Ellipsoidal particles driven by intensity gradients through viscous fluids

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We investigate the drift velocity v_{drift} of ellipsoidal polarizable particles (ellipsoids and coated ellipsoids), driven through a viscous fluid by an electric or electromagnetic field intensity gradient. At low Reynolds number and in the dipole approximation v_{drift} is proportional to the square of the principal axis along the direction of motion multiplied by a form factor, which is weakly depending on the shape of the particle, and by a frequency and shape dependent factor $f(\omega)$. Near frequencies where the real part of $\varepsilon_m f(\omega)$ changes sign (ε_m is the relative dielectric function of the medium in which the partcle is immersed), v_{drift} is sensitive to the shape of the particle. We suggest that our results can be used for the experimental separation of neutral polarizable particles with respect to size or shape.

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I. INTRODUCTION

The manipulation of particles using electromagnetic fields is an area of great current interest. Nobel prizes have recently been awarded in physics for the development of methods that allow trapping and cooling of atoms down to a few μK using laser light [1] and for creating (using this laser cooling technique) the Bose-Einstein condensates [2,3]. When studying biological systems two other techniques have become important. *Optical tweezers* allow manipulation of single dielectric particles; a strong electromagnetic intensity ''spot'' is created by focusing laser beams through a microscope objective. A particle in the spot becomes highly polarized, leading to a large electromagnetic interaction energy. If the spot intensity is large enough, thermal fluctuation cannot move the particle that then becomes trapped. The optical tweezers has, for instance, been used to knot DNA molecules [4], study the motion of motor proteins [5], and to induce shape transformations in cells [6]. In *electrophoresis* a static electric field is used for the separation of particles. If a charged particle is placed in a viscous fluid and is under the action of an external electric field, there is a net force on the particle which (counteracted by the viscous friction force against the fluid molecules) causes the particle to move with a drift velocity $\vec{v}_{\mathrm{drift}}=\vec{\vec{\mu}}\cdot\vec{E}$, where $\vec{E}$ is the electric field at the particle and $\vec{\vec{\mu}}$ is the so-called mobility tensor of the particle. This allows separation of particles of different mobility (of different charges, sizes etc.) [7–10].

In this study we discuss how to separate *neutral* particles such as cells, using electric and electromagnetic fields. Unfortunately, electrophoretic separation requires charged particles and does not work for neutral particles. Optical tweezers, on the other hand, can be applied to electrically neutral particles, but only a few particles at a time can be manipulated. If the particle is immersed in a conducting solvent with a conductivity different from that of the particle, *electromagnetophoretic* separation may be used [11,12]. This technique employs the combined effect of electric and magnetic fields in order to cause motion of the particles. Here we describe a separation method that uses electric or electromagnetic field intensity *gradients* and works for both conducting and nonconducting solvents. An electric or electromagnetic field intensity gradient exerts a force on a neutral particle, which is therefore able to move through a viscous fluid. In the presence of such a gradient, the drift velocity of the particle is (see next section)

$$\vec{v}_{\mathrm{drift}}=\mathrm{Re}(\vec{\vec{K}}\cdot\vec{\nabla}W), \tag{1}$$

where W is the energy density of the external electric field and we have introduced $\vec{\vec{K}}$ which we refer to as the *intensity gradient mobility tensor*. The tensor $\vec{\vec{K}}$ has dimension $[\mathrm{K}]=\mathrm{m}^3\,\mathrm{s/kg}$ and plays the same role for neutral particles as does the mobility tensor for charged particles. As we will see, the intensity gradient mobility tensor depends on the size, shape, and electric properties of the particle. Besides it depends on the viscosity of the fluid in which the particle is immersed. In the rest of this study we investigate $\vec{\vec{K}}$ more closely and in particular: in Sec. II a general expression for $\vec{\vec{K}}$ is derived. This expression depends on the polarizability and the hydrodynamic ''effective radius'' of the particle. In Sec. III standard expressions for the polarizability and effective radius for *ellipsoidal* particles (uncoated and coated ellipsoids) are combined with the result in the previous section to give the intensity gradient mobility tensor. Finally we discuss the separation of neutral particles, such as cells, as a possible application.

II. INTENSITY GRADIENT MOBILITY TENSOR

In this section we derive an expression for the intensity gradient mobility tensor of a particle driven through a viscous fluid in an electric or electromagnetic field intensity gradient (at small Reynolds number) in terms of the polarizability and the hydrodynamic effective radius of the particle.

