3,4}} \right).
\]

Therefore, the uniform tension in the membrane because of excess area is

\[
\gamma_{at}^{\text{ent}} = \gamma_{init, 0} \left( \frac{s_{2,3,4} \gamma_{ex}}{s_{2,3,4}} \right)
\]

where \( \gamma_{init} \) is the initial tension in the membrane. Here, \( \Delta \) contains contribution from all the three modes \( (P_2, P_3 \text{ and } P_4) \).

In the second case, when the induced tension is high, a vesicle is said to be in the enthalpic regime. In this state, the deformation modes are related by the constraint of the vesicle area remaining constant during deformation, \( \Delta = 0 \), leading to

\[
\frac{4}{5} \frac{dx_2}{dt} + \frac{10}{7} \frac{dx_3}{dt} + 2x_4 \frac{dx_4}{dt} = 0.
\]

By substituting the solutions from kinematic conditions for \( dP_2, dP_3, dP_4 \) the enthalpic tension, \( \gamma_{at}^{\text{enth}} \), can be determined and is provided in supplementary material.

4. Dimensionless parameters

All length scales are non dimensionalized by the radius of vesicle \( R_o \), the potential by either \( E_0, R_o \) or \( \Lambda_o R_o \), the tension by \( \kappa_b/R_o \), and the velocity and the stress by \( \varepsilon_{ex} R_o \bar{\Lambda}^2_{as}/\mu_{ex} \) and \( \varepsilon_{ex} R_o \bar{\Lambda}^2_{as} \), respectively. A dimensionless factor \( f = E_0/(\Lambda_o R_o) \) is introduced, which compares the relative strength of the applied uniform and quadrupole electric field, such that \( f = 0 \) represents a pure quadrupole field. Another dimensionless quantity which compares the relative strength of the shape deforming electric stress and shape resisting bending stress is the capillary number \( Ca = \varepsilon_{ex} R_o \bar{\Lambda}^2_{as}/\kappa_b \). There are several time scales in the problem, the hydrodynamic time scale \( t_1 = \mu_{ex}/(\varepsilon_{ex} R_o \bar{\Lambda}^2_{as}) \), the charge relaxation time scale of outer fluid \( t_{ex} = \varepsilon_{ex}/\sigma_{ex} \), the Maxwell–Wagner relaxation time \( t_{MWO} = (2\varepsilon_{ex} + \varepsilon_{in})/(2\varepsilon_{ex} + \varepsilon_{in}) \) and the membrane charging time \( t_{cap} = C_m (\varepsilon_{ex} + \varepsilon_{in}) \), where \( C_m = C_m R_o/\varepsilon_{ex} \). Here we use \( t_{ex} \) for non-dimensionalizing the time in the electrostatics equations and \( \omega = \omega_{ex}/\sigma_{ex} \) represents the non-dimensional frequency. The hydrodynamic equation (kinematic condition) is non-dimensionalised using the slow hydrodynamic time scale \( t_{in} \).

5. Rate of shape change

The non-dimensional equations for the time evolution of the various deformation modes are given by,

\[
\frac{ds_2}{dt} = \frac{2f(-30Y + 18Y + X\bar{Y})}{9Y},
\]

\[
\frac{ds_3}{dt} = -6(6 - 18Y + X\bar{Y} + 14Y_{1a} + 90Y_{3} + 7X_{2a} \bar{Y} + 7Y(2Y_{1b} + X_{2b} \bar{Y}))) / (7Y(32 + 23\mu_e)),
\]

\[
\frac{ds_4}{dt} = -12(-10Y(6 + \gamma_{ex})/\bar{C}a + f(-8Y + X\bar{Y})) / (8Y(85 + 76\mu_e)),
\]

\[
\frac{ds_4}{dt} = -20(-12\bar{Y}(6 + \gamma_{ex})/\bar{C}a + f(-48Y + 7X\bar{Y})) / (63Y(20 + 19\mu_e)),
\]

where constant terms \( X \)’s and \( Y \)’s are parts of the normal and tangential electric stresses, respectively. These are lengthy expressions and therefore not provided in the manuscript.

6. Results and discussion

Although the results are presented in non-dimensional parameters (represented by an over-bar) it is important to mention the typical experimental parameters of relevance [13, 20]. These are mostly borrowed from the values reported in [13, 20] and [11], and have been used to determine the
range of non-dimensional parameters used in this work. An isolated spherical vesicle of \( R_s = 5 \mu m \), motivated by typical size of a biological cell, membrane conductivity \( \sigma_m = 4 \times 10^{-9} \text{ S m}^{-1} \), membrane permittivity \( \varepsilon_m = 10 \varepsilon_0 \), initial membrane tension \( \gamma_{\text{init}} = 2.75 \times 10^{-6} \text{ N m}^{-1} \), and membrane thickness \( d_m = 5 \text{ nm} \), containing a fluid of \( \sigma_f = 1.4 \times 10^{-3} \text{ S m}^{-1} \), \( \varepsilon_f = 80 \varepsilon_0 \) is assumed to be suspended in a medium of \( \sigma_c = 5 \times 10^{-3} \text{ S m}^{-1} \), \( \varepsilon_c = 80 \varepsilon_0 \). Here \( \varepsilon_0 \) is the permittivity of free space.

