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Integral effects of the Debye layer on a sedimenting particle with zeta-potential variations

Philipp G. Marthaler[1,*] and **Andreas G. Class**[1,**]

[1] Institute for Thermal Energy Technology and Safety (ITES), Karlsruhe Institute of Technology (KIT), Herrmann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

Fluid-solid interfaces give rise to electro-hydrodynamic effects at microscale. In a macroscale model, those effects can be represented by jump conditions. This report focuses on the derivation of such jump conditions for integral parameters, like mass flux, forces and charge fluxes. In particular, the effect of spatial and temporal variations in the ζ-potential on the solid's surface are discussed, using the generic problem of a sedimenting particle.

The dynamic behavior induced by varying ζ-potentials in electro-hydrodynamic problems is of great interest for both technical applications and life sciences. Examples are lab-on-a-chip-systems or micro-organisms. In technical systems, as discussed e.g. by Ramos et al. [9], research focuses on the design of applied surface potentials in order to optimize the induced flow pattern. The understanding of micro-organisms is based on the interaction of a fluid electrolyte with charged and flexible bio-membranes. One major topic in this context is the understanding of micro-organism motility as e.g. investigated by Stone & Samuel [11].

Time-dependent behavior originates from variations in the surface charge of the corresponding solid or fluctuations of the macroscale flow field. Consequently, the general problem of interest is an arbitrarily shaped solid subjected to a generic surface charge. Following a number of previous publications [4, 6, 12], we investigate the problem of a small particle sedimenting in an electrolyte liquid. The liquid is assumed to be of constant density and viscosity containing two symmetric species of ions. In particular, both ion species have equal diffusion coefficients and opposite charges. The charged surface of the particle is in direct contact with a liquid electrolyte, i.e. a fluid characterized by its flow and pressure field, and ion concentration fields. The electro-hydrodynamic behavior arises from a small layer of dimensionless thickness δ close to the solid's surface.

Singular problems can be modeled by a method originally developed for flame surfaces by Class et al. [5] and adapted to electro-hydrodynamic problems by Marthaler and Class [8]. The method combines a contravariant description of the corresponding complex-shaped surface following Aris [1] with an asymptotic approach based on Bender [2] and Hinch [7] to find analytical solutions for the set of differential equations.

We use the generic conservation equation as starting point of our analysis. Based on our previous work [5, 8], we describe the arbitrary surface of our particle with the contravariant metric $g^{ij}\left(x^j,t\right)$. Due to the assumption, that the surface normal coordinate x^1 is orthogonal to both tangential coordinates x^α at each point of the surface, the simplifications $g^{11}=1$ and $g^{1\alpha}$ hold. With the volume element $\sqrt{g}\left(x^j,t\right)$ the conservation of a generic scalar $a\left(x^j,t\right)$ takes the form

$$\partial_t\left(\sqrt{g}\,a\right)+\partial_{x^j}\left(\sqrt{g}\,J^j\left(a\right)\right)=\sqrt{g}\,S\left(a\right), \tag{1}$$

where $J^j\left(a\right)$ includes all fluxes which can e.g. be specified as convective, diffusive or electrophoretic. The fluxes are balanced by a source term $S\left(a\right)$ and a transient term. By replacing the generic parameter a by the relevant parameters for our example problem, flow velocity $v^j\left(x^j,t\right)$, pressure $p\left(x^j,t\right)$, electric charges of equal valence and opposite sign $c_\pm\left(x^j,t\right)$ and electric potential $\phi\left(x^j,t\right)$,we find a system of charge conservation equations

$$\mathrm{Pe}\,\partial_t\left(\sqrt{g}\,c_\pm\right)+\partial_{x^j}\left(\sqrt{g}\left(-g^{ij}\partial_{x^i}c_\pm+\mathrm{Pe}\,c_\pm v^j\mp c_\pm g^{ij}\partial_{x^i}\phi\right)\right)=0, \tag{2}$$

complemented by momentum and mass conservation and Gauß's law. The Reynolds number in a typical microfluidic problem is small enough to neglect the transient and convective terms in the momentum equation. Thus, it reduces to a balance of Newton and Maxwell stresses, and the only time derivative in the governing equations appears in the charge balance.
At the solid boundary $x^1=0$, no-slip and impermeability conditions hold. Far from the wall $x^1\to\infty$, we have an attenuation condition to the outer flow field. At the boundaries, the electrical problem is governed by

$$J^1\left(c_\pm\right)\big|_{x^1=0}=0,\quad \phi\big|_{x^1=0}=\zeta\left(x^\alpha,t\right),\quad c_\pm\big|_{x^1\to\infty}\to 1,\quad \phi\big|_{x^1\to\infty}\to\phi_\infty. \tag{3}$$