Let us study a particle in a viscous fluid assuming that there is a time-dependent force $\vec{F}(t)$ on the particle. The molecules in the fluid collides in a stochastic fashion with the particle, which then exhibits a random type of motion that in the general case is described by Langevin's equation [13,14]: $m d\vec{v}(t)/dt=\vec{F}(t)-\vec{\vec{\xi}}\vec{v}(t)+\vec{g}(t)$, where $\vec{v}(t)$ is the velocity of the particle at time t. The term on the left-hand side is the inertial term (m is the mass of the particle). The second term on the right-hand side is a viscous (dissipative) term and $\vec{\vec{\xi}}$ is the *friction tensor* of the particle. $\vec{g}(t)$ is a

stochastic force [the ensemble average is zero, $\langle \vec{g}(t) \rangle = \vec{0}$] and represents the ''thermal collisions'' by the molecules in the fluid and the particle. The ''amplitude'' of the stochastic force is determined by the fluctuation-dissipation theorem [14], which relates this amplitude to the friction.

We are primarily interested in applying our analysis to biological systems such as cells moving with small velocities through viscous fluids. We will therefore through the rest of this study assume that the dimensionless number

$$R = \frac{L v \rho}{\eta} \tag{2}$$

is small, i.e., that $R \ll 1$. L is a characteristic length of the particle and η is the viscosity of the fluid. This condition is assumed to be satisfied *both* for the particle density ($\rho = \rho_{\text{particle}}$) and the fluid density ($\rho = \rho_{\text{fluid}}$). When ρ is taken as the particle density, R is the ratio between the inertial force and the viscous force in Langevin's equation above (the condition $R \ll 1$ hence allows us to neglect particle inertial effects) [15]. When ρ is taken as the fluid density R becomes the so-called *Reynolds number*, which plays an important role in hydrodynamics [16]. As an example of small R motion, consider a bacterium in water. A bacterium is typically one micron ($L \sim 1\,\mu$m) in size. If such an object moves by a speed, $v = 1$ mm/s we have $R \sim 10^{-3}$ (assuming that the density of the bacterium is approximately equal to that of water). We hence neglect inertial effects ($R \ll 1$) in this study, and Langevin's equation then becomes [14]

$$\vec{F}(t) - \overleftrightarrow{\xi}\vec{v}(t) + \vec{g}(t) = 0. \tag{3}$$

This equation is valid for fluids with high viscosity, small particle velocities, and sizes.

Let us now consider the drift velocity of the particle through the fluid. Because of the stochastic nature of the motion of the particle (as contained in Langevin's equation), the particle position $\vec{x}$ must be described in terms of a *probability distribution* $P(\vec{x},t)$ (or in terms of a probability distribution for different velocities). The mean velocity (the drift velocity) $\vec{v}_{\text{drift}} \equiv \langle \vec{v} \rangle$ can, however, be obtained without a full knowledge of the probability distribution $P(\vec{x},t)$. Let us restrict ourselves to motion along some principal axis of the particle. The viscous force is then in the same direction as the velocity of the particle and $\overleftrightarrow{\xi}$ is diagonal. We then take the ensemble average of Eq. (3) in order to obtain the drift velocity of the particle according to

$$v_{\text{drift,u}} = \frac{F_u}{\xi_{uu}}, \tag{4}$$

where ξ_{uu} is the friction constant for motion along the u direction ($u = x,y,z$) [17]. We now consider fluctuations around this mean value as caused by the stochastic force $\vec{g}(t)$. The mean distance, x_{drift}, a particle travels under the influence of the external force during a time t is $x_{\text{drift}} \equiv v_{\text{drift}} t$ [18]. On top of this [due to the stochastic force $\vec{g}(t)$], there is a diffusive motion which tends to spread the positions of the particles around x_{drift}. Since for diffusive motion the ''spread'' varies as $\sqrt{Dt}$ (where $D \equiv k_B T/\xi$ is the diffusion constant for the particle) we can always, by choosing t sufficiently large, separate particles with different v_{drift}.

