## Deformation of Lipid Bilayer Spheres by Electric Fields \*

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In two previous publications <sup>1, 2</sup> we have proposed an elastic theory for simple lipid bilayers which may be viewed as two-dimensional fluids. In particular, we have shown that bilayer spheres in an aqueous medium can be deformed into ellipsoidal bodies if they are submitted to a magnetic field or excess outside pressure. Here we consider a possible deformation by electric fields. It will be seen that the electric effect can be quite strong for large vesicles.

As before, we restrict ourselves to small deformations, assuming the bilayer to be unstretchable and the sphere to become an ellipsoid of revolution. To calculate the ellipticity we minimize the total energy consisting of curvature-elastic and electric parts. The conductivity of the bilayer will in general be very much smaller than that of the aqueous environment, so it seems permissible to treat the membrane as a perfect insulator.

The electric energy of deformation may be obtained from the Maxwell stresses. Those inside the membrane will be balanced by equal but opposite elastic stresses. The membrane is likely to sustain the latter without undergoing an appreciable deformation as they induce neither curvature nor shear flow in the bilayer. The only unbalanced force is due to the Maxwell stress exerted by the electric field just outside the vesicle, since in the enclosed water the field must be identical to zero. We assume here that the membrane is dielectrically isotropic, which implies that the forces caused by the Maxwell stresses are confined to the interfaces with water. If it is not, electrical torque densities may be expected within the bilayer. The torques may induce curvature, but this should be negligible as the polarizabilities of the bilayer should be very much smaller than that of water.

The standard expression for the electric potential  $U_{\rm e}$  around a sphere in a uniform applied field is

$$U_{\rm e} = -F_{\rm eo} z + A_{\rm e} (\cos \Theta/r^2). \tag{1}$$

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The field  $F_{\rm eo}$  is applied in z direction; r and  $\Theta$  are polar coordinates, the polar axis being parallel to the z axis. For an insulating sphere the radial component of the electric field must obey the condition

$$F_{\rm er}(r_{\rm e},\,\Theta) \equiv 0,$$
 (2)

where  $r_e$  is the radius of the sphere, *i. e.*, the outer radius of the bilayer shell. Insertion of (1) yields

$$F_{\rm eo} + (2 A_{\rm e}/r_{\rm e}^3) = 0,$$
 (3)

thus giving the constant  $A_{\rm e}$  .

In order to compute the electric energy of deformation we write down the force density per unit area, f, exerted on the water-sphere interface by the Maxwell stress of the external field. We have

$$f_{\rm r} = -(1/8\,\pi)\,\varepsilon_{\rm w}\,F_{\rm e\Theta}^2\,(r_{\rm e}\,,\,\Theta)\,,\tag{4}$$

where  $\varepsilon_{\rm w}$  is the dielectric constant of the (external) aqueous medium and  $F_{\rm e\theta}(r_{\rm e},\Theta)$  the field component along the meridians. There is no tangential component of the force. From (1) and (3) one obtains

$$f_{\rm r} = -(9/32\,\pi)\,\varepsilon_{\rm w}\,F_{\rm e0}^2\sin^2\Theta\,.\tag{5}$$

The radial displacement of the bilayer for a small ellipsoidal deformation leaving the bilayer area unchanged may be expressed as

$$s = (3/2) s_2(\cos^2 \Theta - 1/3).$$
 (6)

where the amplitude  $s_2$  of the second Legendre polynomial is a measure for the ellipticity. Accordingly, we have for the electric energy of deformation

$$E_{\rm d} = -\int_{0}^{\pi} f_{\rm r} \, s \, 2 \, \pi \, r_{\rm e}^{2} \sin \Theta \, d\Theta$$
$$= -(3/2) \, \varepsilon_{\rm w} \, F_{\rm e0}^{2} \, r_{\rm e}^{2} \, s_{2} \,. \tag{7}$$

The total curvature-elastic energy of the ellipsoidal deformation was calculated to be, for constant membrane area <sup>1</sup>.

