Chapter 2
Electrokinetic Forces in Inhomogeneous Fields

This second chapter will describe the fundamentals of electrokinetic forces on particles suspended in a solvent medium. It starts from the basic observation of electrophoretic motion, which occurs for charged particles and summarizes its main properties. The main focus is on the derivation of a force equation for uncharged but polarizable particles that are subject to a strongly inhomogeneous electric field. In this field, the internal polarization of a micro-object interacts with the field, an effect called *dielectrophoresis*, which forms the basis for particle manipulation in this thesis.

2.1 Electrophoresis and Dielectrophoresis

For particles suspended in an electric field, the first observation in daily life is that a particle of charge q is influenced by other charges and the resulting electric field E:

$$F_{EP} = q \cdot E. \tag{2.1}$$

This effect is termed *electrophoresis*, and it is a standard school experiment for electrostatic interaction to rub a cotton cloth over a glass bar to separate charges on both parts and to demonstrate their mutual attraction. Furthermore, it can be employed in more useful ways to construct lenses in electron microscopy [1] or to manipulate charged matter in an electric field [2]. The predominant properties of electrophoretic movement can be described as follows:

- Particles must be charged to be affected by an electric field.
- Electrophoresis occurs regardless of the spatial structure of the electric field.
- A reverse of the sign of the charge or the orientation of the electric field is accompanied by a reverse of particle motion.
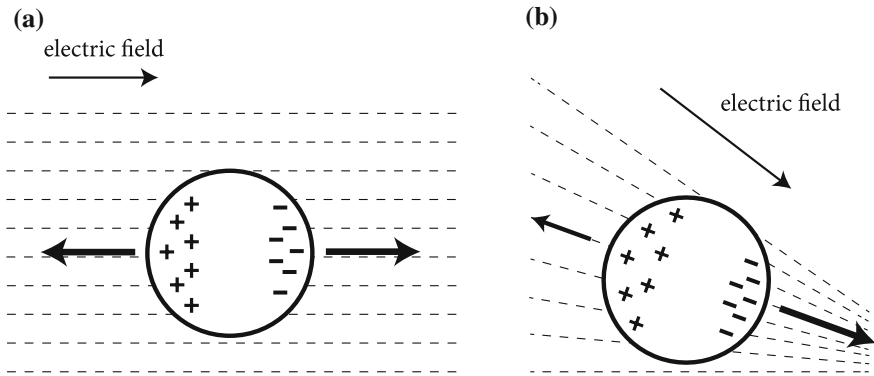- The mean displacement of electrophoretic motion vanishes in an alternating electric field.

(a)   **(b)**

Fig. 2.1 Polarizable particle influenced by an electric field: in **a** the particle is polarized, but the forces on both side counteract, while in **b** a *positive dielectrophoretic force* moves particle towards regions of high field intensity

The charging of particles, while being easy for macroscopic objects, may not be easily accomplished for microscopic objects. Once the micro particle comes into contact with the electrode of opposite charge, electrons may be transferred and the interaction is lost.

Another effect that is able to manipulate matter on the micro scale without the presence of excess charges was first described in 1958 by Pohl under the term *dielectrophoresis* [3], which is an extension of the term "electrophoresis" to include also dielectric, neutral particles. Dielectrophoresis (DEP) occurs when polarizable matter is subject to an inhomogeneous electric field. A descriptive illustration can be found in Fig. 2.1. If a polarizable particle is placed in a homogeneous electric field, free charge carriers on the particle become separated, which means that a dipole is induced. This separation is strongly dependent on the electric properties of the particle and the surrounding medium, as will be seen later. This dipole is influenced by counteracting forces of the same magnitude on both sides, hence it experiences no net force. A different situation occurs when the external electric field is inhomogeneous (cf. Fig. 2.1b). In this case the dipole experiences a force of higher magnitude on the side where the field intensity is higher, which causes the particle to be attracted to this region. This behavior is referred to as *positive dielectrophoresis* for reasons described later in this chapter. Of course this simple image neglects several effects that are important in the description of dielectrophoresis. The induced electrodynamic moment may not always be a dipole but consist of quadrupole or even more complex multipolar moments [4, 5]. Additionally, the electric field does not necessarily have to be constant, but can change in magnitude and even sign; the force may not always be attractive towards the regions of high field intensity but could as well be repulsive. To come to a more accurate description of the forces, the *effective moment method* as suggested by Jones [4] will be described in the following.