Let us now take the external force appearing in Eq. (4) as the the force on a small, neutral polarizable particle as caused by an external electric or electromagnetic field intensity gradient. We also assume that the magnetic susceptibility of the particle equals the susceptibility of the surrounding medium. The general expression in the *dipole approximation* for the time-averaged electric force on a neutral particle is [19] $F_u(\vec{x},\omega) = \Sigma_v \text{Re}[P_v(\vec{x},\omega)\partial E_v^*(\vec{x},\omega)/\partial u]/2$, where $P_v(\vec{x},\omega)$ is the v component of the total induced dipole moment of the particle and $E_v(\vec{x},\omega)$ is the electric field at the position $\vec{x}$ of the particle. We now assume that there is a linear relationship between the induced dipole and the electric field (linear response) at the particle according to $P_v(\omega) = \Sigma_r 4\pi\varepsilon_m\varepsilon_0\alpha_{vr}(\omega)E_r(\omega)$, where ε_0 is the permittivity of vacuum and ε_m is the relative dielectric function of the medium surrounding the particle. We have also introduced the *polarizability* $\alpha_{uv}(\omega)$ of the particle (which has dimension of volume, $[\alpha] = \text{m}^3$). Inserting the above equation into the equation for the force and taking the form of the electric field such that it has only one component, taken to be the u component ($u = x,y,$ or z), and such that this component only depend on u [two examples are (i) the only spatial variation of the electric field is in the x direction, i.e, $\vec{E} = (E_x(x),0,0)$ (ii) the electric field only varies along the z direction and $\vec{E} = (0,0,E_z(z))$], we find [20]

$$F_u = \text{Re}\left(4\pi\alpha_{uu}\frac{\partial}{\partial u}W \right), \tag{5}$$

where W is the energy density of the electric field (the *intensity* is obtained by multiplying the energy density by the speed of light c) at the particle

$$W = \frac{1}{4}\varepsilon_m\varepsilon_0|E_u(u)|^2. \tag{6}$$

The force is determined by the polarizability, which is a property of the particle, and the gradient of the intensity $c\vec{\nabla}W$. The force on a neutral particle is thus towards higher field intensities if $\alpha > 0$.

Let us now consider the friction constant $\overleftrightarrow{\xi}$ occurring in Eq. (4). The flow of an incompressible fluid around a moving particle is in the general case obtained by solving (nonlinear) Navier-Stoke's equation [16]. The boundary condition at the interface between the particle and the fluid is that of no-slip and no penetration. The force exerted by the fluid on the particle (and hence the friction tensor) is thereafter obtained by integrating the hydrodynamic stress tensor over the surface of the particle. For the case of low Reynolds number [see Eq. (2)], Navier-Stoke's equation is linear. The integration of the stress tensor may then be formally performed [16] and the result for the friction tensor is [21]

$$\xi_{uv} = 6\pi\eta R_{\text{eff},uv}, \tag{7}$$

where η is the viscosity of the fluid. The 3×3 tensor $R_{\mathrm{eff},uv}$ (with dimension of length) is the hydrodynamic ''effective radius'' of the particle and is most conveniently obtained by performing an asymptotic expansion of the solution to the fluid equations of motion [16]. For a spherical particle, the effective radius is a scalar equal to the radius of the particle, and the above result is the well-known Stoke's law [16,22]. It is interesting to note that the viscous force is proportional to the effective *radius* (dimension of length) of the particle instead of (as might be naively assumed) the cross sectional *area* of the particle. This can be understood through dimensional arguments; the only parameters entering the analysis are η and u (for low Reynolds numbers Navier-Stoke's equation is independent of the fluid density ρ). The only combination of these quantities giving dimension of force is $\eta L u$ (since $[\eta]=\mathrm{kg/ms}$), where L is a characteristic length scale of the particle.

We are now in a position to obtain a general expression for the intensity gradient mobility tensor of a particle moving (along one of its principal directions) in an electric or electromagnetic field intensity gradient through a viscous fluid. By combining Eqs. (1), (4), (5), and (7) we obtain the intensity gradient mobility tensor

$$\eta K_{uu}=\frac{2}{3}\frac{\alpha_{uu}}{R_{\mathrm{eff},uu}}. \tag{8}$$

To obtain a large flow of particles towards high intensity regions we should have high intensity gradients [see Eq. (1)], large polarizability and small viscosity, and small effective radius of the particle. Notice that the only *particle* parameters entering the expression for the intensity gradient mobility tensor are the polarizability and the hydrodynamic effective radius. Since both of these entities are, in general, dependent on the shape and size of the particle, v_{drift} depends in a nontrivial fashion on the geometrical structure of the particle under consideration. By putting neutral particles into an electric field gradient, it should therefore be possible to separate different shapes and sizes. In the following section we investigate the drift velocity of elliptic particles as an example.