The axisymmetric quadrupole electrodes in experiments typically consist of two end cap electrodes maintained at a certain voltage and a ring electrode which is often ground. An AC electric field can be generated by a peak-to-peak voltage of \( V_{\text{pp}} \) applied to the end-cap (live) and the ring electrode \( (\phi_{\text{eq}} = 0 \ V_{\text{pp}}) \). An additional 10 \( V_{\text{pp}} \) potential can be superimposed ‘between’ two end cap electrodes to produce a simultaneous uniform and quadrupole field between the electrodes. The frequency can be varied from around 500 Hz to more than 10 MHz. A separation of \( L = 20 \mu m \) and \( r_o = \sqrt{2}\sigma_o \), can be maintained between the electrodes (see figure 1), where \( z_o, r_o \) are the distances of the end caps and the ring respectively, from the center of quadrupole system. These trap parameters are similar to the experimental work of Froude and Zhu [20] except while their electrode arrangement is 2D planar quadrupole, the present work deals with axisymmetric quadrupolar system. Similar to [20] the conductivity ratio \( \sigma_r = 280 \) is considered, and additionally, \( \sigma_r = 0.003 \) is also used to investigate the \( \sigma_r < 1 \) case.

A vesicle, initially positioned at the center of electrode system, deforms under applied electric field. Additionally, it undergoes dielectrophoresis for a non-zero value of the parameter \( \bar{f} \). The problem of vesicle deformation under pure quadrupole field \( (f = 0) \) is discussed in detail while vesicle deformation under simultaneous uniform and quadrupole fields \( (\text{non-zero} \ f) \) is discussed in the supplementary material. Both entropic and enthalpic tension approaches are used in the deformation studies and a variety of shapes are reported. A qualitative comparison with the vesicle shapes reported in recent experimental papers [11, 13] is also made.

6.1. Transmembrane potential

The transmembrane potential (TMP) generated by a uniform electric field is equal and opposite at the north and south poles of the vesicle. On the contrary the transmembrane potential due to the quadrupole field is symmetric about the equator and therefore identical at the north and south poles. Therefore for membranes with a finite resting potential (in the absence of field), while uniform field would lead to a difference in poration tendency at the north or south pole, a quadrupole field will cause symmetric poration at the two poles. The variation of the amplitude of the transmembrane potential, plotted in figure 2 for \( f = 1 \), shows that the TMP falls over a frequency equivalent to the reciprocal of the capacitor charging time \( (t_c^{-1} = 0.015 \text{ for } \sigma_r = 280 \text{ and } t_c^{-1} = 2.4 \times 10^{-5} \text{ for } \sigma_r = 0.003) \) for both uniform and quadrupolar parts of the applied fields. The transmembrane potential due to quadrupolar part is greater than that due to the uniform part. Increasing the membrane conductance from 0 to 0.5 reduces the transmembrane potential slightly for \( \sigma_r \gg 1 \). However, the reduction is precipitous for \( \sigma_r < 1 \). The transmembrane potential in the low frequency limit in a non-conducting membrane is a result of the charges built on the membrane to reduce the normal electric field in the outer region to zero. This potential is independent of the conductivity ratio of the two fluids for a non-conducting membrane. In a conducting membrane, the outer electric field need not be zero, since the membrane can allow current to flow through it. This leads to a drop in the transmembrane potential. A reduction in \( \sigma_r \) means a lower electrical conductivity of the inner fluid for the same conductivity of the outer fluid (used in non-dimensionalization). This leads to lower transmembrane current (ohmic) which results in a very small transmembrane potential being able to drive ohmic current through the conducting membrane when \( \sigma_r < 1 \).

6.2. Vesicle dielectrophoresis

An important question of relevance is the stability (with respect to position) of the vesicle inside the quadrupole field. Vesicles are known to undergo dielectrophoresis in non-uniform electric fields. Typically calculation of dielectrophoretic velocity has two parts, the total electrostatic force acting on the vesicle...
and the drag force on a moving vesicle. Dielectrophoresis has been studied using a rigorous Maxwell stress approach [18, 21–25] by integrating the electric stress tensor over the spherical surface of a particle in a leaky media under a slightly nonuniform electric field. On the other hand the more popular dipole moment method [26, 27] considers the force exerted by the gradient of the applied field on the polarization vector induced in a spherical particle assuming it is subjected to the electric field at the center of mass. The dipole moment method, although not exact, is more commonly used due to its applicability to arbitrary fields. On the contrary, for composite, concentric spherical systems gets complicated and the Maxwell stress method might be more straightforward, as used in this work.

The advantage of Maxwell stress method over the dipole moment method is best demonstrated in the case of liquid drops, wherein non uniform electric field has been extensively studied to understand their translation, deformation, levitation, breakup etc [28–35]. The analysis shows that the tangential electric stresses in a leaky dielectric system (both the drop and the fluid medium in which it is suspended are leaky dielectrics), leads to circulation inside the drop, as well as in the outer fluid medium. This alters the drag on the particle. Therefore to obtain the dielectrophoresis velocity, the problem has to be solved using the Maxwell stress approach. A simplified approach, where the DEP force is calculated from the dipole moment method, and the drag say the Hadamard–Rybczynski equation, can lead to erroneous results.

Similar to a drop interface, the vesicle interface, which is typically a bilayer membrane, is deformable as discussed by [36] and demonstrated in this work. There is no a priori reason to not expect this coupling in a vesicle (or a biological cell) as well, since the electric stresses at the interface can in-principle drive fluid motion in the inner and outer side of the vesicle. Thus although the scientific community working on dielectrophoresis of cells and vesicles has been using the net dielectrophoretic force calculated by the dipole moment method, and the net drag as that given by assumption of rigid body hydrodynamics, this assumption of a rigid body drag cannot be a priori assumed, and if true, should be shown rigorously.

It is therefore important to self consistently solve the electrohydrodynamics problem using the Maxwell-stress and low Re approach. A self consistent calculation, presented in the appendix on the simultaneous axisymmetric uniform and quadrupole AC electric fields yields the following results.

