Hot Swimming using Table Salt: The Theory behind Self-Thermoelectrophoresis

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We analytically and numerically study the self-propulsion of a thermoelectrophoretic colloidal Janus swimmer. We show that experimentally significant propulsion velocities may be achieved using relatively small temperature gradients that couple to monovalent ions dissolved in the suspending medium. Our thin screening-layer theory reveals that the thermoelectric effect is strictly due to the nonlinear coupling between the out-of-equilibrium ion concentrations and electric potential distributions, which stem from the temperature heterogeneity, to the equilibrium electrostatic screening profiles. We obtain excellent agreement between our theory and finite-element calculations in the appropriate limits. We use the latter to also explore the effect of nonlinearity for large Debye lengths, as well as study the flow field around such a swimmer. Our results provide a solid theoretical framework, against which further experiments can be realized and analyzed.

I. INTRODUCTION

Almost a decade and a half ago saw the introduction of the first man-made chemical swimmers, colloidal particles that used catalytic decomposition of hydrogen peroxide ($\text{H}_2\text{O}_2$) to achieve self-propulsion [1, 2]. These Janus swimmers where heralded as artificial model systems for studying the complex motion and cooperative behavior observed in biology [3]; such dynamics have by now indeed been reproduced in man-made systems [4–8]. This collective behavior has been the subject of intense theoretical and numerical effort, \textit{e.g.}, see Refs. [9–11]. Chemical swimmers also opened new avenues for gaining insight into out-of-equilibrium statistical physics [9, 12], as well as provided a foundation for the pursuit of advanced nanorobotic and microsurgical ambitions [13, 14]. These swimmers are foreseen to remain the workhorse of the active matter community for some time.

Nevertheless, despite their success, many questions persist concerning the way $\text{H}_2\text{O}_2$-fueled swimmers self-propel. A large number of experimental unknowns leaves significant freedom in fitting current models. This is a double-edged sword: On the one hand, simple models have been extremely successful in capturing qualitatively the interactions between chemical swimmers and their environment, \textit{e.g.}, see Refs [15–17]. From this one can infer a certain robustness to the observed phenomena, \textit{i.e.}, understanding of the full chemical details appears to be unnecessary to capture trends and behaviors. On the other hand, the exact way the surface and bulk chemistry leads to self-propulsion remains unclear [15, 18–22], especially when it comes to understanding the high reported speeds of nanoscopic swimmers [23, 24]. In addition, $\text{H}_2\text{O}_2$ is not biocompatible, as are many other catalytic fuels that have been considered, \textit{e.g.}, see Refs. [25, 26], limiting the \textit{in vivo} applicability. This has led to the exploration of other self-propulsion strategies, which are biocompatible and/or involve more well-understood chemistry [27, 28].

A promising candidate for biocompatibility is thermophoresis, which utilizes local heating to achieve motion through the migration of solute species in a temperature gradient, known as the Soret effect. This means of propulsion has been studied experimentally by several groups [29–32], who have shown that significant self-propulsion may be achieved. From a theoretical perspective, there remain open questions concerning the microscopic origins of the thermophoretic effect and associated Soret coefficients, with attempts to unify thermophoretic theory for colloidal motion as recent as 2018 [33]. Yet, there are systems for which the theory matches the experiment well, \textit{e.g.}, see Ref. [34]. Significant progress has been made theoretically for a specific driving mechanism, where the dominant contribution to thermophoresis comes from electrostatic interactions, \textit{i.e.}, thermoelectrophoresis [32, 35–38]. In this case, dissolved ions interacting with a charged surface are brought out of equilibrium using a temperature gradient and subsequently induce fluid flow. Thermoelectrophoretic theory has been successfully applied to the describe the motion of “hot swimmers” against gravity [32].

Here, we theoretically examine the thermoelectrophoretic self-propulsion mechanism going, for which we go beyond the approximations made in previous works [32, 35, 38]. We describe in detail the associated equation system and solve it using the finite-element method (FEM). In the small Debye-length limit, we further investigate the motion of a “hot swimmer” in an electrolyte using analytic theory, deriving simple expressions for the speed that we validate against our numerical result. Our FEM calculations allow us to study the nonlinear effects in the large Debye-length limit, where we obtain motility reversals that are reminiscent of external electrophoresis. In general, we obtain self-propulsion...
speeds of a few $\mu\text{m} \text{s}^{-1}$ for physiologically relevant (monovalent) salt concentrations $\approx 1 \text{ mmol} \text{ L}^{-1}$ and small local heating of $\Delta T \lesssim 5 \text{ K}$, which holds promise for biological application. Changing the type of ions and bulk salt concentration also allows for sensitive tuning of the speed and flow field around the hot swimmer, through which it interacts with its surrounding. We conclude by detailing the implications of our findings on thermoelectrophoretic hot swimming in simple salt solutions and how these relate to current and potential future experiments.

II. THE MODEL

We consider a single spherical colloid of radius $a$, with its bottom half coated by a thin metal or carbon cap. The colloid is immersed in an electrolyte, comprised of water and a monovalent salt, with reservoir concentration $n^\infty$ and local ionics $n_\pm$($r$), where $r$ is the position vector. By illuminating the colloid with an appropriately chosen light source, the cap can be heated, which leads to a temperature heterogeneity around the colloid which drives the system out of equilibrium. This causes the colloid to self-propel due to the thermoelectrophoresis, see Fig. 1 for a schematic illustration. Here, we also define our radial $r$ and axial $z$ coordinates (unit vectors $\hat{r}$ and $\hat{z}$, respectively), as well as the polar angle $\theta$.

![Fig. 1. Sketch of a charged Janus particle (axisymmetric around $z$) immersed in an electrolyte with an ambient temperature $T_\infty$. Illumination of the capped hemisphere (light yellow) increases its temperature by $\Delta T$. In steady state, the heating leads to an asymmetric distribution of ions around the colloid, resulting in its self-propulsion. In a frame-of-reference co-moving with the colloid, the fluid velocity is then $U$ at infinity.

The governing equations of our system in steady state are as follows. The temperature distribution throughout the system is given by $T(r)$ and obeys the heat equation,

$$\nabla \cdot \left( k(T(r)) \nabla T(r) \right) = 0,$$  \hspace{1cm} (1)

where $k$ is the thermal conductivity, with $k = k_\text{f}$ in the fluid and $k = k_\text{s}$ for the solid colloid. In Eq. (1), we neglected advection in the fluid phase since the typical $O(\mu\text{m} \text{s})$ velocities of microswimmers lead to small thermal Péclet numbers. Note that we take the thermal conductivity in Eq. (1) to be temperature dependent, with the constitutive relation for $k(T)$ given in section IIIA. The temperature dependence will be considered in this work for all physical properties. We leave the $T(r)$ dependence of all fields, such as the fluid velocity, implicit throughout.

Within a continuum framework, the ion dynamics is captured by the classical Poisson-Nernst-Planck equations. The Poisson equation for the electric potential $\Phi(r)$ reads

$$\nabla \cdot (\varepsilon(T(r)) \nabla \Phi(r)) = -e \left( n_+(r) - n_-(r) \right),$$  \hspace{1cm} (2)

where $\varepsilon$ is the medium’s dielectric permittivity and $e$ is the elementary charge. The Nernst-Planck equations for the ion fluxes are $[35, 37]$

$$j_\pm(r) = -D_\pm(T(r)) \left[ \nabla n_\pm(r) \pm \frac{e n_\pm(r)}{k_B T(r)} \nabla \Phi(r) 
+ 2 n_\pm(r) \alpha_\pm(T(r)) \frac{\nabla T(r)}{T(r)} \right],$$  \hspace{1cm} (3)

where $k_B$ is Boltzmann’s constant, $D_\pm$ are the regular diffusion constants and $\alpha_\pm$ are the thermal diffusion coefficients of the respective ions. The latter are related to the intrinsic Soret coefficients via $S_\pm = 2 \alpha_\pm/T$. The equation system is closed by the ionic conservation laws

$$\nabla \cdot j_\pm(r) + u(r) \cdot \nabla n_\pm(r) = 0,$$  \hspace{1cm} (4)

where $u(r)$ is the fluid velocity.