III. ELLIPSOIDAL PARTICLES

In this section we combine the well-known results for the hydrodynamic effective radius and the polarizability for *ellipsoidal* particles (ellipsoids and coated ellipsoids) in order to obtain the drift velocity v_{drift} (illustrations are only given for *spheroidal* particles, i.e. particles with two of the principal axes equal). We consider only cases below where the gradient of the intensity is parallel to v_{drift}, i.e., where the system has already come to a ''stationary'' state concerning torques.

The polarizability α_{uv} for a homogeneous particle is obtained through the solution of Maxwell's macroscopic equations. In the limit of long wavelengths λ of the electric field compared to a typical length scale L of the object, $L/\lambda\ll1$, the *electrostatic approximation* can be used in order to find α_{uv} [23]. The result for the polarizability for an ellipsoid *and* for a coated ellipsoid is then [23,24]

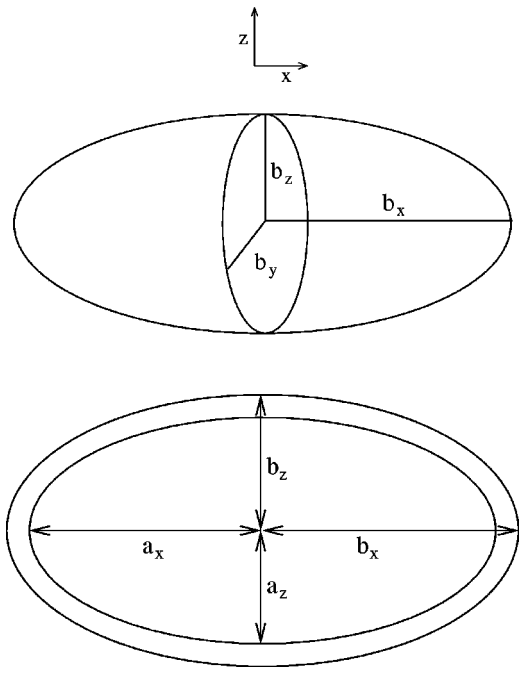

FIG. 1. Model geometry, (top) an uncoated ellipsoid with principal axes b_x, b_y, and b_z. (Bottom) A cut through a coated ellipsoid. The principal axes perpendicular to the paper are of length b_y (the outer ellipsoid) and a_y (the inner ellipsoid). When considering motion of *spheroids* we let $b_y=b_z$ and $a_y=a_z$ and distinguish between (i) motion along the rotationally invariant axis (motion along the *x* axis) and (ii) motion perpendicular to the rotationally invariant axis (motion along the *z* axis).

$$\alpha_{uu}=\frac{V}{4\pi n_u}f(\omega,n_u), \tag{9}$$

where the depolarization factor n_u is defined in Eq. (A3) and depends on the principal radii b_u $(u=x,y,z)$ of the particle. n_u contains all information on the shape of the particle. $V=4\pi b_x b_y b_z/3$ is the total volume occupied by the elliptic particle (see Fig. 1). The function $f(\omega,n_u)$ is different for the ellipsoid and the coated ellipsoid. For the ellipsoid we have

$$f_{\mathrm{ell}}(\omega,n_u)=\frac{\varepsilon_p(\omega)-1}{\varepsilon_p(\omega)-1+1/n_u}. \tag{10}$$

We have defined $\varepsilon_p(\omega)\equiv\varepsilon_{\mathrm{part}}(\omega)/\varepsilon_m(\omega)$, where the (complex) dielectric function for the ellipsoid is denoted by $\varepsilon_{\mathrm{part}}(\omega)$ and the dielectric function for the medium surrounding the particle is as before $\varepsilon_m(\omega)$. Notice that the complex dielectric functions include the effect of both ''free'' (ions or electrons) and ''bound'' charges [25]. Let us now consider $f(\omega,n_u)$ for a coated ellipsoid. Denote the dielectric function of the inner ellipsoid (with principal axes a_x, a_y, and a_z, see Fig. 1) by $\varepsilon_{\mathrm{inner}}(\omega)$. Similarly denote the dielectric function of the coating by $\varepsilon_{\mathrm{coat}}(\omega)$. We then have [23]