1. The dipole moment method and the Maxwell stress method to describe the dielectrophoresis of vesicles are indeed identical for quadrupole fields and yield the same dielectrophoretic force.
2. The correct hydrodynamics in such a case, specifically the drag on a vesicle, is found to obey the drag on a rigid
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sphere (Stokes drag) thereby vindicating the often used, but not explicitly proved, assumption, typically used in the literature.

6.3. Deformation in pure quadrupole electric field

A vesicle deforms in an applied electric field on account of the Maxwell stress, proportional to the square of electric field, acting on it. Thus while a uniform electric field \( P_1 \) produces shapes described by the \( P_2 \) mode, when a pure quadrupolar \( (P_2) \) field is applied, the Maxwell stresses and thereby the resulting shapes are expected to have the \( P_2, P_4 \) modes. At least two experimental results are reported in the literature on vesicle deformation in multipolar fields [11, 13]. Unlike uniform electric fields which typically result in prolate or oblate spheroids, higher order multipolar fields (e.g. quadrupole and octupole) lead to interesting shapes such as square, diamond and even hexagonal. We therefore understand the deformation of a vesicle in quadrupole potentials using the Maxwell stress approach.

To determine realistic parameters for quadrupolar fields that can cause deformation, one can consider around 16V\(_{pp}\) potential applied between the end cap electrodes and the ring electrodes, with \( r_o = \sqrt{2} z_o \) and \( z_o = 10 \, \mu m \). The applied potential generates an axisymmetric quadrupole electric field of strength \( \Lambda_o = -2.12 \times 10^{10} \, V \, m^{-1} \). A vesicle of size \( R_o = 5 \, \mu m \) and excess area \( \Delta = 0.2 \) can be considered to be at the center where the DEP force on the vesicle is zero on account of zero electric field, thereby preventing its translation. This allows a systematic analysis of shape deformations.
due to non-zero electric stress at the vesicle surface. In calculations presented $\kappa_b = 25K_B T$, that gives a capillary number of $Ca = \varepsilon_e R^2 \Delta \gamma_b \bar{\kappa}_b = 9684$. These numbers serve as a reference for the choice of non-dimensional parameters used in the present analysis.

Figure 3 shows the evolution of amplitude of deformation modes, $s_2$ and $s_4$, with time for very high and low $\sigma_r$ at an intermediate frequency ($\omega = 1$) in both entropic and enthalpic regimes. For $\sigma_r > 1$ both $s_2$ and $s_4$ are positive, while for $\sigma_r < 1$, $s_2$ and $s_4$ are negative. The $P_2$ deformation mode is not admitted in the pure quadrupole case ($\bar{s}_3 = 0$). Thus starting with an initial spherical shape, both the enthalpic and entropic regimes show that a final, non-spherical, equilibrium shape is reached. The deformations in the enthalpic regime are higher than those in the entropic regime. The results for the entropic case are presented for two values of $\Delta = 0.02$ and 0.2. A qualitatively similar trend is observed when $\Delta$ is increased from small values of $\Delta = 0.02$ to higher values of $\Delta = 0.2$ which could be debated to be significantly higher for $\Delta < \frac{1}{\sigma_r}$ and $\Delta > \frac{1}{\sigma_r}$, respectively. The results seem to follow the same trend as $\frac{1}{\sigma_r}$ even at high frequencies, a vesicle does show non-zero, although small, tangential and normal electric fields and stresses, on account of the capacitance of the membrane.

In the entropic regime, at low and intermediate frequencies, the variation of the shapes with frequency is qualitatively similar to that observed in the entropic case. However, unlike the entropic case, low deformation (that is nearly spherical shape) is not observed at very high frequencies, since the modes have to conserve the total area and volume simultaneously. The distribution of this area between $s_2$ and $s_4$ amplitudes of the $P_2$, $P_4$ modes with frequency. In the high frequency regime, a nearly spherical shape is seen since the Maxwell stresses become small although do not disappear. Thus unlike a drop even at high frequencies, a vesicle does show non-zero, although small, tangential and normal electric fields and stresses, on account of the capacitance of the membrane.

Similar results are obtained for the case of mixed (uniform and quadrupole) electric fields in the supplementary material.

Figure 5(a) shows the stable shapes for high and low $\sigma_r$ in the entropic regime. It should be noted that while the shapes in the pure entropic regime are obtained by solving the dynamical equations for $s_2$ and $s_4$, calculations in the enthalpic regime lead to different nature of the equations. The steady state solution to the evolution equations (35)–(37)
for $s_2$ and $s_4$ in the enthalpic regime gives multiple roots (as pairs in $s_2, s_4$) of which only one root (and thereby shape) is expected to be stable. It should be noted that each root corresponds to a different shape. To find the stable root, the eigenvalues of the linearized coefficient matrix resulting from the two non-linear differential equations for $s_2$ and $s_4$ (35)–(37) are determined for each of the roots, and the root is deemed stable if the eigenvalues are negative (and unstable if at least one of the eigen value is positive). This is also demonstrated by solving the evolution equations (35)–(37) with respect to time, with the initial shape ($s_2$ and $s_4$ values) being a perturbation around the stable or unstable roots. When the initial condition corresponds to the unstable roots, the system is seen to evolve to the stable roots (and thereby shapes). Figure 7 shows these plots for $\sigma_r > 1$ and $\sigma_r < 1$ respectively. A systematic evolution to the stable shapes is seen, with an interesting period of relative quiescence before the transition.

