

Biophysical Journal, Volume 96

**Supporting Material**

**Electrohydrodynamic model of vesicle deformation in alternating electric fields**

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# Electrohydrodynamic model of vesicle deformation in AC electric fields

## Supplementary material

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### 1 Solution of the hydrodynamic problem

Here we outline the leading-order solution for the velocity field and hydrodynamic stresses. In this case, all quantities are evaluated on a sphere. More details can be found in Refs. (1–4).

Velocity fields are described using basis sets of fundamental solutions of the Stokes equations appropriate for spherical geometry (5),  $\mathbf{u}_{jmq}^{\pm}$ , defined in Section 3:

$$\mathbf{v}^{\text{ex}}(\mathbf{r}) = \sum_{jmq} c_{jmq} \mathbf{u}_{jmq}^{-}(\mathbf{r}), \quad \mathbf{v}^{\text{in}}(\mathbf{r}) = \sum_{jmq} c_{jmq} \mathbf{u}_{jmq}^{+}(\mathbf{r}). \quad (1)$$

$$\sum_{jmq} \equiv \sum_{j=2}^{\infty} \sum_{m=-j}^j \sum_{q=0}^2 \quad (2)$$

The velocity field contains only  $q = 0, 2$  due to the axial symmetry. The local area conservation implies that the velocity field at the interface is solenoidal (1)

$$\nabla_s \cdot \mathbf{v} = 0. \quad (3)$$

Therefore the amplitudes of the velocity field Eq. 1 are related

$$c_{jm0} = \frac{2}{\sqrt{j(j+1)}} c_{jm2}. \quad (4)$$

The component of velocity that is normal to the interface,  $c_{jm2}$ , is determined using the stress balance (Eq. 18 in the manuscript text), which in terms of spherical harmonics reads

$$\delta_{j2} \delta_{m0} \tau_{jmq}^{\text{el}} + \tau_{jmq}^{\text{hd,ex}} - \chi \tau_{jmq}^{\text{hd,in}} = \tau_{jmq}^{\text{mm}}. \quad (5)$$

Tangential stresses correspond to the  $q = 0$  component, and the normal stresses - to  $q = 2$ .  $\delta_{ij}$  is the Kronecker delta function. The hydrodynamic tractions are given by Eq. 30–Eq. 33. The electrical tractions are given by (Eq. 31 in the manuscript text), which is recast in the form

$$\boldsymbol{\tau}^{\text{el}} = 8\sqrt{\frac{\pi}{5}} \tau_r^{\text{el}} \mathbf{y}_{202}(\theta, \phi) - 2\sqrt{\frac{2\pi}{15}} \tau_{\theta}^{\text{el}} \mathbf{y}_{200}(\theta, \phi). \quad (6)$$

The membrane tractions are (1, 2)

$$\tau_{jmq}^{\text{mm}} = Ca^{-1}(\tau_{jmq}^{\kappa} + \tau_{jmq}^{\sigma}) + \chi_s \tau_{jmq}^s. \quad (7)$$

where  $Ca$  is the capillary number and  $\chi_s = \eta_{\text{mm}}/\eta a$  is a surface viscosity parameter. We have included the membrane viscous stresses for the sake of completeness. The surface viscosity of lipid bilayers in the fluid phase is relatively low,  $\eta_{\text{mm}} \sim 10^{-9} \text{Ns/m}$ , and its effects are usually negligible. Surface viscous effects become important in bilayers assembled from polymers (polymersomes), where the membrane viscosity  $\eta_{\text{mm}} \sim 10^{-6} \text{Ns/m}$  (6, 7).

The bending contribution to the membrane traction is

$$\tau_{jm2}^{\kappa} = j(j+1)(j-1)(j+2) f_{jm}, \quad \tau_{jm0}^{\kappa} = 0, \quad (8)$$

the stresses due to membrane tension are

$$\tau_{jm2}^\sigma = 2\sigma_{jm} + \sigma_0(j-1)(j+2)f_{jm}, \quad \tau_{jm0}^\sigma = -\sqrt{j(j+1)}\sigma_{jm}, \quad (9)$$

and the surface viscous stresses have only in-plane shearing component

$$\tau_{jm2}^s = 0, \quad \tau_{jm0}^s = 2(j-1)(j+2)[j(j+1)]^{-1/2}f_{jm}. \quad (10)$$

The non-uniform part of the membrane tension,  $\sigma_{jm}$ , is determined from the tangential component of the stress balance Eq. 5,  $q = 0$ ,

$$\sigma_{jm} = Ca \left[ \frac{\tau_{jm0}^{\text{el}}}{\sqrt{j(j+1)}} + c_{jm2} \frac{2+j+(j-1)(\chi+2(j+2)\chi_s)}{j(j+1)} \right]. \quad (11)$$