For a micron-size colloid self-propelling at a speed that is $O(\mu\text{m} \text{s})$, the Reynolds number $Re \ll 1$, and hence the fluid velocities is governed by the Stokes equations for an incompressible fluid.

$$\eta(T(r)) \Delta u(r) - \nabla p(r) = e \left( n_+(r) - n_-(r) \right) \nabla \Phi(r)$$
$$+ \frac{1}{2} \left| \nabla \Phi(r) \right|^2 \frac{\partial (T(r))}{\partial T(r)} \nabla T(r),$$  \hspace{1cm} (5)

$$\nabla \cdot u(r) = 0,$$  \hspace{1cm} (6)

where $\eta$ is the viscosity of the solvent and $p(r)$ is the hydrostatic pressure; $\Delta$ indicates the vector Laplacian. Here, we use in the right-hand side of Eq. (5) the body-force terms derived by Landau and Lifshitz [39] and also employed by Refs. [38, 40–42], where the first term is the electric body force, which implicitly depends on the temperature through the ionic distributions, and the second term is the thermoelectric coupling due to the permittivity dependence on temperature.

The boundary conditions for our problem are the following. On the swimmer, we have a no-slip condition for the fluid velocity, $u(r_s) = 0$, where $r_s$ is a position vector on the surface of the swimmer; $|r_s| = a$. We choose a frame of reference co-moving with the particle such that, the fluid velocity far away from the particle obeys $u(|r| \uparrow \infty) = -U$, with $U$ the swim velocity and $U = U \cdot \hat{z}$ the swim speed. N.B. Our definition of the swim speed allows it to assume negative
values, which we use throughout to help identify the direction of travel. The Poisson equation has the boundary condition that the electrostatic potential decays to zero in the bulk, i.e., $\Phi(\mathbf{r}) \uparrow \infty = 0$. At the surface, we must distinguish between a conductor and an insulator. For the former, we have $\Phi(\mathbf{r}) = \Phi_0(\mathbf{r})$, with $\Phi_0$ the surface potential. For the latter, we have $\mathbf{n}(\mathbf{r}_s) \cdot \nabla \Phi(\mathbf{r}) \bigg|_{\mathbf{r}=\mathbf{r}_s} = -\sigma(\mathbf{r}_s)/\epsilon(\tau_0(\mathbf{r}_s))$, where $\sigma$ is the surface charge density and $\mathbf{n}$ is the outward unit normal to the surface. The salt concentrations at the edge of the system assume their reservoir value, i.e., $\mathbf{n}(\mathbf{r}_s) \cdot j_\pm(\mathbf{r}_s) = 0$.

Finally, for the heat equation, the temperature far away is given by the reservoir temperature $T(\mathbf{r}) \uparrow \infty = T^\infty$. For the capped surface we must distinguish between constant heat flux and constant temperature, respectively. When the thermal conductivity of the coating $k_{\text{cap}}$ is much larger than that of the fluid and solid colloid, $k_t$ and $k_s$, respectively, there is a constant temperature on the lower hemisphere $T(\mathbf{r}_s) = T^\infty + \Delta T$, with $\Delta T$ the excess temperature induced by heating. This occurs, for instance, for a metallic cap [6, 30]. When thermal conductivity of the coating is much smaller that of the fluid and colloid, $k_{\text{cap}} \ll k_t$, $k_s$, e.g., for a carbon coating [43], heat is immediately conducted to the surroundings such that the illumination leads to a constant heat flux $q$ through the cap. In this case, the boundary condition reads $k_t \mathbf{n}(\mathbf{r}_s) \cdot \nabla T(\mathbf{r}) \bigg|_{\mathbf{r}=\mathbf{r}_s} - k_s \mathbf{n}(\mathbf{r}_s) \cdot \nabla T(\mathbf{r}) \bigg|_{\mathbf{r}=\mathbf{r}_s} = q(\mathbf{r}_s)$.

On the top (uncapped) half of the colloid we have the flux continuity condition $k_t \mathbf{n}(\mathbf{r}_s) \cdot \nabla T(\mathbf{r}) \bigg|_{\mathbf{r}=\mathbf{r}_s} = k_t \mathbf{n}(\mathbf{r}_s) \cdot \nabla T(\mathbf{r}) \bigg|_{\mathbf{r}=\mathbf{r}_s}$.

The system of Eqs. (1)–(6) with the appropriate boundary conditions was solved numerically using the finite element software COMSOL Multiphysics to obtain the self-propulsion speed of the particle. The results of the numerical calculations were compared with those of the linear theory presented in the next section.

III. LINEAR THEORY

To gain deeper insight into our system, we derive expressions for the speed of the thermoelectrophoretic swimmer $U$ by linearizing Eqs. (1)–(6). The approach we employ is similar to that of Ref. [20], but applied here also to the temperature dependencies. Linearization is not required, however, to establish the temperature profile. This can be obtained using the calculations presented in Ref. [44]; for completeness we provide the relevant expressions in our notation in appendix A.

A. Linearizing the Equations

Our first linearization is of the electrostatic potential and the ion distributions, i.e., we make the usual Debye-Hückel approximation. We write

$$n_\pm(\mathbf{r}) = n^\infty \left(1 \pm x_\pm(\mathbf{r})\right),$$  

$$\Phi(\mathbf{r}) = \frac{k_B T^\infty}{e} \phi(\mathbf{r}),$$

where $x_\pm(\mathbf{r})$ and $\phi(\mathbf{r})$ are the dimensionless, linearized ion distributions and potential, respectively.

Our second linearization decomposes the fields and physical quantities into equilibrium ("eq") and non-equilibrium ("neq") parts, where the non-equilibrium parts are due to variations in temperature. Here, we shall expand in the small parameter $\tau = \Delta T/T^\infty$, corresponding to the relative maximum temperature difference $\Delta T$ from the reservoir temperature $T^\infty$. Note that $\tau$ is well-defined for both equi-temperature and equi-flux surfaces; this choice allows us to write to the temperature distribution as