Figure 8. Relative contribution of all stresses (destabilizing forces: solid, stabilizing forces: dashed) acting on a vesicle from $\theta = 0 - 2\pi$ when $\sigma_r = 280$. (a), (c) Entropic approach ($\bar{\omega} = 1, Ca = 9684$); (b), (d) enthalpic approach ($\bar{\omega} = 1, Ca = 9684$); (e) enthalpic approach ($\bar{\omega} = 1000, Ca = 9684$); (f) enthalpic approach ($\bar{\omega} = 1000, Ca = 100$); (g) entropic approach ($\bar{\omega} = 0.0001, Ca = 9684$); (h) enthalpic approach ($\bar{\omega} = 0.0001, Ca = 9684$).
Figure 8 shows the contribution of different stabilizing membrane forces (bending, uniform and nonuniform tension) as well as deforming electric forces and the resulting hydrodynamic forces. Figures 8(a) and (b) show that the deforming normal electric stresses are balanced by the uniform tension in both the entropic regimes at all frequencies and enthalpic regime at intermediate and low frequencies (only $\bar{\omega} = 1$ shown in figure). At high frequencies, in the enthalpic regime though, a small but non-trivial contribution of the bending and nonuniform tension generated normal forces is observed. This is really due to the low absolute value of the normal stresses at high frequencies. The contribution of bending forces is found to increase further and equal to that due to the nonuniform tension at small capillary numbers (figures 8(e) and (f)).

The tangential electric stresses are balanced by the non uniform tension and the tangential hydrodynamic stress in both the entropic and enthalpic regimes at intermediate and low frequencies (only $\bar{\omega} = 1$ shown in figures 8(c) and (d)). At very low frequencies, the tangential electric stress tends to zero, and while in the entropic regime, the small tangential hydrodynamic stress is balanced by the non uniform tension, in the enthalpic regime, the values of tangential stress is even lower, and a balance of all the three stresses, hydrodynamic, electrical and non uniform tension is observed (figures 8(g) and (h)).

Figure 9 presents variation of entropic and enthalpic tension with apparent excess area for three different frequencies. Figure 9(a) shows that in the fluctuation dominated regime, the tension varies exponentially with the apparent excess area, as assumed in equation (32) and in agreement with the prediction of Evan and Rawicz [17]. Figure 9(b) shows that a vesicle in the enthalpic regime, exhibits a decrease in tension with an increase in the apparent excess area ($\gamma_{\text{enth}} \sim \rho^{-1/2}$ where $\rho = \Delta/(4\pi)$), for a given $Ca$ at different frequencies. The tension increases with capillary number and scales as $\gamma_{\text{enth}} \sim Ca$ (figure 9(c)). Figures 9(c) and (d) show that for a given $Ca$, the excess area dependent enthalpic tension is higher for smaller excess area.

The experimental work by Evan and Rawicz [17] indicates that when a vesicle is stretched from a spherical shape due to
application of an external force, the maximum tension could not be more than 0.5 mN m\(^{-1}\) in the low tension regime. In the high tension regime, the maximum allowable vesicle tension is known to be typically around 10 mN m\(^{-1}\), thereby limiting the capillary number. We provide scaling arguments to get such a criteria in the electrodeformation of vesicles in the conclusion section.

7. Conclusions

A systematic analysis of vesicle dielectrophoresis and deformation in non-uniform AC electric field is presented. The deformation of a vesicle in quadrupole field shows a variety of shapes such as cuboid and rhomboid, significantly different from the spheroids seen in uniform field, and these shapes depend upon the regime, entropic or enthalpic, as well as the conductivity ratio and the applied frequency.

It would be appropriate to compare the experimental results in Issadore et al [11] and Korlach et al [13] with the analysis conducted in this work while accounting for the planar (quadrupole) and 3D (octupole) fields in their set-ups respectively as against the axisymmetric quadrupole field in the present case. The squaring of shapes is clearly observed in the experiments of Issadore et al [11] for planar quadrupole. Using their experimental parameters it is seen that two shapes from the enthalpic theory presented in this work (figure 10) are identical to their experimental shapes (figures 6(c) and (e) of [11]).

Similarly as predicted in this work, intermediate frequency squaring and high frequency near spherical vesicles can be seen in figures 3(b) and (c), respectively in Korlach et al [13].

The electrical parameters as well as the quadrupole electrode design suggested in this work, should allow the method to be used for understanding electrodeformation of vesicles and biological cells in non uniform fields that are more commonly used in experiments and applications. The method shows that in the entropic regime, the vesicles admit higher order shapes, indicating influence of quadrupole field. In the enthalpic regime, the electric field as well as the frequency and the conductivity ratio determine the final shape of a vesicle with a given excess area and a high nonlinearity in the shape is observed. When a uniform electric field is employed in the enthalpic regime, the shape is prolate or oblate spheroid that satisfies the excess area constraint, and therefore an interplay of different shape modes cannot be investigated. Thus quadrupole field is the simplest axisymmetric configuration that explores the competition of \(s_2\) and \(s_4\) deformation in determining the final shape.

It should be noted that though highly non linear shapes at high capillary numbers are presented in this work, the electrostatics and hydrodynamics are solved on a sphere though. However, it should be mentioned that the membrane deformation leads to non-linear equations due to area incompressibility conditions even at linear order in deformation. Thus although the drag on the non-spherical shapes could be different, this should only lead to slower dynamics, while keeping the shapes similar to what are predicted in the present work.