$$f_{\mathrm{coat}}(\omega,n_u) = \frac{(\varepsilon_c-1)[\varepsilon_c+(\varepsilon_i-\varepsilon_c)\gamma]+\varepsilon_c(\varepsilon_i-\varepsilon_c)V_i/V}{(\varepsilon_c-1+1/n_u)[\varepsilon_c+(\varepsilon_i-\varepsilon_c)\gamma]+\varepsilon_c(\varepsilon_i-\varepsilon_c)V_i/V}, \tag{11}$$

where $\varepsilon_c(\omega)\equiv\varepsilon_{\mathrm{coat}}(\omega)/\varepsilon_m(\omega)$ and $\varepsilon_i(\omega)\equiv\varepsilon_{\mathrm{inner}}(\omega)/\varepsilon_m(\omega)$. We have also introduced $\gamma\equiv n_u^i-n_uV_i/V$, where V is the total volume of the particle and $V_i=4\pi a_x a_y a_z/3$ is the volume occupied by the inner ellipsoid. n_u^i is the depolarization factor of the inner elliptical surface and is obtained by replacing b_u by a_u in Eq. (A3). Notice that the above expression for $f(\omega,n_u)$ reduces to that given by Eq. (10) as it should, if the dielectric functions for the coating and the inner ellipsoid are equal, $\varepsilon_c=\varepsilon_i$, or if the coating thickness is zero, $V_i=V$.

We now turn to the problem of the viscous force exerted on an ellipsoidal particle moving along one of its principal axes in a viscous fluid. The solution of the low Reynolds number Navier-Stoke's equation and the corresponding viscous force was worked out in Ref. [26]. For motion along the u axis, the hydrodynamic effective radius was found to be [27]

$$R_{\mathrm{eff},uu}=\frac{1}{\pi}\frac{V}{Q+b_u^2n_u}, \tag{12}$$

where b_u is the principal radii along the direction of motion. The depolarization factor is as before given by Eq. (A3) and Q is given in Eq. (A1); both of these entities depend on the shape of the particle. It is interesting to note that both the polarizability and the hydrodynamic effective radius depend on the very same n_u. However, the effective radius depends on the shape also through Q.

The intensity gradient mobility is given by Eq. (8) where, as we have seen, both the polarizability and effective radius depend on the particle shape. We now combine Eqs. (9) and (12) in order to find the intensity gradient mobility tensor

$$\eta K_{uu}=\frac{l_u^2}{6}g_u f(\omega,n_u), \tag{13}$$

where we have introduced $l_u=2b_u$ which is the length of the principal axis along the direction of motion. $f(\omega,n_u)$ is given by Eq. (10) or (11), and we have also introduced

$$g_u\equiv\frac{1}{4}\left(1+\frac{Q}{b_u^2n_u}\right). \tag{14}$$

From Eqs. (1) and (13) we see that the drift velocity is inversely proportional to the viscosity and proportional to the square length of the axis, along which the particle moves, multiplied by a form factor g_u depending on the ellipticity, and by a frequency and shape dependent factor $f(\omega,n_u)$.

Let us first consider g_u. Using the results in the Appendix it is possible to obtain an analytical expression for g_u for the case of spheroidal particles (i.e., ellipsoidal particles which have two of their principal axes equal), see Fig. 1. We choose

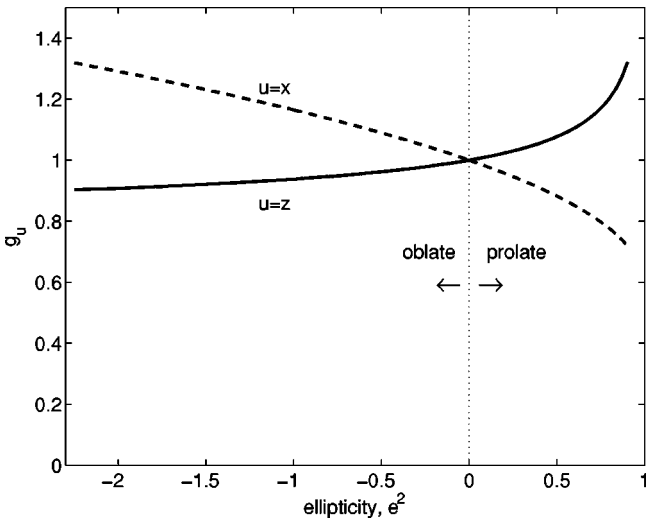

FIG. 2. The form factor g_u [g_u ($u=x,z$) is proportional to the drift velocity for a spheroid along u] as a function of ellipticity, $e^2=1-b_z^2/b_x^2$, see Fig. 1. The dashed ($--$) curve corresponds to g_x and the solid curve ($-$) corresponds to g_z. Notice that g_u is only weakly depending on the ellipticity (shape).