Attenuation conditions for the potential and charges (that take their equilibirum value 1 far from the charged wall) and a no-flux condition at the wall are complemented by a Dirichlet-type boundary condition of the potential. The ζ-potential, representing

* Corresponding author: e-mail philipp.marthaler@kit.edu, phone +49 721 608 25781
** e-mail andreas.class@kit.edu

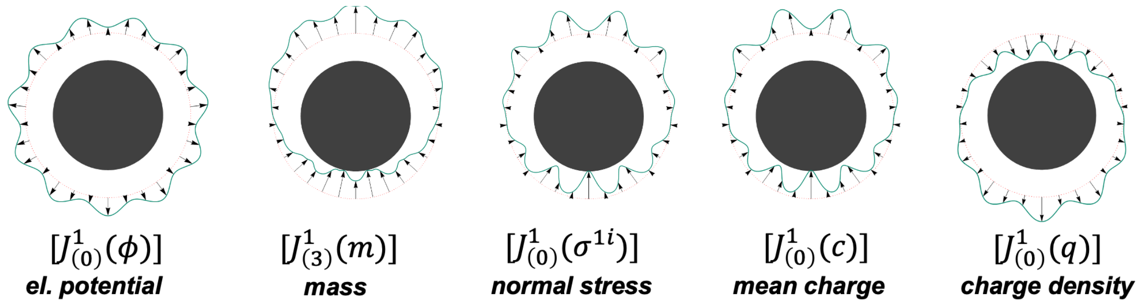

Fig. 1: Qualitative jump conditions for normal fluxes on the sphere surface. The red dotted line represents the value zero. Arrows and green line demonstrate the respective jump conditions at the circumferential position. The reagrding ζ-potential in dimensionless form was chosen as $\zeta = \zeta_0 + \tilde{\zeta}\sin(k\,\theta)$. The used parameters are $\zeta_0 = 1$, $\tilde{\zeta} = 0.1$, wavenumber $k = 11$, and the polar coordinate $\theta \in [0, 2\pi]$. The indices in brackets represent the order of the displyed jump, (3) stands for values of order δ^3. The particle sediments from top to bottom, the value $\theta = 0$ is located in the lee position.

the excitation term in our problem, is assumed to be known. Introducing temporal or spatial varying ζ-potentials, the system's response behavior can be investigated. After introducing the coordinate stretch $x^1 = \delta X$, we compute jump conditions for the integral parameters following the procedures in Marthaler & Class [8] and Class et al. [5]. For the derivation of the general form of the jump condition

$$\sqrt{g}\left[J^1(a)\right] = \int_{X_{\text{surface}}}^{\infty} \left(\sqrt{g}\,(S(A) - S(a)) - \partial_t\left(\sqrt{g}\,(A - a)\right) - \partial_{x^\alpha}\left(\sqrt{g}\,(J^\alpha(A) - J^\alpha(a))\right)\right)\mathrm{d}X \quad (4)$$

we refer to the mentioned publications. The overarching objective of the procedure is the replacement of detailed physics inside the layer by jump conditions. Therefore, the jump condition contains differences of the parameters of the continuous model $A\left(x^j, t\right)$ and the parameters of the jumping model $a\left(x^j, t\right)$. Spatial variations of the ζ-potential along the particle surface are computed with the above relation (4). Examples for the results, considering a stationary case, are displayed in Fig. 1. In particular, the hydrodynamic effects give rise to corrections to the forces acting on the surrounding fluid. The normal stress distribution indicates an extra force acting on the fluid on the floward side of the sedimenting particle and a negative correction on the lee side.

Temporal variations in the ζ-potential are distinguished by their fluctuation frequency. Slow time scales of order $t \sim \delta^{-1}$ or slower let the transient term in the Nernst-Planck equations vanish. This leads to a quasi-stationary system with the well-known Gouy-Chapman solution. Fast time scales of at least$t \sim \delta^0 \tau_{(0)}$ yield the time-dependent basic system

$$\partial_X\left(-\sqrt{g}_{(0)}\partial_X\Phi_{(0)}\right) = Q_{(0)}, \quad \partial_{\tau_{(0)}}\left(\sqrt{g}_{(0)}C_{\pm(0)}\right) + \partial_X\left(-\sqrt{g}_{(0)}\partial_X C_{\pm(0)} \mp \sqrt{g}_{(0)}C_{\pm(0)}\partial_X\Phi_{(0)}\right) = 0, \quad (5)$$

which has to be solved numerically. Developing a numerical code for this problem will increase the understanding for many technical or bio-mechanical problems in the context of electro-hydrodynamics. Yet, the latter requires an extension of the model, incoporating the interaction between soft tissue and the electrolyte. That model extension defines a second roadmap for future research.

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