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## Dielectrophoresis: a spherical shell model

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**Abstract.** The complete algebraic expression in the effective dipole approximation for the dielectrophoretic force acting on a spherical object surrounded by a shell when placed in an alternating electric field is deduced. In the approximation of a thin shell of low conductivity the frequency dependence of the dielectrophoretic force is discussed revealing the contribution of the shell. The electric potential difference across the shell is also calculated.

### 1. Introduction

Alternating or rotating electric fields are largely used in the displacing (Pohl 1978) or spinning (Quincke 1896, Teixeira-Pinto *et al* 1960, Arnold and Zimmermann 1982) of small particles, droplets, bubbles or biological cells. Pulsed electric fields can also be used in inducing the fusion of biological cells (Senda *et al* 1979, Zimmermann 1982) or the permeabilisation of their membranes (Zimmermann *et al* 1976).

All these phenomena, known as dielectrophoresis, electrorotation, electrofusion and electropermeabilisation, are described in terms of dielectrophoretic forces, electric torques and transmembrane potentials appearing when particles or cells are subjected to external electric fields. The main polarisation mechanism effective at frequencies up to  $10^7$  Hz is the Maxwell–Wagner polarisation. It originates in the differences between the electric properties of the suspended particle and those of the suspending medium.

The simplest theoretical model used to find some of these parameters consists of a homogeneous sphere immersed in a conducting dielectric medium. However the model fails to describe phenomena appearing in the case of biological cells which are heterogeneous having a complex internal structure. In such cases single- or multishell models seem to be suitable (Pauly and Schwan 1959).

In the present paper and in the following one (Turcu and Lucaciu 1989) we propose a unitary theoretical approach giving the complete algebraic expressions for the dielectrophoretic force, the electric torque and for the transmembrane potential appearing in a spherical single-shell model.

The comparison of some approximate formulae obtained in the shell model with the corresponding ones from the homogeneous sphere model reveals the essential contribution of the shell.

In this first paper we shall limit ourselves to finding the dielectrophoretic force and the transmembrane potential for the spherical shell model relevant in dielectrophoresis, electrofusion and electropermeabilisation phenomena.

**2. The spherical model**

Let us consider a conducting dielectric sphere suspended in a conducting dielectric medium and subjected to a uniform alternating electric field

$$\hat{E} = E \exp(i\omega t) \tag{1}$$

where  $\omega$  is the angular frequency of the electric field.

The internal and external media are characterised by the complex permittivities

$$\hat{\epsilon}_k = \epsilon_k - i\sigma_k/\omega \quad k = 1, 2. \tag{2}$$

The solution of the Laplace equation

$$\Delta\phi = 0 \tag{3}$$

for a sphere is well known:

$$\begin{aligned} \hat{\phi}_1 &= -[\hat{E}r - (\hat{P}R^3/3\epsilon_1 r^2)] \cos \theta & r \geq R \\ \hat{\phi}_2 &= -\hat{E}_2 r \cos \theta & r < R. \end{aligned} \tag{4}$$

The coefficients  $\hat{E}_2$  and  $\hat{P}$  are calculated using the boundary continuity of the potentials and of the normal components of displacements. The internal electric field  $E_2$  and the polarisation density  $P$  are obtained as the real parts of the corresponding complex quantities:

$$\begin{aligned} \hat{E}_2 &= 3 \frac{\hat{\epsilon}_1}{\hat{\epsilon}_2 + 2\hat{\epsilon}_1} \hat{E} \\ \hat{P} &= 3\epsilon_1 \frac{\hat{\epsilon}_2 - \hat{\epsilon}_1}{\hat{\epsilon}_2 + 2\hat{\epsilon}_1} \hat{E}. \end{aligned} \tag{5}$$