$$T(\mathbf{r}) = T^\infty \left(1 + \tau t(\mathbf{r})\right),$$

where $t(\mathbf{r})$ is the dimensionless temperature. Similarly, for the other physical fields the decomposition yields: $x_{\pm}(\mathbf{r}) = x_{\pm}^\text{eq}(\mathbf{r}) + \tau x_{\pm}^\text{neq}(\mathbf{r})$, $\phi(\mathbf{r}) = \phi^\text{eq}(\mathbf{r}) + \tau \phi^\text{neq}(\mathbf{r})$, $\mathbf{u}(\mathbf{r}) = \tau \mathbf{v}(\mathbf{r})$, and $p(\mathbf{r}) = p^\text{eq}(\mathbf{r}) + \tau p^\text{neq}(\mathbf{r})$. Notice that in equilibrium there is no fluid flow, hence we only have the out-of-equilibrium velocity component. The physical quantities are expanded as: $\epsilon/\epsilon^\infty = 1 + \tau \epsilon^\text{eq}(\mathbf{r}) + \tau \epsilon^\text{neq}(\mathbf{r})$, $T/T^\infty = 1 + \tau T^\text{eq}(\mathbf{r}) + \tau T^\text{neq}(\mathbf{r})$. Here, the "∞" superscript denotes the reservoir value and the "∗" superscript the first-order Taylor expansion coefficient. We have numerically verified that all starred quantities are order unity and that the non-equilibrium fields are much smaller than the equilibrium contributions, see appendix B. To simplify the equations further, we also introduce the conjugate variables to the ionic distributions, see appendix B. To simplify the equations further, we also introduce the conjugate variables to the ionic distributions, see appendix B. To simplify the equations further, we also introduce the conjugate variables to the ionic distributions, see appendix B. To simplify the equations further, we also introduce the conjugate variables to the ionic distributions, see appendix B. To simplify the equations further, we also introduce the conjugate variables to the ionic distributions, see appendix B. To simplify the equations further, we also introduce the conjugate variables to the ionic distributions, see appendix B. To simplify the equations further, we also introduce the conjugate variables to the ionic distributions, see appendix B.
The Stokes equations read

\[ \eta^\infty \Delta \mathbf{u}(r) - \nabla p_{neq}(r) = \]

\[ 2k_BTn^\infty n^\infty \left[ \delta X_{neq}^\infty(r) \nabla \phi_{eq}^\infty(r) - \phi_{eq}^\infty(r) \nabla \phi_{neq}^\infty(r) \right] \]

\[ - \frac{1}{2} (\lambda^\infty)^2 c^* \left( \nabla \phi_{eq}^\infty(r) \right)^2 \nabla t(r) , \] (12)

\[ \nabla \cdot \mathbf{u}(r) = 0 , \] (13)

where we used \( \partial \epsilon/\partial T = \epsilon^* \epsilon^*/T^\infty \) and introduced the Debye length \( \lambda^\infty \equiv 1/\kappa^\infty \). Finally, the ionic fluxes become

\[ \mathbf{j}_{neq}^\pm(r) = -D^\infty n^\infty [\nabla X_{neq}^\infty(r) \pm \nabla \phi_{neq}^\infty(r) \]

\[ + 2 \left( 1 \mp \phi_{eq}(r) \right) \alpha^\infty \nabla t(r) \]

\[ \mp t(r) \nabla \phi_{eq}^\infty(r) , \] (14)

\[ \nabla \cdot \mathbf{j}_{neq}^\infty(r) = 0 , \] (15)

where we have employed the low-Péclet-number approximation, ionic diffusion dominates advection, see appendix B for the justification.

By adding and subtracting the flux expressions in Eq. (14), and employing the conservation equations (15), we obtain

\[ \nabla^2 X_{neq}^\infty(r) = \beta \nabla t(r) \cdot \nabla \phi_{eq}^\infty(r) ; \] (16)

\[ \nabla^2 \delta X_{neq}^\infty(r) + \nabla^2 \phi_{eq}^\infty(r) = (1 + \gamma) \nabla t(r) \cdot \nabla \phi_{eq}^\infty(r) \]

\[ + (\kappa^\infty)^2 t(r) \phi_{eq}^\infty(r) , \] (17)

where we have used \( \nabla^2 t(r) = 0 \) and introduced

\[ \beta \equiv \alpha^\infty - \alpha^\infty , \] (18)

\[ \gamma \equiv \alpha^\infty + \alpha^\infty , \] (19)

with \( \beta \) commonly referred to as the (reduced) Seebeck parameter.

Several points are worth noting for the linearized equations. (i) At linear order in \( \tau \), only the term involving \( \epsilon^* \) in Eq. (10) introduces the temperature dependence of the physical properties into the equations. This is due to a dielectric-permittivity dependence of thermoelectrophoresis has been reported in the literature [40–42, 45]. (ii) The expressions for the non-equilibrium flux contain two temperature-related terms. One coupling the gradient of the temperature to the equilibrium ion distributions via the thermal diffusion constant. The other coupling the temperature itself to the gradient of the equilibrium electrostatic potential, via the ionic mobilities [36, 37]. The latter is represented by the term on the third line of Eq. (14). Ultimately, this implies that there can be thermoelectrophoretic swimming without any thermal-diffusion \( \alpha_{\pm} \)-related effect. From Eq. (17) it follows that a non-equilibrium ionic excess will be present even if the thermal diffusion coefficients are zero, in agreement with Refs. [36, 37]. (iii) Equations (16) and (17) reveal that the cross coupling between temperature fields and equilibrium ionic screening is crucial to obtain thermoelectrophoresis. If we ignore such cross terms, only the trivial solution is obtained. This intrinsic non-linearity complicates obtaining solutions using standard spectral methods. (iv) The fields \( \delta X_{neq}^\infty(r) \) and \( \phi_{neq}^\infty(r) \) form a closed subsystem of equations, to linear order in \( \tau \). The non-equilibrium ion concentration \( X^\infty_{neq} \) in Eq. (16) is only due to coupling between the temperature gradient and the equilibrium ion potential. It is weighted by the difference in thermal diffusivity \( \beta \), meaning that \( X^\infty_{neq} \) vanishes, when there is no thermal-diffusion-based ion accumulation in the double layer (\( \alpha^\infty = \alpha^\infty = 0 \) or \( \alpha^\infty = \alpha^\infty \)). The closed subsystem is the only non-equilibrium part that then remains.

B. The Thin-Screening-Layer Approximation

The above nonlinear system is solved in the thin-screening-layer limit; analytic progress can be made in the thick-screening limit, but is not discussed here. In the former limit, the electrostatic screening length \( \lambda^\infty \) is small compared to the particle radius, \( \kappa^\infty a \gg 1 \). Outside ("out") of the screening layer, we have \( \nabla^2 X_{neq}^\infty(r) = 0 \) and \( \nabla^2 \phi_{neq}^\infty(r) = 0 \), since \( \phi_{eq}^\infty(r) = 0 \) in this region and \( \delta X_{neq}^\infty(r) = 0 \), because any excess charge is screened. This implies that the only solutions for the potential and total ionicity permissible in the region outside of the double layer have a Laplace form. Using the expressions for the fluxes Eq. (14), one finds that \( X_{neq}^\infty(r) = -\gamma t(r) \) and \( \phi_{neq}^\infty(r) = -\beta t(r) \). That is, the thermoelectric effect causes the total ionicity to vary in the bulk and sets up an unscreened electric potential. The latter exactly balances the fluxes of ions induced by thermal diffusion, countering any charge separation in the bulk, in agreement Ref. [35].

Now let \( \lambda^\infty q \) measure distance in the direction perpendicular to the surface, with \( q = 0 \) for \( r = a \). In the thin-screening-layer approximation, the curvature of the sphere can locally be ignored. We can then split the solutions into parallel and perpendicular components: \( \phi_{neq}(r_s, q) = \phi(r_s) e^{-q} \) and \( t(r_s, q) = t(r_s) \). Here, \( \phi(r_s) \) is the potential at the surface and \( t(r_s) \) is the temperature at the surface. On the size of the screening layer, the temperature can be approximated to be radially constant because the temperature in the fluid decays with a power law of leading order \( a/r \).