Fig. 2.2 Illustrative scheme
for the calculation of the
dielectrophoretic force, **a** net
force on a dipole, **b** effective
moment calculation
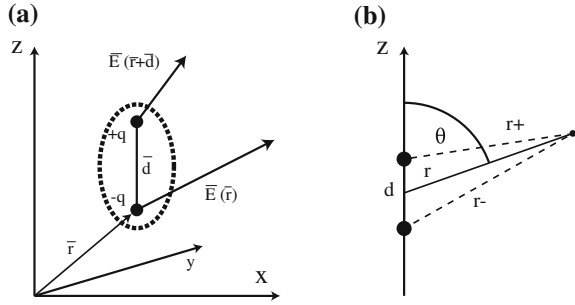
2.2 Dielectrophoretic Force Calculation

The calculation of the dielectrophoretic force on a microscopic particle suspended in
a medium has to account for the polarization of a particle and its ensuing interaction
with the electric field. An intriguing first approach is to consider a point dipole
that is subject to a non-uniform field. In this case the net force can be described as
the difference between the two electrophoretic forces acting on the two ends of the
dipole [4]:

$$\vec{F} = \vec{F}_{EP}(+q) + \vec{F}_{EP}(-q) = q\vec{E}(\vec{r} + \vec{d}) - q\vec{E}(\vec{r}). \tag{2.2}$$

Using a Taylor expansion and introducing ∇ as the gradient operator for the electric
field, the force can be approximated as

$$\vec{F} = \vec{p}\nabla\vec{E} + \ldots, \tag{2.3}$$

where higher order terms have been neglected and $\vec{p} = q\vec{d}$ has been introduced
as the dipole moment of a particle of size d. The challenge in describing the force
is the calculation of the dipole moment for microscopic polarizable particles and
to relate this moment to the particle properties. Jones suggested to use an effective
moment method to derive the dipole moment induced inside a dielectric particle [6].
The effective moment technique calculates the dipolar potential for a infinitesimal
dipole suspended in an isotropic liquid. This potential is compared to the solution
of Laplace's equation for a microscopic particle suspended in the same liquid, but
under the influence of a homogeneous electric field, which induces a very similar
dipole structure inside the dielectric particle.

From Fig. 2.2b, it is obvious that the potential Φ of a dipole can be calculated as
the superposition of two charge potentials separated by a distance d [6]:

$$\Phi(r, \theta) = \frac{q}{4\pi\varepsilon_1 r_+} - \frac{q}{4\pi\varepsilon_1 r_-}, \tag{2.4}$$

where ε_1 is the static permeability of the medium in which the charges are located. Using the geometric relation $r_\pm = (1 + (\frac{d}{2r})^2 \mp \frac{d}{r} \cos\theta)^{-1/2}$ and expanding this using its Taylor series, the potential of a dipole of finite extent can be written as [4]

$$\Phi_{\text{dipole}} = \frac{qd\cos\theta}{4\pi\varepsilon_1 r^2} + \frac{qd^3(5\cos^3\theta - 3\cos\theta)}{32\pi\varepsilon_1 r^4} + \dots, \qquad (2.5)$$

where the first term describes the potential of an infinitely small dipole and the second term is an octupole correction due to the finite extent of the charge separation. In order to relate the effective dipole moment in Eq. 2.5 to the particle properties, it is necessary to solve Laplace's equation with appropriate boundary conditions for a homogeneous sphere of radius R suspended in a constant electric field. In this case, the radial symmetric electrostatic potential can be written as the superposition of the dipole potential and the external electric field E_0 [4]:

$$\Phi_1(r, \theta) = E_0 r \cos\theta \qquad\qquad \text{for r > R}$$

$$\Phi_2(r, \theta) = E_0 r \cos\theta + \frac{A\cos\theta}{r^2} \qquad\qquad \text{for r} \leq \text{R.} \qquad (2.6)$$

It is required that both the potentials and the electromagnetic displacement flux across the boundary of sphere and fluid are continuous. With these assumptions, the unknown constant A can be determined to be [4]

$$A = \frac{\varepsilon_p - \varepsilon_m}{\varepsilon_p + 2\varepsilon_m} R^3 E_0, \qquad (2.7)$$

where $\varepsilon_{p,m}$ are the relative dielectric permittivities of the sphere (particle) and the surrounding medium, respectively.