It is convenient to introduce the dimensionless susceptibility  $\hat{\chi}$  by:

$$\hat{P} = \epsilon_1 \hat{\chi} \hat{E} \tag{6}$$

and to put it in the compact form

$$\hat{\chi} = [K + N/(1 + i\omega\tau)]. \tag{7}$$

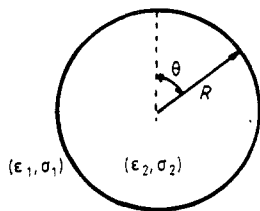
The real coefficients  $k$  and  $N$  are

$$K = 3(\epsilon_r - 1)/(\epsilon_r + 2) \tag{8}$$

$$N = -9(\epsilon_r - \sigma_r)/(\epsilon_r + 2)(\sigma_r + 2)$$

where the following notation has been introduced:

$$\epsilon_r = \epsilon_2/\epsilon_1 \quad \sigma_r = \sigma_2/\sigma_1 \quad \tau = (\epsilon_2 + 2\epsilon_1)/(\sigma_2 + 2\sigma_1). \tag{9}$$



**Figure 1.** The spherical model.

In the effective dipole approximation the dielectrophoretic force acting on a neutral sphere in a non-uniform electric field is given (Pohl 1978, Jones 1979) by

$$\mathbf{F} = (\mathbf{P}_{\text{eff}} \nabla) \mathbf{E} \quad (10)$$

and  $\mathbf{P}_{\text{eff}}$  is calculated as being induced by a uniform electric field. The approximation seems to be excellent for a not very strong non-uniformity.

From (6) and (10) one obtains the following expression for the time-dependent dielectrophoretic force (Benguigui and Lin 1982):

$$\mathbf{F} = \frac{1}{2} V \epsilon_1 [\text{Re } \hat{\chi} \cos \omega t - \text{Im } \hat{\chi} \sin \omega t] \cos \omega t \nabla E^2 \quad (11)$$

where  $V$  is the volume of the sphere and

$$\text{Re } \hat{\chi} = K + N/(1 + \omega^2 \tau^2) \quad \text{Im } \hat{\chi} = -N\omega\tau/(1 + \omega^2 \tau^2). \quad (12)$$

Taking the time average from (11) one gets

$$\langle \mathbf{F} \rangle = \frac{1}{4} V \epsilon_1 \text{Re } \hat{\chi} \nabla E^2. \quad (13)$$

The frequency dependence of the dielectrophoretic force is illustrated in figure 2 for several values of the dimensionless parameters  $\epsilon_r$  and  $\sigma_r$ .

The low-frequency limit is controlled by  $\sigma_r$ :

$$\lim_{\omega \rightarrow 0} \text{Re } \hat{\chi} = 3(\sigma_r - 1)/(\sigma_r + 2) \quad (14)$$

while the high-frequency limit is controlled by  $\epsilon_r$ :

$$\lim_{\omega \rightarrow \infty} \text{Re } \hat{\chi} = 3(\epsilon_r - 1)/(\epsilon_r + 2). \quad (15)$$

By examining some complex dielectrophoretic spectra for several types of biological cells (Pohl 1978) one can see that they are very badly approximated by the simple spectra given by the spherical model. We shall try to improve the theoretical prediction by introducing a spherical shell.

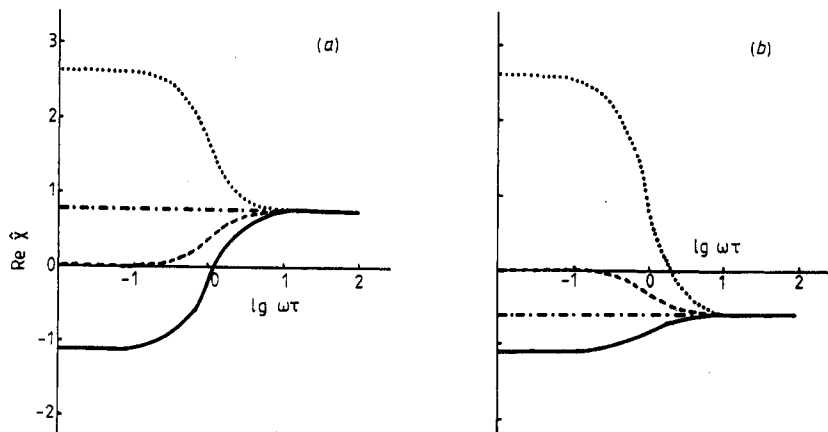


Figure 2. Frequency dependence of the real part of the complex electric susceptibility in the spherical model for (a)  $\epsilon_r = 2$  and (b)  $\epsilon_r = 0.5$  and for several values of the dimensionless parameter  $\sigma_r = 20$  (dotted curve), 1 (broken curve),  $\epsilon_r$  (chain curve), 0.2 (full curve).