Applying the coordinate transformation inside the screening layer, we have for the Laplacian: \( \nabla^2 = \nabla^2_{\parallel} + (\kappa^\infty)^2 \partial^2_{\parallel} \), with \( \nabla^2_{\parallel} \) the gradient in the tangent plane to \( r_s \). A similar transformation for the gradients — taking the limit \( \lambda^\infty \downarrow 0 \) — leads to the following transformed
ionicity and ion excess

\[ \partial_q^2 X_{\text{in}}^{\text{neq}}(r_s, q) = 0; \]  
\[ \partial_q^2 \phi_{\text{in}}^{\text{neq}}(r_s, q) + \partial_q \phi_{\text{in}}^{\text{neq}}(r_s, q) = t(r_s) \phi(r_s) e^{-q}, \]  
with corresponding Poisson equation

\[ \partial_q^2 X_{\text{in}}^{\text{neq}}(r_s, q) + \epsilon^* t(r_s) \phi(r_s) e^{-q} = -\delta X_{\text{in}}^{\text{neq}}(r_s, q), \]  
where the subscript “in” is used to indicate that these fields are within the thin screening layer. Since only derivatives with respect to \( q \) remain in Eqs. (20)–(22), they can be solved using separation of variables.

The limit \( |r_s| \downarrow a \) for the solutions outside of the screening layer gives a set of boundary conditions for the solution inside. Note that by construction this corresponds to \( q \uparrow \infty \). Taking this limit within the layer, we find

\[ X_{\text{in}}^{\text{neq}}(r_s, q \uparrow \infty) = -\gamma t(r_s^+), \quad \delta X_{\text{in}}^{\text{neq}}(r_s, q \uparrow \infty) = 0, \]  
and

\[ \phi_{\text{in}}^{\text{neq}}(r_s, q \uparrow \infty) = -\beta t(r_s^+). \]  
Here, the value of the right-hand side is evaluated at the edge of the double layer.

For the boundary conditions at the surface of the particle (\( q = 0 \)), we linearize the conducting and insulating conditions, leading to \( \phi_{\text{in}}^{\text{neq}}(r_s, 0) = 0 \) and \( \partial_q \phi_{\text{in}}^{\text{neq}}(r_s, 0) \mid_{q=0} = 0 \), respectively. For a conductor, the equilibrium part of the field accounts fully for the surface potential, \( \phi_{\text{in}}^{\text{eq}}(r_s, 0) = \phi_0(r_s) \), with \( \phi_0(r_s) \) the reduced surface potential. For an insulator, the boundary condition of the equilibrium potential

\[ \partial_q \phi_{\text{in}}^{\text{eq}}(r_s, q) \mid_{q=0} = -\lambda^\infty \frac{e \sigma(r_s)}{k_B T \infty}, \]  
(23)
covers any surface charge present. N.B. Here, we do not consider temperature-dependent charge regulation.

The above conditions, together with the linearized Eqs. (20)–(22), lead to the following solutions within the screening layer. As the temperature decays very little inside the screening layer, the added total ionicity due to the heating therein is therefore \( X_{\text{in}}^{\text{neq}}(r_s, q) = -\gamma t(r_s) \). The effect on the net salt concentration in the double layer is, however, sufficiently small that local corrections to the Debye length do not have to be accounted for, since \( X_{\text{in}} = X_{\text{in}}^{\text{eq}} + \tau X_{\text{in}}^{\text{neq}} \) with \( \tau \ll 1 \). The non-equilibrium ion excess decays with \( q \), and we must consider conducting (equipotential) and insulating (fixed charge) surfaces separately.

For an equipotential surface, we find

\[ \delta X_{\text{in}}^{\text{neq}}(r_s, q) = -\beta t(r_s) e^{-q} \]
\[ + \frac{1}{2} \phi(r_s) t(r_s) (2 - (1 + e^*) (q) e^{-q} \]  
(24)
where the electrostatic potential is given by

\[ \phi_{\text{in}}^{\text{neq}}(r_s, q) = -\beta t(r_s) (1 - e^{-q}) \]
\[ + \frac{1}{2} \phi(r_s) t(r_s) (1 + e^*) q e^{-q} \]  
(25)
such that \( \phi(r_s) = \phi_0(r_s) \), because \( \phi_{\text{in}}^{\text{eq}}(r_s, 0) = 0 \). The Seebeck effect thus leads to the development of a surface thermocharge, which is given by \( \delta X_{\text{in}}^{\text{neq}} = (\phi_0 - \beta t) \), as follows from Eq. (24). This expression differs from the one found by Majee and Wirger [35] in that we include a non-zero imposed surface potential. Moreover, by the surface thermocharge, we mean the charge that is imposed directly at the surface, rather than the integral form that is employed in Refs. [35, 38], which gives the effective bulk thermocharge built up around the swimmer due to thermophoresis. The charging for \( \beta = 0 \) follows from the right-hand side of Eq. (21). Suppose for convenience that \( e^* = 0 \) then Eqs. (21) and (22) combine to give an inhomogeneous Helmholtz equation for \( \delta X_{\text{in}}^{\text{neq}} \), for which the particular solution exactly corresponds to minus the right-hand side of Eq. (21). Physically, the effect is due to the difference in temperature dependence between regular diffusion and electric migration [36, 37], which leads to a surface (and bulk) thermocharge on top of that induced by the Seebeck effect [35, 38].

For a fixed surface charge, we find

\[ \delta X_{\text{in}}^{\text{neq}}(r_s, q) = \frac{1}{2} \phi(r_s) t(r_s) (2 - (1 + e^*) (1 + q)) e^{-q} \]
(26)
where the associated electrostatic potential reads

\[ \phi_{\text{in}}^{\text{neq}}(r_s, q) = -\beta t(r_s) + \frac{1}{2} \phi(r_s) t(r_s) (1 + e^*) (1 + q) e^{-q} \]
(27)
and we have via Eq. (23) that

\[ \phi(r_s) = \phi_0(r_s) \equiv \frac{\lambda^\infty e \sigma(r_s)}{k_B T \infty}; \]
(28)
with \( \sigma(r_s) \) the surface charge density. Again there is a thermocharging effect \( \delta X_{\text{in}}^{\text{neq}}(r_s, q) \neq 0 \) as described in Ref. [35, 38]. However, due to the difference in boundary conditions, there is only a non-Seebeck contribution, which means that uncharged surfaces cannot pick up a surface thermocharge. However, they may pick up a bulk thermocharge, as follows from using the definition in Refs. [35, 38]. In general, we will restrict our analysis to the case \( \phi(r_s) \neq 0 \).

C. The Thermoelectrophoretic Swimming Speed

To compute the swim speed, we do not follow the standard thin slip-layer approach [46]. The force contributions parallel to the surface lead to speeds that are comparable in magnitude to the ones that result from the perpendicular components, see appendix D. This implies that the slip-layer reduction of separating out parallel flow and perpendicular pressure contributions to the Stokes equations is not applicable. Instead, we employ Teubner’s method [47] of integrating the (out-of-
equilibrium) body force density with an integration kernel \( K(r) \) to obtain the reduced swim speed

\[
\tilde{U} = -\frac{1}{6\pi\eta a} \int_V K(r) \cdot f^{neq}(r) \, dr; \tag{29}
\]

\[
K(r) = \left( \frac{3a}{2r} - \frac{a^3}{2r^3} - 1 \right) \cos \theta \hat{r} - \left( \frac{3a}{4r} + \frac{a^3}{4r^3} - 1 \right) \sin \theta \hat{\theta}, \tag{30}
\]

where integration takes place over the volume \( V \) outside of the particle. The integral in Eq. (30) can be split into a part inside and outside of the screening layer. Then, an expansion in terms of \( \lambda^\infty \) is performed on the integration kernel \( K(r) \) for the inner part, and the perpendicular and parallel components are computed separately. We also assume that the swimmer is either entirely an equipotential surface, or an entirely insulating surface such that only the first term in the Legendre-Fourier modes of the temperature expansion contributes to the speed generation, see appendix A. The laborious calculation for the swim speed is reproduced in appendix E.