Comparing this to the potential of a point dipole in Eq. 2.5, an expression can be derived for the effective dipole moment p_{eff}

$$p_{\text{eff}} = 4\pi\varepsilon_m R^3 E_0 \frac{\varepsilon_p - \varepsilon_m}{\varepsilon_p + 2\varepsilon_m}. \qquad (2.8)$$

Inserting this expression for the effective dipole moment into Eq. 2.3 and making use of the relationship $E_0\nabla E_0 = \frac{1}{2}\nabla E_0^2$, one arrives at the often quoted expression for the dielectrophoretic force on a homogeneous sphere in a dielectric medium [3, 6, 7]:

$$F_{\text{DEP}} = 2\pi R^3 \varepsilon_1 K(\varepsilon_m, \varepsilon_p)\nabla E_0^2, \qquad (2.9)$$

where $K(\varepsilon_m, \varepsilon_p)$ is the material- and shape-dependent Clausius-Mossotti factor that determines the magnitude and direction of the dielectrophoretic force. Note that this derivation of the force is only valid for a perfectly insulating sphere in a dielectric, non-conducting liquid. Using the same approach as before with minor extensions,

a finite conductivity and alternating electric fields of frequency ω can be included to arrive at a more general form of the force [6]:

$$\langle F(\omega)_{\text{DEP}} \rangle = 2\pi R^3 \varepsilon_m \Re(K(\omega)) \nabla \left\langle E_{\text{rms}}^2 \right\rangle, \tag{2.10}$$

where $\langle \dots \rangle$ defines the temporal average, $\Re$ the real part and E_{rms} the root mean square amplitude of the alternating electric field. For a particle of spherical shape, the Clausius-Mossotti factor is defined as [3]

$$K(\omega) = \frac{\varepsilon_p^* - \varepsilon_m^*}{\varepsilon_p^* + 2\varepsilon_m^*}, \tag{2.11}$$

with ε_k^* as the complex permittivity of the medium and sphere, respectively:

$$\varepsilon_k^* = \varepsilon_k + i\frac{\sigma_k}{\omega}. \tag{2.12}$$

2.3 Clausius-Mossotti Factor

Based on the electric properties of the materials under investigation, the real part of the complex Clausius-Mossotti factor determines the magnitude and direction of the dielectrophoretic force. If the real part is positive, the suspended sphere is attracted towards regions of high field intensity, commonly referred to as *positive dielectrophoresis*. The opposite case of *negative dielectrophoresis* occurs if the real part is negative and matter is repelled by high field intensities. By looking at Eq. 2.11, it becomes obvious that regardless of the electric properties of the materials under investigation, the real part of the Clausius-Mossotti factor for a spherical particle is fixed between $-\frac{1}{2} \leq K(\omega) \leq 1$. It is important to note that this restriction does not apply in all cases. For example, elongated structures like nanowires can possess values of $K(\omega)$ exceeding 1 by orders of magnitude [8], accompanied by substantially higher dielectrophoretic forces. Reference [4] gives further analytical expressions for the Clausius-Mossotti factor in the case of elliptical particles where the polarization along the longer and shorter axes are different and torques are exerted onto the structures. Even more complex forms, for which there is no analytical solution for the induced multipolar moments, have been treated numerically [9].