**3. The spherical shell model**

Let us introduce a third medium between the internal and the external one having a spherical shell shape and being characterised by the complex permittivity

$$\hat{\epsilon}_m = \epsilon_m - i\sigma_m/\omega \tag{16}$$

where the index *m* stands for membrane.

The solution of the Laplace equation for a spherical shell is also well known:

$$\begin{aligned} \hat{\phi}_1 &= -(\hat{E}r - \hat{P}R^3/3\epsilon_1 r^2) \cos \theta & r \geq R \\ \hat{\phi}_m &= -(\hat{E}_m r - \hat{P}_m R_i^3/3\epsilon_m r^2) \cos \theta & R_i < r < R \\ \hat{\phi}_2 &= -\hat{E}_2 r \cos \theta & r \leq R_i \end{aligned} \tag{17}$$

where  $R_i = R - d$  with *d* being the thickness of the shell. The parameters  $\hat{P}$ ,  $\hat{E}_m$ ,  $\hat{P}_m$ ,  $\hat{E}_2$  are calculated using the following boundary conditions at the interfaces:

$$\begin{aligned} \hat{E} - \hat{P}/3\epsilon_1 &= \hat{E}_m - (1 - \delta)^3 P_m/3\epsilon_m \\ \hat{E}_m - \hat{P}_m/3\epsilon_m &= \hat{E}_2 \\ \hat{\epsilon}_1(\hat{E} + 2\hat{P}/3\epsilon_1) &= \hat{\epsilon}_m[\hat{E}_m + 2(1 - \delta)^3 P_m/3\epsilon_m] \\ \hat{\epsilon}_m(\hat{E}_m + 2\hat{P}_m/3\epsilon_m) &= \hat{\epsilon}_2 \hat{E}_2 \end{aligned} \tag{18}$$

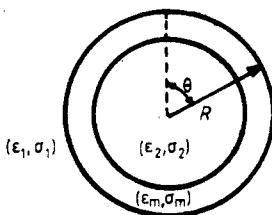
where  $\delta = d/R$ .

After some labourious but straightforward algebra the following expressions of the complex coefficients are obtained:

$$\begin{aligned} \hat{E}_m &= \frac{\hat{\epsilon}_1(\hat{\epsilon}_2 + 2\hat{\epsilon}_m)}{\hat{\epsilon}_m(\hat{\epsilon}_2 + 2\hat{\epsilon}_1) + 2\alpha(\hat{\epsilon}_1 - \hat{\epsilon}_m)(\hat{\epsilon}_2 - \hat{\epsilon}_m)} \hat{E} \\ \hat{E}_2 &= 3 \frac{\hat{\epsilon}_1 \hat{\epsilon}_m}{\hat{\epsilon}_m(\hat{\epsilon}_2 + 2\hat{\epsilon}_1) + 2\alpha(\hat{\epsilon}_1 - \hat{\epsilon}_m)(\hat{\epsilon}_2 - \hat{\epsilon}_m)} \hat{E} \\ \hat{P}_m &= 3\epsilon_m \frac{\hat{\epsilon}_1(\hat{\epsilon}_2 - \hat{\epsilon}_m)}{\hat{\epsilon}_m(\hat{\epsilon}_2 + 2\hat{\epsilon}_1) + 2\alpha(\hat{\epsilon}_1 - \hat{\epsilon}_m)(\hat{\epsilon}_2 - \hat{\epsilon}_m)} \hat{E} \\ \hat{P} &= 3\epsilon_1 \frac{\hat{\epsilon}_m(\hat{\epsilon}_2 - \hat{\epsilon}_1) - \alpha(\hat{\epsilon}_1 - \hat{\epsilon}_m)(\hat{\epsilon}_2 - \hat{\epsilon}_m)}{\hat{\epsilon}_m(\hat{\epsilon}_2 + 2\hat{\epsilon}_1) + 2\alpha(\hat{\epsilon}_1 - \hat{\epsilon}_m)(\hat{\epsilon}_2 - \hat{\epsilon}_m)} \hat{E} \end{aligned} \tag{19}$$

