Hot Microswimmers

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Abstract. Hot microswimmers are self-propelled Brownian particles that exploit local heating for their directed self-thermophoretic motion. We provide a pedagogical overview of the key physical mechanisms underlying this promising new technology. It covers the hydrodynamics of swimming, thermophoresis and -osmosis, hot Brownian motion, force-free steering, and dedicated experimental and simulation tools to analyze hot Brownian swimmers.

1 Introduction

All physicists know Newton’s first law of motion, namely that (to inertial observers) all things move at a constant velocity $v$, unless acted upon by an external force $F$. It certainly holds also for particles immersed in a viscous fluid of shear viscosity $\eta$, at a fundamental level. Yet, if you do not see the solvent, e.g. because it is transparent, the world of these particles looks very much “Aristotelian”, in the sense that things usually do not move without forcing. The reason is that momentum is constantly dissipated to the solvent (assumed to be at rest). The law of motion of the particles therefore depends both on the particle and on the solvent properties, the best-known example being Stokes’ law, $F = 6\pi \eta a v$, for the forced stationary motion of a spherical bead of radius $a$. Now, turn a small colloidal particle into a microswimmer, e.g. by coating it asymmetrically with a gold cap and heating it by laser light \cite{1}. Then it does move without external forcing. Swimming is a form of autonomous motion. Axially symmetric swimmers, the simplest examples for active colloids, do seem to obey Newton’s first law. They are in uniform force-free motion, despite being immersed in a dissipative solvent. However, in contrast to Newton’s particles in vacuum, their uniform drift arises from a complex dissipative solvent flow around the particle and relies on a constant supply of energy, e.g. in the form of chemical fuel, acoustic actuation, or laser heating, that maintains the system far from equilibrium \cite{2}. Moreover, the solvent motion never is entirely deterministic but introduces Brownian fluctuations in the position and orientation of the particle that will sooner or later randomize the

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particle path. In the following, we gather some facts and ideas about how such Brownian swimming motion actually comes about and how it is quantitatively described. Within the broad class of artificial micro- and nano-swimmers, which can nowadays be fabricated in large numbers and with a great variety of propulsion mechanisms [3], we focus on so-called hot Brownian swimmers — thermally anisotropic Brownian particles fueled by (optical) heating. Briefly, the propulsion of heated particles works as follows. First, a geometric/material asymmetry of the particle is exploited to establish an asymmetric temperature profile in the surrounding solvent upon particle heating. There ensues a thermoosmotic flow along the surface of the particles. It can either be harnessed for pumping, if the particle is held fixed in space by some external force, or for phoretic self-propulsion, if the particle is mobile. Boiling is strongly suppressed by the Laplace pressure (which is inversely proportional to the radius of curvature and can therefore be quite substantial for small particles) and heating is highly localized for nanoparticles. Therefore, substantially larger temperature gradients and, accordingly, more efficient thermophoretic transport can be realized with self-thermophoresis than with conventional macroscopic thermophoresis. Self-thermophoresis is thus arguably an interesting and technologically promising propulsion mechanism, with some important advantages over other designs. i) Universality, Availability, Biocompatibility: it does not rely on exotic (maybe poisonous) solvents or fuels, but exploits a comparatively “universal” mechanism. It does not run out of fuel and is minimally invasive, since the heating is local and sizable motion can already be achieved with minor heating of the surroundings. ii) Control: the propulsion speed can be regulated continuously and propulsion can instantly be switched on and off, e.g. by using conventional lasers and microscopy equipment. Thanks to emerging efficient cooling mechanisms for colloidal particles [4], one can even imagine fabricating particles with a reverse gear. Besides, efficient force-free steering mechanisms such as photon nudging [5] are already available. iii) Versatility: heating can be realized by a variety of methods, such as the absorption of laser light by metal or carbon parts, or of microwaves by super-paramagnetic parts, which opens up the possibility of combining several independent and independently addressable thermophoretic propulsion mechanisms into one microstructure. A whole community of researchers is moreover interested in micro- and nano-particle heating for its own sake [6]. iv) Scalability: downscaling does not reduce the propulsion speed [5] but increases the efficiency [7]. Synthesis, speed control, and steering [5] of self-thermophoretic swimmers, as well as their photothermal detection [8], are all scalable to nanoscopic dimensions.