$$E_{\rm c} = \frac{8\,\pi}{5} \ k_{\rm c} \ \frac{s_2^2}{r_0^2} \left(6 - r_0 \, c_0\right) \tag{8}$$

where  $k_{\rm c}$  is the curvature-elastic modulus and  $c_0$  the spontaneous curvature of the bilayer. The mean radius  $r_0$  and the outer radius  $r_{\rm e}$  of the shell are interchangeable whenever the thickness of the bilayer  $(50-100\,{\rm \AA})$  is much smaller than the radii. We restrict ourselves to this case, the only one of interest, as is to be seen immediately. Minimizing  $E_{\rm d}+E_{\rm c}$  yields the desired formula for the ellipticity induced by the electric field:

$$s_2 = \frac{3}{64 \pi} \varepsilon_w F_{e0}^2 \frac{r_0^4}{k_c} \frac{1}{(6 - r_0 c_0)} . \tag{9}$$



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The deformation is always prolate. For  $6 - c_0 r_0 < 0$  the spherical shape is unstable even without field <sup>2</sup>.

An earlier estimate  $^{1,\,2}$  of the curvature-elastic modulus is  $k_{\rm c}=5\cdot 10^{-13}\,{\rm erg}$ . In experiments  $F_{\rm eo}$  must be small enough to prevent excessive heat in the medium. For  $F_{\rm eo}=30~{\rm Vcm}^{-1}=0.1~{\rm cgs},~r_0=3\cdot 10^{-4}\,{\rm cm},~\varepsilon_{\rm w}=80,~{\rm and}~c_0=0$  one obtains  $s_2=3\cdot 10^{-5}\,{\rm cm}$ . This fairly strong deformation is near the limit of validity of the present approximation. However, since  $s_2\propto r_0^4$ , the effect decreases with decreasing vesicle radius more rapidly than its magnetic counterpart  $^{1,\,2}$  which varies at  $|s_2|\propto r_0^3$ .

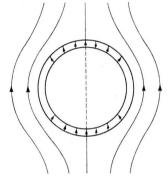


Fig. 1. Schematic field distribution around spherical vesicle.

It has been discussed previously <sup>2</sup> that strong deformations are likely to be hindered by slow permeation. In the case of large vesicles, pores or inserted tubes may be useful to avoid such difficulties. With strong fields and large vesicles one might hope that the needed holes can be generated, perhaps with a limited lifetime, by electric breakthroughs <sup>3</sup>. Within the membrane one has for the radial field strength

$$F_{\rm mr} = \frac{3}{2} \frac{r_{\rm o}}{b} F_{\rm eo} \cos \Theta , \qquad (10)$$

<sup>1</sup> W. Helfrich, Physics Letters **43 A**, 409 [1973].

<sup>2</sup> W. Helfrich, Z. Naturforsch. 28 c, 693 [1973].

where b is the thickness of the bilayer. The formula, valid for  $r_0 \gg b$ , is readily derived by expressing the potential within the bilayer by a form like (1). (The electric potential in the aqueous interior is, of course, identical to zero). Clearly,  $F_{\rm mr}$  can be very high for large vesicles.

The space charge (ion cloud) sustaining the high field in the membrane will be spread over a surface layer of water. Its thickness as well as that of any electric double layers is roughly given by the Debye screening length

$$a = (\varepsilon_{\mathrm{w}} \, k_{\mathrm{B}} \, T/4 \, \pi \, q^2 \, n)^{1/2}$$

where n is the ion concentration and q the ionic charge. The spreading of space charge can certainly be disregarded in calculating  $s_2$  if  $a \le b$ , as is the case with high ion concentrations. Despite the complexity of the situation, involving hydrostatic pressure and shear flow in the water, Eqn. (9) possibly remains valid for larger Debye lengths. This may be inferred from the fact that the internal Maxwell stresses in the membrane including the space charge layers can in principle be balanced, if we were dealing with a solid, by equal but opposite elastic stresses, and from the assumption that the bilayer is unstretchable. A detailed discussion of the limit of validity would be quite difficult and is not attempted here.

A bilayer may be electrically polar if its two sides are chemically different. Polarity may also result from curvature. Polar interaction with the applied field can be shown not to contribute to the deformation, at least not to a first approximation. Experimentally, it can be ruled out by the use of AC fields.

which diminishes the strength of the ellipsoidal deformation. However, the influence can be shown to be negligible for large vesicles  $(r_0 \gg b)$ , provided the holes are very few and small.

<sup>3</sup> Holes not on the vesicle "equator" (with respect to the applied field) will allow the field to penetrate the vesicle