A unified description of the tension incorporating simultaneously the enthalpic and the entropic effects of a vesicle in an external forcing such as shear flow or electric field, or even applied tension due to micropipettes, has eluded solution with few exceptions [16]. While the shear flow studies are typically based on enthalpic approach [37], entropic [2, 38] as well as enthalpic approach have been used for describing the tension in electric fields [2]. However, a comparison of the two approaches is lacking. Bridging the two regimes, or predicting a continuous transition from the entropic to enthalpic regimes could be a difficult task. For example even in the analysis of Seifert [16], it is difficult to predict the exact shear rate for a transition from entropic to enthalpic regime (as observed to

\[
\frac{R_o}{\sigma} = 5 \, \mu m, \sigma_{ex} = 0.1 \, S \, m^{-1}, \sigma_{ex} = 0.001 \, S \, m^{-1}, E_o = 10^5 \, V \, m^{-1}, C_m = 10^{-2} \, F \, m^{-2}. \text{ The corresponding non-dimensional parameters in our model are: for (a) } \sigma = 100, C_m = C_o R_o / \varepsilon_{ex} = 70, C_o = \varepsilon_{ex} R_o^2 E_o^2 / \kappa_b = 86.08, \bar{\omega} = \omega / \varepsilon_{ex} / \sigma_{ex} = 0.014, \text{ and for (b) on reversing fluid conductivity on each side and increasing frequency) } \sigma = 0.01, C_m = C_o R_o / \varepsilon_{ex} = 70, C_o = \varepsilon_{ex} R_o^2 E_o^2 / \kappa_b = 86.08, \bar{\omega} = 100 \text{ (here we have assumed } \kappa_b = 25 \kappa_b T \text{ and } \varepsilon_{ex} = 80 \varepsilon_o, \text{ excess area } \Delta = 0.035, \text{ due to lack of data in [11]).}
\]

\[
\begin{align*}
\bar{\lambda} & = 0.014, \\
\bar{\omega} & = 0.014, \\
\bar{\Delta} & = 0.014,
\end{align*}
\]
be around 0.5 mN m\(^{-1}\) for example in micropipette experiments [17].

Inspired by the work of Seifert [16], we propose that a possible transition criteria for the tension from the entropic to the enthalpic regime for uniform electric fields by observing that the tension due to the electric field can be expressed in a generic way as

\[
\bar{\Delta} = \Psi(\omega, \chi, \sigma) C_a \bar{\Delta}^{-1/2}.
\]  

(38)

According to Seifert [16], the area under thermal undulations (fluctuations) is related to the tension by equation (32). As suggested by Seifert [16] the effect of external field on the vesicle can be assumed to be significant when the total excess area is shared equally by the thermal undulations as well as that due to the external force experienced by the field. In the case of a vesicle in electric field, this would amount to \(\Delta_{\text{enthal}} = \Delta_{\text{th}} = \Delta/2\). Substituting the tension due to the electric field in the expressions for tension and excess area due to thermal undulations, we get the electric capillary number required for transition from entropic to enthalpic regime as

\[
C_a = \Psi(\Delta/2)^{-1/2} \gamma_{\text{init}} e^{-\frac{4\pi \bar{\Delta}}{\gamma_{\text{init}}}}. 
\]  

(39)

We demonstrate the methodology for non dimensional \(\bar{\omega} = 1\) data shown in figures 9(b) and (c). The data can be fitted as \(\gamma = (\text{mN m}\(^{-1}\)) = 1.32 \times 10^{-6} C_a \Delta^{-1/2}\). Thus, one can estimate the transition capillary number which varies from around 500 to 17 000 as the \(\Delta\) is changed from 0.01 to 0.05 (using an estimate of \(\gamma_{\text{init}} = 2.75 \times 10^{-3}\) mN m\(^{-1}\) used in this work). Thus while vesicles with small \(\Delta\) will exhibit an easy cross over to the enthalpic regime, the vesicles with large \(\Delta\) would most likely continue to be in the entropic regime.

Appendix A. Evaluation of surface convection in membrane electrohydrodynamics

The dimensional electrostatic boundary conditions are

\[
\phi_m - \phi_{\text{ex}} = V_m. 
\]  

(A.1)

\[
\varepsilon_{\text{ex}} \left( \frac{d}{dt} E_{\text{ex}} + \nabla \cdot \mathbf{u}_{\text{ex}} \right) + \sigma_{\text{ex}} E_{\text{ex}} 
\]

\[
= \varepsilon_{\text{in}} \left( \frac{d}{dt} E_{\text{in}} + \nabla \cdot \mathbf{u}_{\text{in}} \right) + \sigma_{\text{in}} E_{\text{in}},
\]

\[
(A.2)
\]

\[
\varepsilon_{\text{ex}} \left( \frac{d}{dt} E_{\text{ex}} + \nabla \cdot \mathbf{u}_{\text{ex}} \right) + \sigma_{\text{ex}} E_{\text{ex}} 
\]

\[
= C_m \left( \frac{d}{dt} V_m + \nabla \cdot \mathbf{u}_m \right) + G_m V_m.
\]

(A.3)

In the case of AC electric fields, the time dependence of the potentials and thereby the electric fields can be expected to be of the type \(e^{i\omega t}\) and higher harmonics, premultiplied by their complex amplitudes. Therefore the dimensional equation can be written as

\[
\varepsilon_{\text{ex}} (i\omega E_{\text{ex}} + \nabla \cdot \mathbf{u}_{\text{ex}}) + \sigma_{\text{ex}} E_{\text{ex}} 
\]

\[
= \varepsilon_{\text{in}} (i\omega E_{\text{in}} + \nabla \cdot \mathbf{u}_{\text{in}}) + \sigma_{\text{in}} E_{\text{in}},
\]

\[
(A.4)
\]

\[
\varepsilon_{\text{ex}} (i\omega E_{\text{ex}} + \nabla \cdot \mathbf{u}_{\text{ex}}) + \sigma_{\text{ex}} E_{\text{ex}} 
\]

\[
= C_m (i\omega V_m + \nabla \cdot \mathbf{u}_m) + G_m V_m.
\]

(A.5)