Grouping the expressions for the individual contributions together, we obtain for the total speed of a swimmer

\[
\tilde{U} = -\frac{k_B T^\infty n^\infty}{6\pi\eta a} (\lambda^\infty)^2 \left[ 8\beta \phi_i - (1 - \epsilon^*) \phi_i^2 \right] \tilde{t}_1, \tag{31}
\]

to leading order in \( \lambda^\infty \), with \( \phi_i = \phi_0 \) for the conducting surface and \( \phi_i = \phi_s \) for the insulating surface, respectively. Surprisingly, we find that the form of the speed is independent of the surface type, despite the large difference in the intermediate expressions in our derivation (appendix E). We will compare the above expression to other results found in the literature in section V.

The dimensionful expressions for the thermoelectrophoretic self-propulsion speed can be obtained by multiplying \( \tilde{U} \) with \( \Delta T/T^\infty \). They read

\[
U = -\frac{k_B T^\infty n^\infty}{6\pi\eta a} (\lambda^\infty)^2 \left[ 8\beta \phi_i - (1 - \epsilon^*) \phi_i^2 \right] \frac{\Delta T}{T^\infty} \text{ isothermal cap} + \frac{3\pi a q}{4k_f(2 + K)T^\infty} \text{ constant heat flux cap}, \tag{32}
\]

where we made use of Eqs. (A7) and (A9) in appendix A. Note that the dimensionful expression for the constant heat flux condition \( q \) is not dependent on the particle radius \( a \) here. However, this is not the case in practice, since typically \( q \) depends on \( a \). In general, \( q \propto I \sigma_{abs}/a^2 \), where \( I \) is the illumination intensity and \( \sigma_{abs} \) is the absorption cross section. The dependence of \( \sigma_{abs} \) on \( a \), however, is non-trivial. \( \sigma_{abs} \propto a^3 \) for small particles with \( a \sim O(0.01 \mu m) \), while for big particles with \( a \sim O(10 \mu m) \), \( \sigma_{abs} \propto a^2 \) [48]. Therefore, \( q \) varies from \( q \propto I/a \) to \( q \propto I \), while being more complex in between. We only consider a fixed value of \( a \) here and will ignore such dependencies in the following.

IV. RESULTS

In this section, we discuss our numerical FEM results and show that these correspond to the expressions of our analytic calculations in the appropriate limits. We will predominantly use dimensionful units to make the connection with experiments and to highlight those regimes wherein we expect measurable results.

![FIG. 2. Contour plot of the excess temperature \( t\Delta T \) around a hot \((K = 1)\) Janus swimmer in the \(xz\)-plane. In the left halfplane, we show \( t \) when the heat flux \( Q \) is fixed on the cap (white, \( z \leq 0 \)), such that the maximum temperature on the bottom hemisphere is \( \approx 5 \) K. The right halfplane shows the temperature field with \( \Delta T = 5 \) K fixed on the heated cap.](image)

Throughout, we assume a colloidal particle diameter of \( 1 \mu m \). We consider three types of swimmer material for the hot swimmers: no conductivity contrast with water \( K = 1 \); polystyrene (PS), \( K_{PS} \equiv k_{PS}/k_f = 0.0847 \); and silica (\( \text{SiO}_2 \)) \( K_{\text{SiO}_2} = 2.34 \). For the fluid, we use the physical properties of water at \( T^\infty = 298.15 \) K (room temperature): \( \epsilon^\infty = \epsilon_0 \epsilon_r \) with \( \epsilon_0 \) the vacuum and \( \epsilon_r = 78.4 \) the relative permittivity, \( \eta^\infty = 8.9 \cdot 10^{-4} \) Pa.s, and \( k^\infty = k_f = 0.591 \) W m\(^{-1}\) K\(^{-1}\) [49]. The ambient pressure is specified to be \( p^\infty = 1 \cdot 10^5 \) Pa, approximately one atmosphere.

We consider three types of ions to determine the effect of thermoelectrophoresis, one cation, sodium \( \text{Na}^+ \), and two anions, chloride \( \text{Cl}^- \) and hydroxide \( \text{OH}^- \). This choice is based on the commonplaceness of these ions, as well as the fact that the \( \text{Cl}^- \) anion has a much smaller Soret coefficient than \( \text{OH}^- \), allowing us to probe the effect thereof on the motion of the swimmer. The ionic diffusion coefficients are \( D_{\text{Na}^+} = 1.3 \cdot 10^{-9} \) m\(^2\) s\(^{-1}\), \( D_{\text{OH}^-} = \ldots \).
FIG. 3. The net charge $\delta X = \delta X^{\text{eq}} + \tau \delta X^{\text{neq}}$ along the particle contour parameterized by the polar angle $\theta$. The curves are for a Janus swimmer with $\Delta T = 5 K$, $K = 1$, and an equipotential surface with $\phi_0 = 0.05$ (a) and $\phi_0 = 0.5$ (b); this corresponds to $\approx 2.6 \text{ mV}$ and $\approx 13 \text{ mV}$, respectively.

$2.0 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$, and $D^\infty_{\text{Cl}^-} = 5.3 \cdot 10^{-9} \text{ m}^2 \text{ s}^{-1}$ [50]. The thermal diffusion coefficients are given by $\alpha^\infty_{\text{Na}^+} = 0.7$, $\alpha^\infty_{\text{Cl}^-} = 0.1$, $\alpha^\infty_{\text{OH}^-} = 3.4$ [35, 51, 52]. In all cases, we set $\epsilon^* = 0$, dropping any thermal polarization effects, in order to facilitate the discussion of the results. The actual value $\epsilon^* \approx 1.3$, see appendix B, would give an appreciable contribution to the swim speed whenever $\beta = 0$, see Eq. (32). As we will see, we obtain good correspondence between our numerical results and analytic expressions in the appropriate limits. The case $\epsilon^* \neq 0$ may thus be readily analyzed using the analytic theory.

Let us first examine the temperature profile around a heated ($K = 1$) swimmer. Fig. 2a shows the temperature excess for both types of thermal boundary condition, where we chose the heat flux such that the maximum deviation from the reservoir temperature, $\Delta T = 5 K$, is comparable to the imposed excess temperature for the equi-temperature surface, $\Delta T = 5 K$. The two temperature fields differ only slightly, and we will therefore focus on constant-heat-flux boundary conditions in the following, unless stated otherwise.