$b_y=b_z$ (and $a_y=a_z$). The shape dependent factor g_u is then completely described by the ellipticity $e^2\equiv1-b_z^2/b_x^2$ (where b_x and b_z are the two independent principal radii, see Fig. 1). When evaluating g_u for a spheroid we need to distinguish between two cases: (i) g_x determines v_{drift} for motion along the rotationally invariant axis (motion along x axis in Fig. 1) (ii) g_z gives the velocity for motion perpendicular to the rotationally invariant axis, i.e., the spheroid moves along the z axis in Fig. 1. Using Eqs. (A11) and (A12) we then find

$$g_x=\frac{1}{4}\left[1+e^2+\frac{1}{n_x(e)}(1-e^2)\right] \tag{15}$$

and

$$g_z=\frac{1}{4(1-e^2)}\left(1-3e^2+\frac{1}{n_z(e)}\right), \tag{16}$$

where $n_x(e)$ is given by Eqs. (A5) and (A7). $n_z(e)$ is given by Eqs. (A6) and (A8). Notice that the above results reduce to the result for a sphere $g_u=1$ in the limit $e\to0$. In Fig. 2 the ''correction'' form factors g_x and g_z are plotted. The correction form factors are only weakly dependent on e; for the interval shown in Fig. 2 the deviation from the sphere result is less than 30%. Corrections to the l_u^2 dependence of the drift velocity (for fixed l_u and f) are found for a very elongated (''cigar-shaped'') prolate spheroids, where the correction factor is less than one (<1) for motion along their long axis and larger than one (>1) for motion along their shortest axis. For very thin (''pancake-shaped'') oblate spheroids the correction factor is larger than one (>1) for motion along the shortest axis and less than one (<1) for motion along the large axis. Hence for fixed values of f (far from any resonance frequency of the particle, see below), the intensity gradient mobility tensor is insensitive to the shape and in-

stead the squared length of the axis along the direction of motion predominantly determines the drift velocity.

The frequency dependent part of the drift velocity is contained in $f(\omega,n_u)$. We notice from Eqs. (1), (6), (8), and (9) that if there exists a frequency ω_0 such that $\mathrm{Re}[\varepsilon_m f(\omega,n_u)]$ changes sign, then for $\omega<\omega_0$ the particle moves in the opposite direction compared to the case when $\omega>\omega_0$ (for $\omega=\omega_0$ the drift velocity is zero). Since $f(\omega,n_u)$ depend on the geometry of the particle but in general not its size, the frequency ω_0 will depend on the shape of the particle. This forms the basis for shape dependent separation of ellipsoidal particles; by tuning the frequency of the electric field appropriately one may, for instance, make particles of different shapes go in opposite direction. One example of when this happens is if $\varepsilon_m f(\omega,n_u)$ has a pole at the frequency ω_0 (the imaginary part of $\varepsilon_m f$ is large). Then (through the Kramer-Kronig relation and hence causality) the real part of the polarizability changes sign [28]. Precisely at the resonance frequency ω_0 we have that $\mathrm{Re}(\varepsilon_m f)=0$, and therefore the electric force on the particle is zero [see Eqs. (1) and (13)]. However, the imaginary part of $\varepsilon_m f$ is large at ω_0 and a strong absorption occurs either in the solvent [$\mathrm{Im}(\varepsilon_m)$ is large] or in the particle [$\mathrm{Im}(f)$ is large]. One then gets heating of the solvent and corresponding convection effect may be important at this frequency. One must therefore be careful to tune the frequency sufficiently far below or above ω_0 in order to avoid convection effects that may affect the shape dependent separation discussed in this paragraph. From Eq. (10) we see that f_{ell} has poles at frequencies ω_0 such that $\varepsilon_p(\omega_0)=1-1/n_u$. From Eq. (11) we notice that a coated ellipsoid, in general, has poles at different frequencies than does an uncoated particle. What frequency range that is most appropriate for separation is thus determined by the frequency dependence of the dielectric functions. We wish to keep our results as general as possible, and do not consider any specific dielectric function (which in turn would require a microscopic treatment).