It should be noted that the above quantities, although the same notations are used for convenience, now represent the complex amplitudes of the physical quantities, potentials and electric fields. Here, the electric fields etc are now the complex amplitude of the real electric field. In general, any real quantity \(X = \frac{1}{2} (X e^{i\omega t} + X^* e^{-i\omega t})\), then \(X\) is the complex amplitude. Dividing by the conductivity of the outer fluid yields

\[
t_{\text{ex}} (i\omega E_{\text{ex}} + \nabla \cdot \mathbf{u}_{\text{ex}}) + E_{\text{ex}} 
\]

\[
= \varepsilon_{\text{ex}} t_{\text{ex}} (i\omega E_{\text{in}} + \nabla \cdot \mathbf{u}_{\text{in}}) + \sigma_{\text{ex}} E_{\text{in}},
\]

\[
(A.6)
\]

\[
t_{\text{ex}} (i\omega E_{\text{ex}} + \nabla \cdot \mathbf{u}_{\text{ex}}) + E_{\text{ex}} 
\]

\[
= C_m/t_{\text{ex}} (i\omega V_m + \nabla \cdot \mathbf{u}_m) + G_m/\sigma_{\text{ex}} V_m.
\]

(A.7)

The non-dimensionalisation can now be done using the charge relaxation time of the outer fluid \(t_{\text{ex}} = \varepsilon_{\text{ex}}/\sigma_{\text{ex}}\), with velocity scale being \(R_o/\eta_t\), where \(\eta_t = \mu_{\text{ex}}/\varepsilon_{\text{ex}} E_o^2\), one gets the equations

\[
\left( i\omega E_{\text{ex}} + \frac{t_{\text{ex}}}{\eta_t} \nabla \cdot \mathbf{u}_{\text{ex}} \right) + E_{\text{ex}} 
\]

\[
= \varepsilon_{\text{ex}} \left( i\omega E_{\text{in}} + \frac{t_{\text{ex}}}{\eta_t} \nabla \cdot \mathbf{u}_{\text{in}} \right) + \sigma_{\text{ex}} E_{\text{in}},
\]

\[
(A.8)
\]

\[
\left( i\omega E_{\text{ex}} + \frac{t_{\text{ex}}}{\eta_t} \nabla \cdot \mathbf{u}_{\text{ex}} \right) + E_{\text{ex}} 
\]

\[
= \bar{C}_m \left( i\omega V_m + \frac{t_{\text{ex}}}{\eta_t} \nabla \cdot \mathbf{u}_m \right) + \bar{G}_m V_m,
\]

(A.9)

where in the above equation, now the quantities represent non-dimensional quantities such that \(\omega = \omega t_{\text{ex}}\), with potentials and electric fields non-dimensionalised by \(E_o R\) and \(E_o\) respectively. For typical dielectrophoresis and electro-deformation parameters, the conductivities are in the range of \(\sigma_{\text{ex}} = 10^{-2} \text{ S m}^{-1}\), for \(E_o = 0.1\ \text{kV cm}^{-1}\), leads to \(t_{\text{ex}} = 0.1\ \mu\text{s}\), \(\eta_t = 14\ \text{ms}\). For this parameter range, with the non-dimensional membrane capacitance of the order 50–125, the current due to charge convection can be conveniently ignored as compared to the displacement and ohmic currents of both the bulk fluids and the membrane.

Appendix B. Asymptotic analysis and scaling

The governing equations for electrostatics in the Stokes limit are linear. Therefore the solution to these equations are expressed in the Gegenbauer functions and Legendre
polynomials respectively. The scaling for different quantities are therefore determined by the boundary conditions.

B.1. Enthalpic regime

In the analysis carried out in this work, \( \delta_t \sim \Delta^{-1/2} = \delta \) is used as a small parameter for perturbation expansion of the membrane shape and thereby the membrane force. In the enthalpic limit, the excess area of the membrane is conserved, irrespective of the capillary number. Therefore, the electrostatics and hydrodynamics are assumed to be \( O(1) \) and are expressed as an asymptotic series as

\[
\phi = \phi_0 + \delta \phi_1 + \cdots \tag{B.1}
\]

\[
v = v_0 + \delta v_1 + \cdots \tag{B.2}
\]

This indicates that the Maxwell and hydrodynamic stresses also scale as \( O(1) \). The boundary conditions should now be visited for a consistent analysis. The velocity continuity conditions are consistently satisfied. The tangential stress boundary condition suggests

\[
\gamma^{nu} = \delta \gamma^{nu} + \delta^2 \gamma^{nu}_{1} \tag{B.3}
\]

\[
\gamma^{mu} = \gamma^{mu}_0 + \delta \gamma^{mu}_1. \tag{B.4}
\]

Since the non-dimensional curvature scales as

\[
C = 2 + \delta C_1 + \delta^2 C_2 + \cdots, \tag{B.5}
\]

a balance of the viscous and elastic stresses with the normal stresses suggests a scaling of

\[
p = \frac{1}{\delta} p_0 + p_1 + \delta p_2 + \cdots \tag{B.6}
\]

\[
\gamma^u = \frac{1}{\delta} \gamma^u_0 + \gamma^u_1 + \delta \gamma^u_2 + \cdots \tag{B.7}
\]

\[
\kappa_b = \frac{1}{\delta} \kappa_{b0}. \tag{B.8}
\]

With the above scalings, one gets the leading order solution such that the electrostatics and hydrodynamics are calculated over an undeformed sphere, and the membrane forces are calculated over the perturbed interface, i.e. up to curvature \( C_1 \). The electrostatics and the hydrodynamics are estimated on an undeformed sphere and balance the membrane forces generated by the deformed sphere.