Next, we turn our attention to the net charge at the surface of a hot ($K = 1$) swimmer with equipotential boundary condition, see Fig. 3. When the particle is not heated ($\Delta T = 0 K$), $\delta X$ is fixed, and equal and opposite to the imposed value of $\phi_0$ in our approximation. The agreement is good for $\phi_0 = 0.05$ (in the linear regime), but there is an appreciable nonlinear effect for $\phi = 0.5$. Heating of the particle in a $1 \text{ mmol L}^{-1}$ NaOH solution leads to an increase in the anion concentration at the hot surface. Recall that for $\epsilon^* = 0$ the thermocharge at the surface is given by $\delta X^{\text{neq}} = (\phi_0 - \beta)t$ to first order, see Eq. (24). Here, $\beta = -2.7$ and $\phi_0 = 0.05$, which gives $\delta X^{\text{neq}} = 2.75t$, and $\phi_0 = 0.5$, which gives $\delta X^{\text{neq}} = 3.2t$, respectively. Hence, we expect $\delta X$ to increase at the heated cap — it is nearly constant over that hemisphere — and be minimal at the pole of the particle, where the surface temperature is the lowest. The predictions of our linearized theory are qualitatively correct for both potentials, but we only have quantitative agreement for $\phi = 0.05$. The thermocharging effect is much smaller for a $1 \text{ mmol L}^{-1}$ NaCl solution due to the smaller Soret coefficient of the Cl$^-$ anion ($\beta = 0.6$); here we find $\delta X^{\text{neq}} = (\phi_0 - \beta)t = (0.05 - 0.6)t = -0.55t$ and $(0.5 - 0.6)t = -0.1t$, respectively. In the linear regime, our theory predicts the correct sign change of the thermocharging with respect to the NaOH solution, but in the nonlinear regime there is no qualitative agreement. The reason for this is revealed by examining the situation where no Soret effect is included (purple curve). Here, we should obtain $\delta X^{\text{neq}} = \phi_0t = 0.05t$ and $0.5t$, respectively. Clearly, the effect of nonlinearity is much stronger for the $\beta = 0$ thermocharging.

One of the most important properties of the swimmer is the flow field generated by the non-equilibrium effect, as this governs to first order the interaction of the swimmer with its environment. This aspect was previously explored by Bickel et al. [44] for a hot swimmer that had a Seebeck-related slip velocity. Here, we include all terms in our equations and let go of the slip-layer approximation. Fig. 4 shows representative flow fields for several swimmer and environmental configurations. We find that by lowering the ionicity the puller type flow is suppressed, leaving a more neutral-squirmer flow field, see Fig. 4a for the effect for a hot swimmer in a NaOH solution. Changing the anion type and leaving the other parameters the same can be used to change the direction of motion and to change from a puller- to a pusher-type
FIG. 4. Fluid velocity magnitude $|u|$ divided by the absolute swim speed $|U|$ and streamlines for several hot ($K = 1$) swimmers in the laboratory frame of reference. In (a), the electrostatic potential is fixed at $\phi_0 = 0.5$ ($\approx 13$ mV) and we impose $\Delta T = 5$ K at the heated cap. The concentration of NaOH is $1$ mmol L$^{-1}$ in the left panel and $1 \cdot 10^{-3}$ mmol L$^{-1}$ in the right panel. In (b), the surface charge of the particle is fixed at $5 \cdot 10^{-3} e$ nm$^{-2}$ and $\Delta T = 5$ K. The electrolyte in the left panel is $1$ mmol L$^{-1}$ NaOH and, while in the right panel it is $1$ mmol L$^{-1}$ NaCl. Notice the opposite direction of the streamlines in the two panels as the swimmers translate in opposite directions. The large arrows in the center of the swimmer indicate the direction of motion.

flow field, thereby strongly modifying the interaction of the hot swimmer with its environment.

We will explore the dependence of the swim speed in more detail in the following. Fig. 5 shows the swim speed as a function of the bulk salt concentration for four representative swimmer/salt combinations and an equipotential boundary condition. The effect of the difference in thermal conductivity is quantitative, leading to an appreciable increase in absolute speed with reduced $K$. The direction of swimming is reversed between the two types of salt, as previously shown in Fig. 4. In all cases we obtain significant swimming speeds, several µm s$^{-1}$, in physiological to high salt concentrations. Note that we accurately capture the analytic limit for our equipotential swimmer, despite the fact that we do not resolve the thermocharge correctly, see Fig. 3. In the analytic theory we find that for such a swimmer $U \propto n^\infty (\lambda^\infty)^2 \propto 1$ (in terms of $n^\infty$). Our result implies that the swim speed is independent of the reservoir concentration to first order. This is borne out by our numerical data in Fig. 5, which is almost constant over a large range in $n^\infty$. Higher-order terms, proportional to $n^\infty (\lambda^\infty)^3$ etc., would capture the departures from the constant value of $U$ near the limit $\lambda^\infty \downarrow 0$. Physically, the constant speed of the equipotential swimmer in this limit is due to the small variation of the ionic profiles across the screening layer, over which the temperature hardly varies. The majority of the out-of-equilibrium nature of the ion profiles is imposed in the bulk, due to the temperature variation therein whenever $\beta \neq 0$. In those cases that $\beta = 0$, the ion variation is in the bulk couples back to the surface, leading to a dependency $U \propto \phi^2_0$, but with the same constancy in $n^\infty$.

Fig. 6 shows the effect of the electrostatic boundary condition and the Soret effect on the motion of the hot swimmers as a function of the bulk salt concentration. Comparing the two panels of Fig. 6 we find a significant impact of the surface properties of the swimmer.
FIG. 6. Silica swimmer speed $U$ as a function of salt concentration $n^\infty$ for two electrolytes, NaCl (red) and NaOH (black), and when the Soret effect is neglected ($\beta = \gamma = 0$; blue). In (a) we use an equipotential electrostatic boundary condition with $\phi_0 = 0.5 ~\text{(}\approx 13 \text{mV)}$ while in (b) we used a constant surface charge one $\sigma = -5 \times 10^{-3} e \text{nm}^{-2}$. In both panels $\Delta T = 5 \text{K}$ is fixed at the heated cap. The dashed lines indicate the prediction of our analytic theory.

V. DISCUSSION AND OUTLOOK

Summarizing, we have numerically and analytically computed the self-thermoelectrophoretic propulsion speed of a hot swimmer for various boundary conditions and environmental parameters. Here, we have taken into account the difference in temperature response between the diffusion coefficient and the ionic mobilities [36, 37], which leads to contributions that scale with the zeta potential squared. In addition, we have explored the limit of large electrostatic screening layers around the colloid numerically. In the limit of small Debye length, we obtain excelled agreement for the swim speed between our analytic theory and numerical calculations. Interestingly, this proved to be the case even in a limit where our linearized theory did not accurately capture the thermocharging effect.

For an equipotential boundary condition, we obtain $\mu m s^{-1}$ swimming speeds, in physiological to high (monovalent) salt concentrations $n^\infty \gtrsim 1 \text{mmol L}^{-1}$, which are nearly independent of the salt concentration, in the thin screening-layer limit. For an insulating swimmer propulsion speeds are low in this regime and they drop off with increasing bulk ionicity, while they are constant and appreciable in the limit of large Debye length. The direction of swimming can change between these two limits, reminiscent of mobility reversals found in external electrophoresis [53]. Finally, we have show that the flow field around the swimmer is similarly sensitive to the bulk ionicity. This could potentially be exploited in studies of the collective dynamics of self-thermoelectrophoretic swimmers to fine tune their long-ranged interactions.

Recently, Ly et al. [38] performed similar analytic calculations of thermoelectrophoresis, but they only focused on the local slip velocity, rather than the swim velocity that this results in. Like these authors, we observe that there is a substantial difference in the dependence of the potential and ion excess on the temperature for insulating and conducting surfaces, while the (in our case) total speed has an identical form when expressed in terms of the surface potential. However, unlike standard practice in the literature, we accounted for the tangential component to the force density acting over the slip layer in our calculations to arrive at this result, following the ap-
approach of Refs. [20, 47] instead. This raises the question whether Teubner’s expressions for the propulsion speed [47] need to be applied in other settings where the slip-layer is thin, but there nonetheless exists the possibility of significant tangential forcing, due to a complicated non-linear coupling resulting in phoretic motion.