where

$$\alpha = \frac{1}{3}[1 - (1 - \delta)^3]. \tag{20}$$



**Figure 3.** The spherical shell model.

We shall also define the new quantity

$$\Delta\hat{\phi} = \hat{\phi}_2(R_i) - \hat{\phi}_1(R) \quad (21)$$

describing the potential difference on the shell. From (17), (19) and (21) one obtains

$$\Delta\hat{\phi} = 3\hat{\varepsilon}_1 \frac{\delta\hat{\varepsilon}_m + \alpha(\hat{\varepsilon}_2 - \hat{\varepsilon}_m)}{\hat{\varepsilon}_m(\hat{\varepsilon}_2 + 2\hat{\varepsilon}_1) + 2\alpha(\hat{\varepsilon}_1 - \hat{\varepsilon}_m)(\hat{\varepsilon}_2 - \hat{\varepsilon}_m)} \hat{E}R \cos \theta. \quad (22)$$

The expressions (19) and (22) are the main results of this paper. It is hard to use them in their full generality but there are many physically relevant cases when simplifying approximations can be done. In the next section we shall treat the case of thin shells with low conductivity, as specific to biological cells.

#### 4. Single-shell biological cell model

The interior of biological cells behaving like single-shell spheres can be well approximated by a homogeneous medium. Although, in most cases, living cells are nucleated and contain vacuoles and cellular organites and therefore more elaborate models seem to be appropriate, we consider that the systematic investigation of the single-shell model would be very useful for describing the basic facts.

In order to find explicit approximate expressions for the relations (19) and (22) in the case of thin low-conductivity membranes we introduce the dimensionless parameters

$$\varepsilon_{rm} = \varepsilon_m / \varepsilon_1 \quad \sigma_{rm} = \sigma_m / \sigma_1 \quad (23)$$

and we shall expand all the quantities of interest in powers of the dimensionless parameters  $\delta$  and  $\sigma_{rm}$  considered much smaller than unity:

$$\delta \ll 1 \quad \sigma_{rm} \ll 1. \quad (24)$$

The common denominator in all expressions (19) and (22):

$$\begin{aligned} \varepsilon_m(\varepsilon_2 + 2\varepsilon_1) \left(1 - \frac{i}{\omega} \frac{\sigma_m}{\varepsilon_m}\right) \left(1 - \frac{i}{\omega} \frac{\sigma_2 + 2\sigma_1}{\varepsilon_2 + 2\varepsilon_1}\right) \\ + 2\alpha(\varepsilon_1 - \varepsilon_m)(\varepsilon_2 - \varepsilon_m) \left(1 - \frac{i}{\omega} \frac{\sigma_1 - \sigma_m}{\varepsilon_1 - \varepsilon_m}\right) \left(1 - \frac{i}{\omega} \frac{\sigma_2 - \sigma_m}{\varepsilon_2 - \varepsilon_m}\right) \end{aligned} \quad (25)$$

can be put in the simple form

$$\Gamma(1 - i\Omega_1/\omega)(1 - i\Omega_2/\omega). \quad (26)$$

Equating (25) and (26) and retaining only the first-order terms in the small parameter  $\delta$  we find