It is then substituted into the normal component of the stress balance Eq. 5,  $q = 2$ , to obtain the normal velocity  $c_{jm2}$

$$c_{jm2} = C_{jm} + Ca^{-1}(\Gamma_1 + \sigma_0\Gamma_2)f_{jm}, \quad (12)$$

where

$$C_{jm} = -\frac{\sqrt{j(j+1)}}{d(\chi, \chi_s, j)} \left[ 2\tau_{jm0}^{\text{el}} + \sqrt{j(j+1)}\tau_{jm2}^{\text{el}} \right], \quad (13)$$

$$\Gamma_1 = -(j+2)(j-1)[j(j+1)]^2 d(\chi, \chi_s, j)^{-1}, \quad (14)$$

$$\Gamma_2 = -(j+2)(j-1)j(j+1)d(\chi, \chi_s, j)^{-1}, \quad (15)$$

and

$$d(\chi, \chi_s, j) = (4 + 3j^2 + 2j^3) + (-5 + 3j^2 + 2j^3)\chi + 4(-2 + j + j^2)\chi_s. \quad (16)$$

Finally, the motion of the interface is determined from the kinematic condition (Eq. 17 in the manuscript text)

$$\frac{\partial f_{jm}}{\partial t} = c_{jm2} \quad \text{at } r = 1. \quad (17)$$

Substituting  $c_{jm2}$  in Eq. 17 yields the evolution equation for the shape parameters (Eq. 37 in the manuscript text).

The normal velocity Eq. 12 and the shape evolution Eq. 17 include the yet unknown isotropic membrane tension. It is expressed in terms of the shape modes and other known parameters in the problem using the area constraint (2)

$$\sigma_0 = -\frac{\sum_{jm} a(j) \left[ C_{jm} f_{jm}^* + Ca^{-1}\Gamma_1 f_{jm} f_{jm}^* \right]}{Ca^{-1} \sum_{jm} a(j)\Gamma_2 f_{jm} f_{jm}^*}. \quad (18)$$

The complicated dependence of the tension on the shape modes makes the shape evolution equations nonlinear.

In order to clarify the physical significance of the isotropic tension, let us consider the particular case when only the ellipsoidal deformation modes,  $j = 2$ , are present. Eq. 18 simplifies to

$$\sigma_0(t) = -6 + CaC_{20} \frac{32 + 23\chi + 16\chi_s}{12} f_{20}(t) \quad (19)$$

where we have emphasized that the time dependent shape modes give rise to time-dependent membrane tension. We see that the tension varies with deformation. At rest, the tension of a quasi-spherical vesicle is negative (8) and increases with forcing. Once all excess area is transferred to the  $f_{20}$  mode, the tension increases with the field strength  $Ca$  as

$$\sigma_0 \approx CaC_{20} \frac{(32 + 23\chi + 16\chi_s)\sqrt{2}}{12} \Delta^{-1/2} \quad (20)$$

Similar behavior is observed with vesicles in shear flow (1).

## 2 Spherical harmonics

The normalized spherical scalar harmonics are defined as (9)

$$Y_{jm}(\theta, \varphi) = \left[ \frac{2j+1}{4\pi} \frac{(j-m)!}{(j+m)!} \right]^{\frac{1}{2}} (-1)^m P_j^m(\cos \theta) e^{im\varphi}, \quad (21)$$

where  $\hat{\mathbf{r}} = \mathbf{r}/r$ ,  $(r, \theta, \varphi)$  are the spherical coordinates, and  $P_j^m(\cos \theta)$  are the Legendre polynomials. For example

$$Y_{10} = \sqrt{\frac{3}{4\pi}} \cos \theta. \quad (22)$$

The vector spherical harmonics relevant to our study are defined as (10)

$$\mathbf{y}_{jm0} = [j(j+1)]^{-\frac{1}{2}} r \nabla_{\Omega} Y_{jm}, \quad \mathbf{y}_{jm2} = \hat{\mathbf{r}} Y_{jm} \quad (23)$$

where  $\nabla_{\Omega}$  denotes the angular part of the gradient operator. For example

$$\mathbf{y}_{200} = -\sqrt{\frac{15}{32\pi}} \sin(2\theta) \hat{\boldsymbol{\theta}}, \quad \mathbf{y}_{202} = \frac{1}{8} \sqrt{\frac{5}{\pi}} [1 + 3 \cos(2\theta)] \hat{\mathbf{r}} \quad (24)$$