In relation to experiments, our work predicts opposite trends in swim speed with varying bulk ionicity for conducting and insulating surface boundary conditions, respectively. To the best of our knowledge this limit has not yet been systematically explored. Simoncelli et al. [32] observed a decrease of the height above the surface for their thermophoretic swimmers with increased salt concentration, which might be explained in terms of such a thermoelectrophoretic effect. These authors also observe a decreased height by switching from NaCl to NaOH, which could be related to a reversal in swim direction when using this salt [32], potentially in agreement with our prediction of such an inversion. However, the combined thermophoretic and thermoosmotic contributions from the presence of a wall [32], as well as the wall’s own response to changes in pH and salt concentration, complicate interpretation of the data. There is thus a clear need for more detailed experimentation that removes the influence of the boundaries.

Comparison of our results with experiments is further complicated by the typically more complex electrostatic boundary conditions that gold- or carbon-coated hot swimmers can possess. These hot Janus swimmers may be partially conducting and partially insulating. It is conceivable that this leads to a “best of both worlds” scenario, where a relatively high swim speed is maintained over all values of $n^\infty$. Interestingly, Ly et al. [38] pointed out that an equipotential boundary condition may not be applicable at all, as such a coating may be granular, also see the experimental work in Ref. [54]. How such non-uniformity impacts the above results will be left to future investigations. However, these changes in boundary condition can be readily explored within the framework which we have detailed here.

It is our hope that the results presented in this paper will aid in the design of the next generation of biocompatible swimmers and inspire the development of new experiments on collective behavior, wherein superior control is achieved over the underlying self-propulsion mechanism.

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Appendix A: Temperature Profiles

To obtain the temperature profiles in our system, we follow Ref. [44] and reproduce their results here in our notation for completeness. We examine two boundary conditions for the coated hemisphere. For a cap maintained at constant temperature, we assume for the thermal conductivities $k_l = k_a$, and obtain for the temperature field outside of the swimmer

\[
T(r) = T_i + \frac{a q}{2 k_l} \left[ \frac{a}{r} \right] + \sum_{i=0}^{\infty} \tilde{t}_i \left( \frac{a}{r} \right)^{i+1} P_i (\cos \theta),
\]

\[
\tilde{t}_{i=2k} = 0,
\]

\[
\tilde{t}_{i=2k+1} = -\frac{1}{\pi} \left( -1 \right)^k \frac{k+1}{2k+1},
\]

where, $P_i$ is the $i$-th Legendre polynomial.

For a constant heat flux into the cap, the temperature field reads

\[
T(r) = T_i + \frac{a q}{2 k_l} \left[ \frac{a}{r} \right] + \sum_{i=0}^{\infty} \tilde{t}_i \left( \frac{a}{r} \right)^{i+1} P_i (\cos \theta),
\]

\[
\tilde{t}_{i=2k} = 0,
\]

\[
\tilde{t}_{i=2k+1} = -\frac{4k+3}{(2k+2) + (2k+1)K} \left( -1 \right)^k (2k)! ,
\]

where $K = k_a/k_l$ is the conductivity contrast. In this case, the maximum temperature difference appearing in our $\tau^2$ expansion can be written as

\[
\Delta T = \frac{a q}{2 k_l} \left[ 1 - \sum_{i=0}^{\infty} \tilde{t}_i \right],
\]

leading to a reduced temperature field in a more convenient form for our purposes,

\[
t(r) = \left( \frac{a}{r} \right) \left[ 1 - \sum_{j=0}^{\infty} \tilde{t}_j \right]^{-1} \sum_{i=0}^{\infty} \left( \frac{a}{r} \right)^{i+1} P_i (\cos \theta),
\]

\[
\tilde{t}_i = \tilde{t}_i \left[ 1 - \sum_{j=0}^{\infty} \tilde{t}_j \right]^{-1}.
\]

Appendix B: Justification of Linearization Assumptions

In this appendix, we justify the reductions we made in the main text. We refer to the work by Dietzel and Hardt [37] and references therein for a full discussion of the first-order Taylor expansion coefficients to the physical quantities. Here, we reproduce the values listed in Ref. [37] in terms of our notation. For the medium they found $\eta^* \approx -5$, $k^* \approx 0.7$, and $\epsilon^* \approx -1.3$. For the “typical” ions $Na^+$, $K^+$, and $Cl^-$ they obtained $D_+^* \approx 6$ [37]. For the variation in the thermal diffusion coefficient of the ions only limited data is available in the literature. We refer to the work of Caldwell [55], from which we obtain $D_+^* \approx 1$ for NaCl and a temperature dependence for the thermal diffusivity given by $\alpha_+^* \approx 2$. There is clearly some variation in the literature values, but importantly all these numbers are order unity and we are therefore justified in ignoring these temperature dependencies. They come into the differential equations for the potential and concentration at $O(\tau^2)$, with $\tau^2 \approx 3 \cdot 10^{-4}$ for $\Delta T \leq 5 \text{K}$, leading to minute variations. For the speed they come into the expression at $O(\tau)$, which leads to a change of at most 10%. The only exception to this rule is $\epsilon^*$, which enters the theory at linear order and contributes as a constant to the speed, see Eq. (32). Finally, the Péclet number for the ions in our systems are
given by \( Pe = Ua/D \), with \( U \leq 10 \mu m/s^{-1} \) the typical velocity, \( a = 1 \mu m \) the radius of the colloid, and \( D \geq 1.0 \cdot 10^{-9} m^2/s \) the smallest ion diffusivity for convenience. Using the numbers provided in section IV, we find that \( Pe \leq 10^{-2} \), therefore we can safely ignore advection terms in Eq. (15).

**Appendix C: The Equilibrium Linearization**

The linearized equations for the equilibrium in terms of our reduced quantities are as follows. The heat equation reduces to a constant temperature \( T^\infty \) throughout the system. The Stokes equations reduce to zero fluid velocity, with the following pressure condition

\[
\nabla p^eq(r) = -k_B T^\infty n^\infty \left(x^eq_+(r) - x^eq_-(r)\right) \nabla \phi^eq(r).
\]

That is, the hydrostatic pressure exactly cancels the ionic pressure terms induced by electrostatic screening of any charge or potential on the colloid. The linearized equilibrium Poisson equation reads

\[
\nabla^2 \phi^eq(r) = -\frac{1}{2}(\kappa^\infty)^2 \left(x^eq_+(r) - x^eq_-(r)\right).
\]

Lastly, the ionic fluxes become

\[
\mathbf{J}^eq_\perp = -D^eq_\perp n^\infty \left[ \nabla x^eq_+(r) + \nabla \phi^eq(r) \right],
\]

with the closure \( \mathbf{J}^eq_\perp = \mathbf{0} \). The latter follows from the fact that in equilibrium the fluxes vanish. Using the closure, we find that \( x^eq_\pm(r) = \mp \phi^eq(r) \) and \( \nabla^2 \phi^eq(r) = (\kappa^\infty)^2 \phi^eq(r) \). The hydrostatic pressure condition reduces to \( \nabla p^eq(r) = 2k_B T^\infty n^\infty \phi^eq(r) \nabla \phi^eq(r) \).