$$\begin{aligned} \Gamma &= \varepsilon_m(\varepsilon_2 + 2\varepsilon_1) + 2\delta(\varepsilon_1 - \varepsilon_m)(\varepsilon_2 - \varepsilon_m) \\ \Omega_1 &= \frac{\sigma_m}{\varepsilon_m} + 2\delta \frac{[(\sigma_2 - \sigma_m)\varepsilon_m - (\varepsilon_2 - \varepsilon_m)\sigma_m][(\sigma_1 - \sigma_m)\varepsilon_m - (\varepsilon_1 - \varepsilon_m)\sigma_m]}{\varepsilon_m^2[(\sigma_2 + 2\sigma_1)\varepsilon_m - (\varepsilon_2 + 2\varepsilon_1)\sigma_m]} \\ &\quad [(\sigma_2 - \sigma_m)(\varepsilon_2 + 2\varepsilon_1) - (\varepsilon_2 - \varepsilon_m)(\sigma_2 + \sigma_1)] \\ \Omega_2 &= \frac{\sigma_2 + 2\sigma_1}{\varepsilon_2 + 2\varepsilon_1} - 2\delta \frac{\times [(\sigma_1 - \sigma_m)(\varepsilon_2 + 2\varepsilon_1) - (\varepsilon_1 - \varepsilon_m)(\sigma_2 + 2\sigma_1)]}{(\varepsilon_2 + 2\varepsilon_1)^2[(\sigma_2 + 2\sigma_1)\varepsilon_m - (\varepsilon_2 + 2\varepsilon_1)\sigma_m]} \end{aligned} \quad (27)$$

Even in this approximation the expressions of the two characteristic frequencies  $\Omega_1$  and  $\Omega_2$  are very intricate and therefore we have expanded them in powers of the second small parameter  $\sigma_{rm}$ . By retaining again only the first-order terms we have obtained

$$\begin{aligned} \Omega_1 &= \frac{\sigma_m}{\epsilon_m} + \frac{2\delta\sigma_2}{\epsilon_m(\sigma_r+2)} \\ \Omega_2 &= \frac{\sigma_2+2\sigma_1}{\epsilon_2+2\epsilon_1} \left[ 1 + \frac{2\delta}{\epsilon_r+2} \left( \frac{2(\epsilon_r-\sigma_r)^2}{\epsilon_{rm}(\sigma_r+2)} + \frac{\epsilon_r-\sigma_r}{\sigma_r+2} - \epsilon_{rm} \right) \right]. \end{aligned} \tag{28}$$

By introducing the third medium between the internal medium and the external one, a new characteristic frequency, controlled by the membrane parameters, appears. The relative shift of the spherical model characteristic frequency is much smaller than unity.

We shall now expand the effective polarisation  $\hat{P}$  in terms of the two small parameters in order to find the approximate expression of the dielectrophoretic force.

For convenience we have put the corresponding dimensionless susceptibility in the form

$$\hat{\chi} = \left( k + \frac{N_1}{1+i\omega\tau_1} + \frac{N_2}{1+i\omega\tau_2} \right) \tag{29}$$

where  $\tau_i = \Omega_i^{-1}$ ,  $i = 1, 2$ .

After a relatively straightforward algebra we have obtained the following expressions for the coefficients  $K$ ,  $N_1$  and  $N_2$ :

$$\begin{aligned} k &= 3 \frac{\epsilon_r-1}{\epsilon_r+2} \\ N_1 &= -\frac{9}{2} \frac{\sigma_r}{(\sigma_r+2)} \left( 1 + \frac{\sigma_{rm}(\sigma_r+2)}{2\delta\sigma_r} \right)^{-1} \\ N_2 &= -9 \frac{(\epsilon_r-\sigma_r)}{(\epsilon_r+2)(\sigma_r+2)} \end{aligned} \tag{30}$$

where for simplicity only the main contributions were retained. Equation (29) describes the dielectric effective susceptibility of a shelled sphere. The first and third terms correspond to the simple sphere; the second one describes the membrane contribution.

The mechanism responsible for this effective response is the interfacial polarisation at the two interfaces. The frequency dependence of  $\hat{\chi}$  can be described by the superposition of two simple Debye-type behaviours. This type of dependence seems to be characteristic for the effective dielectric response of layered media (Pohl 1978).