A particularly interesting case of a coated particle is a cell (the coating being the cell membrane). We are currently investigating these kinds of structures using microscopic models for the dielectric functions of the cell membrane and the cell interior. In particular, we want to find out whether there are frequencies such that $\mathrm{Re}[\varepsilon_m f(\omega,n_u)]$ changes sign, which would allow for efficient separation of cells with respect to shape. An additional complication in the study of cells is that their membranes are ''soft,'' and hence electric field induced deformations can play a role in their response to an external field. Furthermore, the dielectric function of the cell membrane need not be isotropic.

The method described so far separates particles into *moving* bands (where the particles in different bands have different sizes, shapes, or electric properties). Let us finally discuss the possibility of separating particles into stable (nonmoving) distinct bands, i.e., the particles becoming trapped (having zero drift velocity and not being able to diffuse out of the band) at different positions in the solvent depending on their properties. One such stable band separation technique is based on the idea of using a gradient in the dielectric function of the surrounding medium. As noted above the drift velocity is zero (the particle is ''trapped'') if the frequency of the electric or electromagnetic field equals a resonance frequency in the combined particle-surrounding medium system or if the dielectric function of the particle equals that of the surrounding medium. A dielectric gradient therefore allows separation of neutral particles into stable bands, where the positions of the bands depend on shape or dielectric properties of the particles [12].

We conclude that our scheme allows experimental separation of particles with different lengths of the axes along the direction of motion by using off-resonant electric fields. By applying a field near a frequency ω_0 (for instance a resonance frequency), at which $\mathrm{Re}[\varepsilon_m f(\omega)]$ changes sign [ε_m is the relative dielectric function of the medium and $f(\omega)$ is the frequency dependent part of the polarizability] particles of different shape can be separated.

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APPENDIX: ELLIPTIC SHAPE FUNCTIONS

In this appendix we give the definitions of different entities used in the paper.

Let us first define

$$Q\equiv\frac{b_x b_y b_z}{2}\int_0^\infty\frac{ds}{R(s)},\tag{A1}$$

where

$$R(s)=[(b_x^2+s)(b_y^2+s)(b_z^2+s)]^{1/2},\tag{A2}$$

b_u $(u=x,y,z)$ are the principal radii of an ellipsoid. We also define the so-called *depolarization factors*

$$n_u\equiv\frac{b_x b_y b_z}{2}\int_0^\infty\frac{ds}{(b_u^2+s)R(s)}.\tag{A3}$$

It is straightforward to show that the depolarization factors satisfy the sum rule [24],

$$n_x+n_y+n_z=1.\tag{A4}$$

From this sum rule we directly obtain (by symmetry) the depolarization factors for a sphere: $n_x=n_y=n_z=1/3$.

The entities Q and n_x, n_y, and n_z can be analytically evaluated for a spheroid $b_y=b_z$. In order to be able to evaluate the above integrals, we have to distinguish between two different shapes: *prolate* spheroids $1\geqslant e^2\equiv 1-b_z^2/b_x^2\geqslant 0$ and *oblate* spheroids $e^2\leqslant 0$ (see Fig. 1). In the case of a prolate spheroid we have

$$n_x|_{\mathrm{prolate}}=\frac{1-e^2}{2e^3}\left[\ln\left(\frac{1+e}{1-e}\right)-2e\right].\tag{A5}$$

By the sum rule, we furthermore have

$$n_z|_{\text{prolate}} = n_y|_{\text{prolate}} = \frac{1}{2}(1 - n_x|_{\text{prolate}})$$

$$= \frac{1}{2e^2}\left[1 - \left(\frac{1-e^2}{2e}\right)\ln\left(\frac{1+e}{1-e}\right)\right]. \quad \text{(A6)}$$

In the case of an oblate spheroid, we have ($q^2 = -e^2 \geqslant 0$)

$$n_x|_{\text{oblate}} = \frac{1+q^2}{q^3}(q - \arctan\, q). \quad \text{(A7)}$$

By the sum rule we find

$$n_z|_{\text{oblate}} = n_y|_{\text{oblate}} = \frac{1}{2}(1 - n_x|_{\text{oblate}})$$

$$= \frac{1}{2q^2}\left(\frac{1+q^2}{q}\arctan\, q - 1\right). \quad \text{(A8)}$$

Let us now evaluate Q. Let us start with the case of a prolate shape. Equation (A1) then becomes

$$Q|_{\text{prolate}} = \frac{b_z^2}{2e}\ln\left(\frac{1+e}{1-e}\right). \quad \text{(A9)}$$

In the case of an oblate shape, we get ($q^2 = -e^2 \geqslant 0$)

$$Q|_{\text{oblate}} = \frac{b_z^2}{q}\arctan\, q. \quad \text{(A10)}$$

The results above for Q and n_z, n_y, and n_x completely determine g_u [Eq. (14)] for oblate and prolate shapes.