Appendix C. Dielectrophoretic force and translational velocity of a vesicle in quadrupole field

A vesicle subjected to a non-uniform AC electric field, experiences a DEP force due to up-down asymmetric electric stresses. The electric traction acting on a unit area of a vesicle in the \( z \)-direction can be written as \( F_{\text{DEP}} = \frac{T_z}{2} \sin \theta - \frac{T_\theta}{2} \cos \theta \). The total \( z \)-directional DEP force is determined by integrating the electric traction over the vesicle surface as

\[
F_{\text{DEP}} = \int_0^{2\pi} \int_0^{\pi} F_{\text{DEP}}^E R_0^2 \sin \theta d\theta d\phi = 2\pi \varepsilon_0 R_0^2 \text{Re}[\text{CMF}] \nabla E_{\infty}^2. \tag{C.1}
\]

Equation (C.1) represents two approaches to estimate the DEP force. The integral expression is obtained from a more fundamental approach of Maxwell stress as employed in this work. The right hand side is the classical expression for DEP force obtained by the dipole moment approach. Here Re[CMF] is the real part of Clausius–Mossotti factor which depends upon the frequency of the applied electric field as well as on the electric properties of the particle and the suspending media [26]. A positive value of Re[CMF] implies that a vesicle experiences positive DEP force and moves towards region of high electric field whereas negative Re[CMF] leads to negative DEP which moves a vesicle to a region of low electric field.

Classical theory [26] based on dipole moments suggests positive or negative DEP (movement towards region of higher
or lower electric fields respectively) if \( \text{Re}[\text{CMF}] \) is positive or negative respectively. In this work, \( \text{Re}[\text{CMF}] \) is calculated by equating the classical expression for dielectrophoretic force to the dielectrophoretic force calculated by the Maxwell stress approach. A cross over frequency for transition from negative to positive DEP is obtained by setting \( \text{Re}[\text{CMF}] = 0 \), which gives a lower cross over (LCO) frequency

\[
\tilde{\omega}_{\text{LCO}} = \frac{2\sqrt{\sigma_r}}{\sqrt{3\sqrt{C_m(2\sigma_r + C_m(2 + \sigma_r))}}}
\]  

(C.2)

Figure C1. \( \text{Re}(\text{CMF}) \) versus frequency plot. (a) Role of permittivity ratio when \( \sigma_r = 280 \) (solid) and \( \sigma_r = 0.003 \) (dashed) \((C_m = 125, G_m = 0)\), \( \bar{\epsilon}_r = 0.1 \), \( \bar{\epsilon}_r = 1 \), \( \bar{\epsilon}_r = 10 \). (b) Role of membrane conductance \((C_m = 125, \bar{\epsilon}_r = 1, G_m = 0 \) (solid), \( G_m = 0.5 \) (dashed)), \( \sigma_r = 280 \), \( \sigma_r = 0.003 \).

Figure C2. DEP velocity versus frequency. (a) Role of low permittivity ratio when \( \sigma_r = 280 \) (black) and \( \sigma_r = 0.003 \) (red) \((\bar{\epsilon}_r = 0.1 \) (dashed), \( \bar{\epsilon}_r = 1 \) (solid), \( C_m = 125, f = 0.5, G_m = 0 \)). (b) Role of high permittivity ratio when \( \sigma_r = 280 \) (black) and \( \sigma_r = 0.003 \) (red) \((\bar{\epsilon}_r = 10 \) (dashed), \( \bar{\epsilon}_r = 1 \) (solid), \( C_m = 125, f = 0.5, G_m = 0 \)). (c) Role of membrane conductance when \( \sigma_r = 280 \) (black) and \( \sigma_r = 0.003 \) (red) \((C_m = 125, \bar{\epsilon}_r = 1, f = 0.5, G_m = 0 \) (solid), \( G_m = 0.5 \) (dashed)).
valid for $\sigma_r > 1$ (expression for $\sigma_r < 1$ is complicated and not provided here) whereas the upper cross over (UCO) frequency for transition from positive to negative DEP is given by

$$\omega_{UCO} = \frac{\sqrt{3} \sqrt{C_m(2\varepsilon_r + C_m(\varepsilon_r - \sigma_r)(2 + \sigma_r) + 2C_m(\varepsilon_r - \sigma_r)(2\varepsilon_r + \sigma_r))}}{\sqrt{(C_m(-1 + \varepsilon_r) - \varepsilon_r)(2\varepsilon_r + C_m(2 + \varepsilon_r))^2}}.$$

(C.3)

The expression for the CMF (as provided by Jones [26]) is identical to that obtained in the present work by the Maxwell stress approach [39], although some discrepancy in the reported values of the CMF in literature is noticed [20, 26].

The solution of kinematic condition, on integration with respect to the first Legendre polynomial, over a vesicle surface, gives steady dielectrophoretic velocity of the vesicle.

$$U_{DEP} = \frac{d\vec{s}}{dt} = \frac{\int_0^{\pi} \vec{v}_p \mu P_1(\cos \theta) \sin \theta d\theta}{\int_0^{\pi} P_1(\cos \theta)^2 \sin \theta d\theta} = \frac{2R_o(X_1Y + 6R_o(-5Y_0 + 3Y_2)\varepsilon_{ex}\Lambda_{ex})}{9Y\mu_{ex}}.$$

(C.4)

(C.5)

Substitution of electric stress coefficients (all $X$'s and $Y$'s) and non-dimensionalization of equation (C.5) shows that the DEP velocity varies linearly with $f$ (a dimensionless factor described below). The DEP velocity shows dependency on conductivity as well as permittivity ratio along with the membrane properties $C_m, G_m$. 

Figure C3. Effect of viscosity ratio on the velocity profile for a spherical vesicle undergoing dielectrophoresis (a) $\mu_r = 0.001$, (b) $\mu_r = 1$, (c) $\mu_r = 1000$ ($\bar{\omega} = 1, f = 0.1$).