#### 4.1. The dielectrophoretic force

In the effective dipole approximation the time-averaged dielectrophoretic force acting on a spherical shell is given by

$$F = \frac{1}{4} V \epsilon_1 \left( K + \frac{N_1}{1+\omega^2\tau_1^2} + \frac{N_2}{1+\omega^2\tau_2^2} \right) \nabla E^2. \tag{31}$$

The first and the third terms in the bracket are similar to those of the spherical model. Although the shell is very thin, its relative contribution to the susceptibility is of the order of unity. It also introduces a new characteristic frequency located somewhere in the lower-frequency range.

The dielectrophoretic spectra predicted by the spherical shell model for various values of dimensionless parameters  $\epsilon_r$  and  $\sigma_r$  are shown in figure 4.

By comparing figures 2 and 4 it is easy to see that in the high-frequency range the spectra remain almost unchanged while for low frequencies they are drastically modified. The low-frequency limit becomes negative and parameter independent:

$$\lim_{\omega \rightarrow 0} \operatorname{Re} \hat{\chi} = -\frac{3}{2} \quad (32)$$

when the inequality

$$\sigma_{rm}/\delta \ll 1 \quad (33)$$

also becomes true.

Because in almost all the cases of interest

$$\Omega_1/\Omega_2 \ll 1 \quad (34)$$

in the middle-frequency range ( $\Omega_1 < \omega < \Omega_2$ ) the spherical-shell dielectrophoretic spectra have a shape similar to that predicted by the spherical model in the low-frequency range. The main consequence of the introduction of the shell is the appearance of a maximum in this middle-frequency range for the cases in which the inequality  $\sigma_r > \epsilon_r$  is accomplished. It is this type of behaviour that can qualitatively describe the experimentally measured spectra for biological cells.

#### 4.2. The electric potential on the shell

The potential across the membrane can be put in the handy form

$$\Delta \hat{\phi} = \left( Q + \frac{M_1}{1+i\omega\tau_1} + \frac{M_2}{1+i\omega\tau_2} \right) \hat{E}R \cos \theta. \quad (35)$$

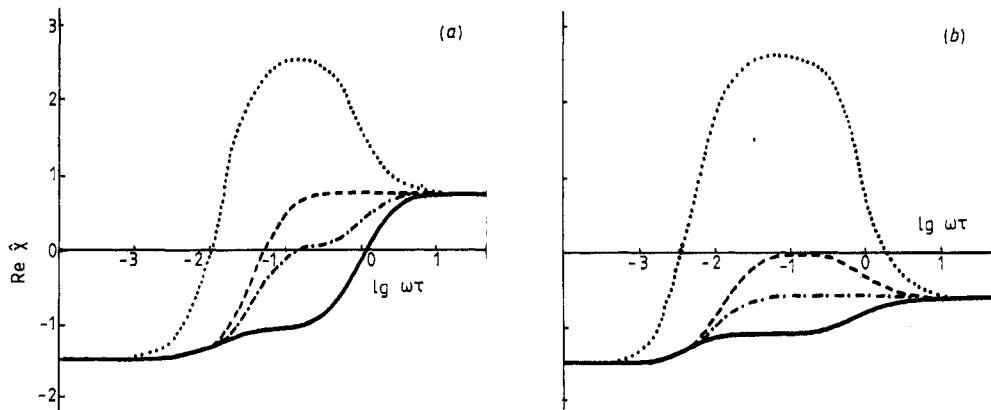


Figure 4. Frequency dependence of the real part of the complex electric susceptibility in the spherical shell model  $\sigma_{rm} = 0$ ,  $\delta = 10^{-3}$ ,  $\epsilon_{rm} = 0.1$  and (a)  $\epsilon_r = 2$ , (b)  $\epsilon_r = 0.5$ . Curves are drawn for four different values of  $\sigma_r = 20$  (dotted curve), 1 (broken curve),  $\epsilon_r$  (chain curve), 0.2 (full curve).