By the above relations it is possible to relate Q to the depolarization factors. Combining Eqs. (A5), (A7), and (A9), we find

$$\frac{Q}{b_x^2} = e^2(n_x - 1) + 1. \quad \text{(A11)}$$

Notice that this result is valid for both prolate ($e^2 > 0$) and oblate ($e^2 < 0$) shapes. It is also straightforward to show that

$$\frac{Q}{b_z^2} = \frac{1 - 2e^2 n_z}{1 - e^2}. \quad \text{(A12)}$$

Also this result is valid for both prolate and oblate spheroids.

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[17] There is, in general, a gravitational force acting on the particle which (counteracted by buoyancy and viscous forces) leads to a drift velocity $v_y = v_{0,y}\triangle$, where $v_{0,y} \sim L^2 g \rho_{\text{fluid}}/\eta$ (g is the acceleration of gravity and L is a characteristic length of the particle) and the relative density is $\triangle \equiv (\rho_{\text{particle}} - \rho_{\text{fluid}})/\rho_{\text{fluid}}$ [8]. We have above chosen the coordinate system such that the gravitational field is in the y direction. If the relative density is zero ($\triangle = 0$), gravitational and buoyancy forces cancel each other leading to zero drift velocity (as seen in the above equation). For a μm sized particle in water, $v_{0,y} \sim 0.01$ mm/s. The above gravitational contribution to the drift velocity does not appear in Eq. (4) if we choose the external force perpendicular to the gravitational field, i.e., in the x or z direction.

[18] The Fokker-Planck equation (the so-called *Smoluchowski equation*) for $P(\vec{x},t)$, which corresponds to the low Reynolds number Langevin equation, Eq. (3), is (we limit the discussion to 1D motion for the ease of argument) [14]

$$\frac{\partial P}{\partial t} = D\frac{\partial^2 P}{\partial x^2} + v_{\text{drift}}\frac{\partial P}{\partial x},$$

where we have introduced the diffusion constant $D \equiv k_b T/\xi$. Assuming that the particle initially is at $x = 0$ [i.e, $P(x, t = 0) = \delta(x)$], this equation can be solved for the probabilty distribution $P(x,t)$. The result is [14]

$$P(x,t) = \exp[-(x - v_{\text{drift}}t)^2/4Dt]/(4\pi Dt)^{1/2},$$

where $D \equiv k_B T/\xi$ is the diffusion constant. The mean position of the particle is hence $x_{\text{drift}} \equiv v_{\text{drift}}t$. There is also a diffusional spread $\sim\sqrt{Dt}$ which tends spread the positions of the particles around the mean value.

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[20] It is possible to add a constant electric field $\vec{E}_0$ without changing the expression for the force. There are then two consequences that we want to point out: (i) The torque on a particle (in the dipole approximation) is $\vec{\tau}=\vec{p}\times\vec{E}$. Thus by choosing the constant part of the electric field appropriately, one can always align the molecules in any desired direction. (ii) The gradient of the electric intensity gets an extra ''cross-term'' contribution [see Eq. (6)] $\varepsilon\varepsilon_0 E^*_{0,u}\partial E_u/\partial u/2$ (this contribution originates from the fact that the constant field ''helps'' to polarize the particle) compared to a situation without $\vec{E}_0$.

[21] Strictly speaking Eq. (7) is valid only for steady motion. However, it can be shown [8] to be valid also for *unsteady* motion as long as the characteristic time for particle motion is longer than t_{relax}, where $t_{\mathrm{relax}}\equiv\rho_{\mathrm{fluid}}L^2/\eta$ (L is a characteristic length of the particle). For 1 μm sized particles in water at room temperature we have $t_{\mathrm{relax}}\sim 1$ μs. For most practical situations, Eq. (7) is therefore valid also for unsteady motion.

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[25] Any complex dielectric function can be written [28] as

$$\varepsilon(\omega)=\varepsilon^0(\omega)+\frac{i\sigma(\omega)}{\omega},$$

where ε^0 is the dielectric constant and represent the response of bound charges to the external field. $\sigma(\omega)$ is the conductivity that corresponds to the response of free charges. The distinction between free and bound charges is physically meaningful only for the response to DC and low-frequency fields.

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