Figure C4. Dependency of non-uniform membrane tension on Boussinesq number ($C_m = 125, G_m = 0, \varepsilon_r = 1, \sigma_{ex} = 280, Ca = 71 033, f = 0.5, \bar{\omega} = 1$).
The expression for $\text{Re}[\text{CMF}]$ indicates that in the low frequency regime, a vesicle with a non-conducting membrane shows a negative value essentially due to a vesicle acting as a dielectric drop in a conducting fluid. In the intermediate frequency regime, the vesicle acts as a leaky dielectric drop when $\epsilon_1^{-1} \leq \omega < \epsilon_1^{-1}$ or $\epsilon_1^{-1}$ (which is $=1$), whichever greater. 

In this range, when $\sigma_\tau < 1$, that is outer medium more conducting, a drop shows negative dielectrophoresis, whereas for $\sigma_\tau > 1$ positive dielectrophoresis is seen. At very high frequencies, the membrane is uncharged, and the vesicle behaves as a perfect dielectric inner fluid in a perfect dielectric outer fluid. The response is then governed by $\epsilon_r$. When the outer medium has more permittivity, it shows negative dielectrophoresis, whereas when the inner fluid has more permittivity, positive dielectrophoresis can be seen. These features are demonstrated in figure C1.

Figure C1(a) shows the effect of dielectric constant ratio $\epsilon_r$, on $\text{Re}[\text{CMF}]$. It is seen that for $\sigma_\tau > 1$ there is an upper and a lower cross over frequency for $\epsilon_r < 1$. However, for $\epsilon_r > 1$ only lower cross over frequency is observed. This shows that a vesicle undergoes a transition from negative to positive DEP at low frequencies, whereas at very high frequencies the $\text{Re}[\text{CMF}]$ and thereby the dielectrophoretic velocity tend to zero (figure C2(a)). While LCO frequency is independent of $\epsilon_r$ when $\sigma_\tau > 1$, and accurately predicted by equation (C.2) the LCO frequency has $\epsilon_r$ dependency for $\sigma_\tau < 1$. The UCO frequency has $\epsilon_r$ dependency for all values of $\sigma_\tau$ and this is predicted well by equation (C.3). When $\sigma_\tau < 1$ (figure C1(a)) there is no upper and lower cross over frequency for $\epsilon_r \leq 1$ and the vesicle always shows negative DEP in the entire frequency range (figure C2(a)). For $\epsilon_r > 1$ there is a LCO frequency which results in negative to positive DEP with an increase in frequency.

Figure C1(b) shows the effect of membrane conductivity on the $\text{Re}[\text{CMF}]$. It indicates that the membrane conductivity predominantly affects the low frequency behavior. For sufficiently high membrane conductance, a vesicle can behave like a drop in DC field in the low frequency regime, thereby exhibiting negative dielectrophoresis for $\sigma_\tau < 1$ and positive dielectrophoresis for $\sigma_\tau > 1$.

It is important to self consistently solve the electrohydrodynamics problem using the Maxwell stress and low Re approach, even for an undeformed sphere, if one is interested in knowing the dielectrophoretic velocity of a deformable particle such as a vesicle. This is essentially due to the coupling between the non-uniform tensions associated with different Legendre modes. Thus, for an undeformed sphere, the $P_2$ and $P_4$ electric stresses generated by the electric fields, need to be satisfied for an undeformed sphere. Although the normal stresses associated with the non-uniform tensions are decoupled, the non-uniform tensions get coupled in the tangential stress balance. In an undeformed sphere, the electric stresses are balanced by the non-uniform tension, leading to fluid flow associated with the third to fifth Gegenbauer streamfunctions (these flows eventually lead to deformation which is discussed next). Although these do not contribute to the dielectrophoretic velocity, they give rise to fluid flow, which has a dependence on the viscosity ratio (figure C3). The non-uniform tension associated with the translational mode $\gamma_{mn}$, leads to a rigid body like motion. Therefore the drag resisting the dielectrophoretic motion is the same as a rigid sphere ($6\pi\mu U \sigma_\tau$), and is independent of the viscosity ratio, although it can be calculated only after solving the coupled problem. Therefore the dielectrophoretic velocities calculated by the Maxwell stress approach in this work are in agreement with that calculated by the dipole moment method with a ‘rigid body’ drag (equation (C.5)). Despite the dielectrophoretic velocity showing a rigid body like drag, the velocity streamlines are quite different than that caused by translation of a rigid spherical particle, and show multiple rolls associated with the $P_2$ and $P_4$ electric stresses associated with the applied field (figure C3). Similarly for a membrane with surface viscosity, the membrane viscous stress is absorbed by the non-uniform tension such that the dielectrophoretic velocity remains independent of the Boussinesq number too. The non-uniform tension though depends linearly on the Boussinesq number (figure C4).

These results can be presented in a phase diagram (figure C5) which shows a negative dielectrophoretic region at low frequencies for all conductivity ratios (due to vesicle acting as a dielectric drop in a perfect conductor) and at all frequencies at low conductivity ratios (due to negative dielectrophoresis in the intermediate frequency range (since the polarization vector is opposite to the applied field)). At moderate and high conductivity ratios, positive dielectrophoresis is seen in an intermediate range of frequencies, essentially in agreement with liquid drops of same conductivity ratio (having polarization vector in the direction of the applied field). The above discussion indicates that the dipole moment method with a rigid particle Stokes drag assumption should suffice to describe the dielectrophoretic motion of a vesicle in non-uniform electric field. The experimental results in [20] are in qualitative agreement with the present model, although quantitative studies cannot be done due to the axisymmetric electrode configuration considered in this work. It should be mentioned here that experiments are typically conducted in pure quadrupole field. In such
a field, no dielectrophoresis will be seen for a vesicle kept exactly at the center of the electrode system. However, an off-center vesicle, say in the $z$ direction will experience a uniform field given by $E_z = -2\Lambda\varepsilon_0\omega$ and a quadrupole potential of $\Lambda\omega$, thereby enabling the use of the results derived in this work.

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