By equating (22) and (35) and using the by now common approximations  $\delta \ll 1$  and  $\sigma_{rm} \ll 1$ , we obtain

$$\begin{aligned}
 Q &= 3 \frac{\delta \varepsilon_r}{\varepsilon_{rm}(\varepsilon_r + 2)} \\
 M_1 &= \frac{3}{2} \left( 1 + \frac{\sigma_{rm}(\sigma_r + 2)}{2\delta\sigma_r} \right)^{-1} \\
 M_2 &= 6 \frac{\delta(\varepsilon_r - \sigma_r)}{\varepsilon_{rm}(\varepsilon_r + 2)(\sigma_r + 2)^2}.
 \end{aligned} \tag{36}$$

By extracting the real part of (35) we get

$$\Delta\phi = \left[ \left( Q + \frac{M_1}{1 + \omega^2\tau_1^2} + \frac{M_2}{1 + \omega^2\tau_2^2} \right) \cos \omega t + \left( \frac{M_1\omega\tau_1}{1 + \omega^2\tau_1^2} + \frac{M_2\omega\tau_2}{1 + \omega^2\tau_2^2} \right) \sin \omega t \right] ER \cos \theta \tag{37}$$

which can be put into the more compact form

$$\Delta\phi = V \cos(\omega t - \zeta). \tag{38}$$

Taking into consideration that  $Q$  and  $M_2$  are much smaller than  $M_1$  and keeping only the main contributions, the parameters  $V$  and  $\zeta$  are given by

$$V = \frac{M_1 ER \cos \theta}{(1 + \omega^2\tau_1^2)^{1/2}} \quad \zeta = \tan^{-1} \omega\tau_1. \tag{39}$$

The expression for the amplitude  $V$  commonly given in the literature (Zimmermann 1982) is

$$V = \frac{3}{2} ER \cos \theta (1 + \omega^2\tau_1^2)^{-1/2}. \tag{40}$$

This formula is indeed an excellent approximation because in the biological cell case the inequality (33) is always true.

Usually the electrofusion and electroporabilisation techniques are based on pulsed electric fields. The value of the transmembrane potential difference induced by a square pulse having a strength  $E$  and a length  $T$  is given by

$$\Delta\phi = \frac{3}{2} ER \cos \theta [1 - \exp(-T/\tau_1)]. \tag{41}$$

The maximum value is attained at the poles and is controlled by the field strength and by its duration.

## 5. Conclusions

The aim of this paper was to investigate from a theoretical point of view the dielectrophoretic force acting on a single spherical object surrounded by a shell. We have found the complete algebraic expressions for the dielectrophoretic force and for the potential difference on the shell appearing when the particle is subjected to external alternating electric fields.

In order to give specific predictions in some physically relevant cases, we have also found approximate formulae appropriate for a very thin shell of low conductivity. These restrictive conditions are very well accomplished by biological cells.

The approximate expression for the dielectrophoretic force and its frequency dependence was compared with that obtained for simple spherical objects in order to reveal the contribution brought by the shell. As has been shown, the presence of the shell modifies drastically the dielectrophoretic spectra in the low-frequency range. Besides the spherical model characteristic frequency, a second characteristic frequency appears, controlled by the shell and located somewhere in the lower-frequency range. It is between these two characteristic frequencies that, for  $\sigma_r > \epsilon_r$ , the dielectrophoretic spectra have a pronounced maximum. The low-frequency limit of the dielectrophoretic force which in the spherical model case is controlled by  $\sigma_r$  becomes negative and parameter independent.

Although very thin, the shell modifies in an essential manner the shape of the dielectrophoretic spectra.

From a phenomenological point of view this fact can be understood remembering that the polarisation originates in the interfacial charge accumulation. The presence of a shell, even very thin, has as a direct consequence the appearance of two interfaces both of which contribute to the polarisation. The effects are stronger for larger differences between the electric properties of each of the two pairs of adjacent media.

Starting from the expression of the potential difference across the shell we have systematically derived an approximate expression for a thin shell of low conductivity. The resulting expression is identical to that usually given in the literature.

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