**Appendix D: The Out-of-Equilibrium Force Density**

In this appendix, we provide the force density onto the fluid in the Stokes equations. For simplicity and analytic convenience, we will assume that the reduced surface potential \( \phi(r_s) \) is locally uniform, i.e., \( \nabla \phi(r_s) = \mathbf{0} \) except possibly at singular points. Writing for the force density \( f^eq \) we then arrive at the following expression

\[
\frac{f^{neq}}{k_B T^\infty n^\infty} = -2\delta X^{neq}(r) \nabla \phi^{eq}(r) + 2\phi^{eq}(r) \nabla \phi^{neq}(r) - (\kappa^\infty)^2 \epsilon^* |\nabla \phi^{eq}(r)|^2 \nabla t(r).
\]

Outside the screening layer, the force density vanishes

\[
\frac{f^{neq}}{k_B T^\infty n^\infty} = 0,
\]

to linear order in \( \tau \). Higher-order terms would lead to a force in the bulk, as there is an unscreened electric field and a temperature gradient. These couple to the temperature-variation of the dielectric permittivity to generate a force on the fluid, see Eq. (5), but this contribution is small, scaling with \( \tau^3 \). Inside of the screening layer, we can split the driving forces into components parallel and perpendicular to the surface. These read

\[
\frac{f_{in,||}}{k_B T^\infty n^\infty} = 2\phi(r_s)e^{-q} \nabla \phi^{neq}(r_s, q) - \epsilon^* \phi(r_s)^2 e^{-2q} \nabla t(r_s);
\]
\[
\frac{f_{in,\perp}}{k_B T^\infty n^\infty} = 2\kappa^\infty \phi(r_s)e^{-q} \delta X^{neq}(r_s, q) + \partial_q \phi^{neq}(r_s, q) \hat{q},
\]

where we have used that \( t(r) \) is almost constant in the perpendicular direction over the length of the double layer.

The expressions for the force density inside the screening layer can be rewritten using the expressions for the density and potential. For the perpendicular component we find

\[
\frac{f_{in,\perp}}{k_B T^\infty n^\infty} = -\kappa^\infty \left[ 4\beta - (3 + \epsilon^* - 2(1 + \epsilon^*) q) \phi(r_s) \right] \times \phi(r_s)e^{-2q} \nabla t(r_s) \hat{q},
\]

which holds for the equipotential \( \phi(r_s) = \phi_0 \) surface, and

\[
\frac{f_{in,\parallel}}{k_B T^\infty n^\infty} = \kappa^\infty \left[ 1 - \epsilon^* - 2(1 + \epsilon^*) q \right] \times \phi(r_s)^2 e^{-2q} \nabla t(r_s) \hat{q},
\]

which holds for insulating \( \phi(r_s) = \phi_s \) surface. Under the same assumption, we obtain

\[
\frac{f_{in,||}}{k_B T^\infty n^\infty} = -\left[ (2 - 2e^{-q}) \beta + (\epsilon^* - (1 + \epsilon^*) q) \phi(r_s)e^{-q} \right] \hat{q} \times \phi(r_s)e^{-q} \nabla t(r_s);
\]
\[
\frac{f_{in,\perp}}{k_B T^\infty n^\infty} = -[2\beta - (1 + (1 + \epsilon^*) q) \phi(r_s)e^{-q}] \times \phi(r_s)^2 e^{-2q} \nabla t(r_s),
\]

for the equipotential and insulating surface, respectively.

**Appendix E: The Expansion of Teubner’s Integration**

Here, we rewrite the expression for the speed given by Teubner [47], see Eq. (29), in terms of the body force inside and outside the screening layer — using that the latter is vanishing and that the system is axisymmetric — to arrive at

\[
\bar{U} = -\frac{1}{3\mu \alpha} \int_a^\infty \int_{\pi}^\pi \sin \theta K(r) \cdot f^{neq}(r) d\theta dr;
\]
\[
= -\frac{1}{3\mu \alpha} \int_a^\infty \int_{\pi}^\pi \sin \theta K(r) \cdot f_{in}(r) d\theta dr,
\]
where \( a^+ \) marks the edge of the screening layer. We have that \( \mathbf{K}(\mathbf{r}_s) = 0 \) and we must therefore perform a perturbative analysis in terms of \( \lambda \) to leading order in \( \lambda \).

For the force, we may write

\[
\hat{F}(\mathbf{r}_s + \lambda^\infty \mathbf{q}) = A(q)\mathbf{t}(a, \theta) + B(q)a^{-1}\partial_\theta t(a, \theta). 
\]

(E4)

where the term \( t(a, \theta) \equiv t(\mathbf{r}_s) \) is the temperature at the surface. Here, \( A(q) \) accounts for all the prefactors in Eqs. (D5) and (D6); \( B(q) \) accounts for all the relevant prefactors in Eqs. (D7) and (D8); and \( a^{-1}\partial_\theta t(a, \theta) \equiv \nabla_t(\mathbf{r}_s) \). Note that here we have used our assumption that \( \phi(\mathbf{r}_s) \) is homogeneous over the surface to avoid \( q \) dependence in the factors \( A \) and \( B \).

Now taking everything together, we may rewrite the expression for the speed contribution due to the region inside of the thin screening layer as

\[
\tilde{U}_\perp = \frac{(\lambda^\infty)^2}{2\eta a} \int_0^\infty \int_0^{\pi} \lambda^\infty q^2 \cos \theta \sin \theta A(q)t(a, \theta) d\theta dq - q^2\theta B(q)\partial_\theta t(a, \theta) d\theta dq. 
\]

(E5)

Spitting the integrand into the \( A(q) \) (\( \perp \)) and \( B(q) \) (\( \parallel \)) terms, we evaluate these contributions separately. Starting with the perpendicular component, we find

\[
\tilde{U}_\perp = \frac{(\lambda^\infty)^3}{2\eta a} \int_0^\infty \int_0^{\pi} q^2 \cos \theta \sin \theta A(q)t(a, \theta) d\theta dq - \frac{(\lambda^\infty)^3}{2\eta a} \int_0^\infty q^2 A(q) dq \int_0^{\pi} t(a, \theta) \cos \theta \sin \theta d\theta 
\]

(E6)

where only the first-order Legendre Polynomial contributes. Note that if we had not assumed homogeneous electrostatic surface properties, the splitting of the integration could not have been done in the same way and all Legendre-Fourier modes would have contributed. Evaluating the integral over \( q \) gives us for a conducting surface

\[
\tilde{U}_\perp = -\frac{k_BT^\infty n^\infty}{6\eta a} (\lambda^\infty)^2 \left[ 2\beta \phi_0 + \epsilon^* \phi_0^2 \right] \tilde{t}_1. 
\]

(E7)

The result for an insulating surface is

\[
\tilde{U}_\perp = -\frac{k_BT^\infty n^\infty}{6\eta a} (\lambda^\infty)^2 \left[ 1 + 2\epsilon^* \right] \phi_s^2 \tilde{t}_1. 
\]

(E8)

where \( \phi_s \) is given by Eq. (28).

Similarly, we obtain for the parallel component

\[
\tilde{U}_\parallel = \frac{(\lambda^\infty)^2}{2\eta a} \int_0^\infty \int_0^{\pi} q^2 \sin^2 \theta B(q)\partial_\theta t(a, \theta) d\theta dq - \frac{(\lambda^\infty)^2}{2\eta a} \int_0^\infty qB(q) dq \int_0^{\pi} \sin^2 \theta \partial_\theta t(a, \theta) d\theta 
\]

(E9)

Evaluating the integral over \( q \) leads to the desired expression for a conducting surface

\[
\tilde{U}_\parallel = -\frac{k_BT^\infty n^\infty}{6\eta a} (\lambda^\infty)^2 \left[ 6\beta \phi_0 - \phi_0^2 \right] \tilde{t}_1. 
\]

(E10)

and for an insulating surface

\[
\tilde{U}_\parallel = -\frac{k_BT^\infty n^\infty}{6\eta a} (\lambda^\infty)^2 \left[ 8\beta \phi_s - (2 + \epsilon^*) \phi_s^2 \right] \tilde{t}_1. 
\]

(E11)