For many objects, however, the assumptions of a spherical shape is a good approximation and allows to predict dielectrophoretic forces. If objects under investigation deviate strongly from this shape, the force calculation has to be changed accordingly. Looking at Eqs. 2.11 and 2.12, it is obvious that the Clausius-Mossotti factor has two limiting values of

$$K(\omega \to 0) = \frac{\sigma_p - \sigma_m}{\sigma_p + 2\sigma_m} \tag{2.13}$$

for low frequencies, which means that in the case of slowly varying alternating fields the Clausius-Mossotti factor can be calculated solely knowing the respective conductivities, whereas for very high frequencies only the dielectric permittivities govern its magnitude:

$$K(\omega \to \infty) = \frac{\varepsilon_p - \varepsilon_m}{\varepsilon_p + 2\varepsilon_m}. \tag{2.14}$$

In between these limiting cases, the Clausius-Mossotti factor must be calculated taking into account both conductivity and permittivity. In particular, it can change its sign when the frequency is changed, for example, if a medium and particle are used where $\varepsilon_2 > \varepsilon_1$, but $\sigma_2 < \sigma_1$. Then for low frequencies, a suspended particle is repelled by field gradients into regions of low field intensity, whereas for high frequencies, it is attracted into high intensity areas. Analogous to the phenomenon of electrophoresis, the most important properties of dielectrophoresis can be summarized as follows:

- Dielectrophoresis acts on polarizable matter, even when it is uncharged.
- It occurs only in inhomogeneous electric fields.
- Due to the fact that the force is proportional to the gradient of E^2, its direction does not change upon repoling the electric field.
- Depending on the sign and magnitude of the Clausius-Mossotti factor, suspended particles are either attracted towards or repelled from regions of high field intensity.

Through these properties, DEP has lended itself to a multitude of sorting and manipulation concepts. These concepts either exploit that for suitable combinations of two different particles in a medium, the sign of the Clausius-Mossotti factor can be different [10], which leads to the spatial separation of particles or they aim at the strong size dependence of the force, which strongly affects the height to which particles can be levitated. This behavior enables a different speed of transport in pressure-driven Hagen-Poiseuille flows, a technique commonly denoted as flow-field fractionation [11, 12]. It is an interesting but often neglected fact about dielectrophoresis and optical tweezers, which were mentioned in the history of optically-induced particle trapping, that—while both tools are often described as two separate phenomena—both forces originate from the same description in the approximation of particles smaller than the electric field inhomogenity and that both can be reduced to the same form. It has been previously mentioned in Eq. 2.14 that in the limit of very high frequencies the Clausius-Mossotti factor is calculated by the relative permittivities. Considering that the refractive index for non-magnetic matter ($\mu_r = 1$) is defined as $n = \sqrt{\varepsilon_r}$, Eq. 2.14 becomes:

$$K(\omega \to \infty) = \frac{n_p^2 - n_m^2}{n_p^2 + 2n_m^2}, \tag{2.15}$$

which is nothing else than the polarizability α that is typically used in the field of optical tweezers to describe the magnitude and direction of optical forces in the

so-called Rayleigh regime [13, 14], which corresponds to the case of subwavelength particles. It should be noted however that the term *refractive index* is typically only used for the electromagnetic spectrum from UV to infrared, while in DEP literature the electric properties are summarized in the term *complex permittivity*, as described before.

2.4 Generalization of DEP for Large Objects and Continuous Media: Multipoles and Polarization Force Density

So far the calculation of the dielectrophoretic forces has only considered dipolar moments, which is valid if the field inhomogeneity is much larger than the particle itself. However, for the sake of completeness, it should be mentioned that situations may arise where this approximation does not hold any more, for example if the particles size is comparable to the electrode structure size or if the object under investigation is not a discrete sphere any more, but rather a continuous medium. The cases of a large sphere or any other object can be described by the general theory of multipoles of order n [5]:

$$\stackrel{\leftrightarrow}{\bar{p}}{}^{(n)} = \frac{4\pi \varepsilon_m R^{2n+1}}{(2n-1)!!} K^{(n)} (\nabla)^{n-1} \vec{E}, \tag{2.16}$$

with the higher order Clausius-Mossotti factor

$$K^{(n)} = \frac{\varepsilon_p^* - \varepsilon_m^*}{n\varepsilon_p^* + (n+1)\varepsilon_m^*}. \tag{2.17}$$