## 3 Fundamental set of velocity fields

Following the definitions given in Blawdziewicz *et al.*(10), we list the expressions for the functions  $\mathbf{u}_{jmq}^{\pm}(r, \theta, \varphi)$ . The velocity field outside the vesicle is described by

$$\mathbf{u}_{jm0}^{-} = \frac{1}{2} r^{-j} (2 - j + jr^{-2}) \mathbf{y}_{jm0} + \frac{1}{2} r^{-j} [j(j+1)]^{1/2} (1 - r^{-2}) \mathbf{y}_{jm2}, \quad (25)$$

$$\mathbf{u}_{jm2}^{-} = \frac{1}{2} r^{-j} (2 - j) \left( \frac{j}{1+j} \right)^{1/2} (1 - r^{-2}) \mathbf{y}_{jm0} + \frac{1}{2} r^{-j} (j + (2 - j)r^{-2}) \mathbf{y}_{jm2}. \quad (26)$$

The velocity field inside the vesicle is described by

$$\mathbf{u}_{jm0}^{+} = \frac{1}{2} r^{j-1} (-(j+1) + (j+3)r^2) \mathbf{y}_{jm0} - \frac{1}{2} r^{j-1} [j(j+1)]^{1/2} (1 - r^2) \mathbf{y}_{jm2}, \quad (27)$$

$$\mathbf{u}_{jm2}^{+} = \frac{1}{2} r^{j-1} (3 + j) \left( \frac{j+1}{j} \right)^{1/2} (1 - r^2) \mathbf{y}_{jm0} + \frac{1}{2} r^{j-1} (j + 3 - (j+1)r^2) \mathbf{y}_{jm2}. \quad (28)$$

On a sphere  $r = 1$  these velocity fields reduce to the vector spherical harmonics defined by Eq. 23

$$\mathbf{u}_{jmq}^{\pm} = \mathbf{y}_{jmq}. \quad (29)$$

Hence,  $\mathbf{u}_{jm0}^{\pm}$  is tangential, and  $\mathbf{u}_{jm2}^{\pm}$  is normal to a sphere. In addition,  $\mathbf{u}_{jm0}^{\pm}$  defines an irrotational velocity field.

The hydrodynamic tractions associated with the velocity fields Eq. 1 are (2)

$$\tau_{jm0}^{\text{hd,in}} = (2j+1)c_{jm0} - 3 \left( \frac{j+1}{j} \right)^{\frac{1}{2}} c_{jm2} \quad (30)$$

$$\tau_{jm2}^{\text{hd,ex}} = -(2j+1)c_{jm0} + 3 \left( \frac{j}{j+1} \right)^{\frac{1}{2}} c_{jm2} \quad (31)$$

$$\tau_{jm0}^{\text{hd,ex}} = 3 \left( \frac{j}{j+1} \right)^{\frac{1}{2}} c_{jm0} - \frac{4+3j+2j^2}{j+1} c_{jm2} \quad (32)$$

$$\tau_{jm2}^{\text{hd,in}} = -3 \left( \frac{j+1}{j} \right)^{\frac{1}{2}} c_{jm0} + \frac{3+j+2j^2}{j} c_{jm2} \quad (33)$$

## 4 Deformation of a prolate vesicle in strong fields

Consider an initially non-spherical, non-fluctuating vesicle. This situation can occur in strong electric fields, where the vesicle is already maximally deformed and then the field direction is changed. The evolution to the new stationary shape is no longer described by Eq.42 in the main text because the tension is no longer in the entropic regime. The effective tension has to be determined self-consistently along with the field-induced changes in shape to keep the total area constant (2), as discussed in Section 1 of the Supplementary material, see Eq. 19. The leading order vesicle electrohydrodynamics becomes non-linear in contrast to the corresponding results for drops and capsules (11–13). This feature of non-equilibrium vesicle dynamics has been noted by several authors in relation to vesicle dynamics in shear flow (2, 4, 14).

Vesicle deformation is approximated by

$$\frac{\partial f_{20}}{\partial t} = C^{\text{el}}(1 - 2\Delta^{-1}f_{20}^2) \quad \frac{\partial f_{2m}}{\partial t} = -2C^{\text{el}}\Delta^{-1}f_{20}f_{2m} \quad (34)$$

where the dot denotes time derivative. The modes  $f_{2m}$  are slaved to the  $f_{20}$ , which is forced to change by the electric field. Eq. 34 can be integrated to yield

$$f_{20}(t) = \delta \tanh \left[ \frac{C^{\text{el}}}{\delta} t + \tanh^{-1} \left( \frac{f_{20}(0)}{\delta} \right) \right]. \quad (35)$$

This equation shows that the maximum possible deformation is

$$\delta = \sqrt{\frac{\Delta}{2}}. \quad (36)$$

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