In the case of non-conducting dielectric fluids, a generalization of the force is made by introducing the *Kelvin polarization force density* $\vec{P}$, where the molecules of the fluid are considered as infinitesimal dipoles. $\vec{P}$ is equivalent to density of the dipoles N_p, multiplied by their dipole value $\vec{p}$ and the total force is calculated as the integral over the continuous volume that is to be actuated [15]:

$$\vec{F} = \int (N_P \vec{p}) \nabla \vec{E} dV = \frac{1}{2} \varepsilon_0 \int (\varepsilon_l - \varepsilon_m) \nabla \vec{E}^2 dV, \tag{2.18}$$

where $(\varepsilon_l - \varepsilon_m)$ describes the *excess polarization* of a dielectric substance l surrounded by a medium m [16]. Note that this derivation treats each dipole as independent and neglects the mutual interaction of different dipoles. Nevertheless, the Kelvin polarization density can be used to qualitatively explain the behavior of liquids later in this thesis. In general, it states that the same assumptions as before also apply to continuous media, namely that a medium is attracted to high field intensity regions in the case of a positive force, i.e. when it possesses a higher polarizability than its surrounding, and repelled if the total force is negative.

References

1. J. Orloff, L. Swanson, Asymmetric electrostatic lenses for field-emission microprobe applications. J. Appl. Phys. **50**(4), 2494–2501 (1979)
2. A. Cohen, W. Moerner, Method for trapping and manipulating nanoscale objects in solution. Appl. Phys. Lett. **86**(9), 093109 (2005)
3. H. Pohl, Some effects of nonuniform fields on dielectrics. J. Appl. Phys. **29**, 1182–1188 (1958)
4. T. Jones, *Electromechanics of Particles*. (Cambridge University Press, Cambridge, 1995)
5. T. Jones, M. Washizu, Multipolar dielectrophoretic and electrorotation theory. J. Electrost. **37**(1–2), 121–134 (1996)
6. T. Jones, Basic theory of dielectrophoresis and electrorotation. IEEE Eng. Med. Biol. Mag. **22**(6), 33–42 (2003)
7. H.A. Eggert, F.Y. Kuhnert, K. Buse, J.R. Adleman et al., Trapping of dielectric particles with light-induced space-charge fields. Appl. Phys. Lett. **90**, 241909 (2007)
8. A. Jamshidi, P.J. Pauzauskie, P.J. Schuck, A.T. Ohta et al., Dynamic manipulation and separation of individual semiconducting and metallic nanowires. Nat. Photonics **2**(2), 85–89 (2008)
9. N.G. Green, T.B. Jones, Numerical determination of the effective moments of non-spherical particles. J. Phys. D Appl. Phys. **40**(1), 78–85 (2007)
10. H. Morgan, M. Hughes, N. Green, Separation of submicron bioparticles by dielectrophoresis. Biophys. J. **77**(1), 516–525 (1999)
11. J. Rousselet, G. Markx, R. Pethig, Separation of erythrocytes and latex beads by dielectrophoretic levitation and hyperlayer field-flow fractionation. Coll. Surf. A Physicochem. Eng. Asp. **140**(1–3), 209–216 (1998)
12. H.-S. Moon, K. Kwon, S.-I. Kim, H. Han et al., Continuous separation of breast cancer cells from blood samples using multi-orifice flow fractionation (MOFF) and dielectrophoresis (DEP). Lab Chip **11**(6), 1118–1125 (2011)
13. P. Smith, A. Ashkin, W. Tomlinson, 4-wave mixing in an artifical Kerr medium. Opt. Lett. **6**(6), 284–286 (1981)
14. A. Ashkin, J.M. Dziedzic, J.E. Bjorkholm, S. Chu, Observation of a single-beam gradient force optical trap for dielectric particles. Opt. Lett. **11**, 288–290 (1986)
15. H. Hauser, J. Melcher, Electromagnetic fields and energy (2013). Prentice-Hall, NJ, http://ocw.mit.edu. (1989)
16. S. Schmid, C. Hierold, A. Boisen, Modeling the Kelvin polarization force actuation of micro- and nanomechanical systems. J. Appl. Phys. **107**(5), 054510 (2010)

Springer