The remainder of the paper is structured as follows. The next section reviews the distinction between self-phoretic, phoretic, and passive motion and the concomitant flow fields excited in the solvent, on a hydrodynamic level. This means that the physical mechanism that actually drives the motion is confined to such a narrow boundary layer that it is sufficiently well captured by a mere hydrodynamic boundary condition for the solvent flow. On this level, the theory of swimming is universal and independent of the actual propulsion mechanism. The following section provides a closer look at the engine of the swimmer, namely the processes of phoresis and self-phoresis of heated particles. The basic principles of self-thermophoretic propulsion and the underlying osmotic processes are very similar to those in other phoretic phenomena, such as diffusiophoresis and electrophoresis. In fact, in real-world realizations, one often encounters a complex mix of several such mechanisms, which can be hard to disentangle for small particles in water, which is arguably the most interesting system for many applications. The particles are usually charged and surrounded by counterions and dissolved salts, all effects being sensitive to temperature. And, more often than not, everything is observed close to a surface, to which similar considerations apply. Next, we also consider the fluctuating part of the motion of a hot Brownian swimmer, namely its hot Brownian motion. This complex nonequilibrium motion lim-
its the deterministic swimming motion by randomizing the swimming direction and therefore needs to be understood. With regard to the mentioned experimental and conceptual difficulties encountered when working with hot Brownian simmers, the ability to numerically simulate models of reduced complexity is particularly valuable. We therefore provide a brief overview over a nonequilibrium molecular dynamics approach that is suitable to simulate hot nano-swimmers on a coarse-grained, yet atomistic basis. Finally, we discuss some dedicated experimental techniques for the detection (photothermal imaging and correlation spectroscopy) and force-free steering (photon-nudging) of hot swimmers.

2 Hydrodynamics: Dragging, Swimming, Phoresis

The contents of this section have in principle been known for more than a century, explicitly at least for some decades, and something similar holds for Sec. 3 (see e.g. [9, 10]). So almost everything has been said, though not yet by everybody. This lack of eloquence has been cured by the more recent literature, which moreover has stirred up some confusion by muddling with the distinction between dragging, swimming, and phoresis (i.e., some writers do not bother to distinguish sail boats from motor boats). This motivated us to include this pedagogic material here, although there are already a number of recent technical reviews clarifying these classical topics for interested contemporary readers (e.g. [11–13]).

For any small suspended particles moving at slow speeds — technically speaking, at low Reynolds numbers $|v|a/\nu \ll 1$ — the spatio-temporal solvent velocity field $u(r, t)$ reacts basically instantaneously in the region where it takes any sizable values, and therefore follows [14] from the stationary Stokes equations,

$$\nu \nabla^2 u = \nabla P - f, \quad \nabla \cdot u = 0,$$

Here, $\nu \equiv \eta/\rho$ denotes the kinematic viscosity, $\rho$ is the mass density of the solvent, and $f$ an optional external force density. The hydrodynamic “pressure” $P$ (actually $P$ is the pressure divided $\rho$) can be thought of as a Lagrange multiplier that serves to account for the second equation. The latter asserts that (without imposed hydrostatic pressure gradients) stationary flows have constant density and therefore must be divergence-free to respect mass conservation: $0 \equiv \partial_t \rho = -\rho \nabla \cdot u$. Equation (1) is the universal basis for a hydrodynamic description of passive and active colloidal particles. On a coarse-grained phenomenological level (i.e. not looking too closely into what is actually really happening very near to the particle surface), passive particles and swimmers merely differ by the hydrodynamic boundary conditions imposed on Eq. (1) at the particle surface. The generic boundary condition on solid surfaces is $u|_S = v$, i.e., the velocity of the fluid and particle match at the particle surface (“no slip, no influx”). Yet, allowing the solvent to slip at the particle surface is the easiest way for a theoretician to turn a passive particle into a swimmer — and usually a very good coarse-grained model for artificial phoretic swimmers and all kinds of animalcules propelled by their multi-ciliated or dynamically wrinkling skins. Boundary conditions and flow fields corresponding to passive drag and active swimming can simply be superimposed by virtue of the linearity of Eq. (1).

Two further remarks seem in order, here. First, note that the solutions of Stokes’ equations depend on the density and viscosity of the fluid only in the combination of the kinematic viscosity $\nu = \eta/\rho$. This has an interesting consequence. Namely, while air and water feel very differently to human swimmers because of their large density ratio ($\simeq 10^3$) and the high Reynolds numbers involved ($\simeq 10^6$), their kinematic viscosities are on the same order; hence, microswimmers actually would not mind
the difference. Further, note that $\nu$ has the physical units of a diffusion coefficient and does indeed describe the diffusion of vorticity through the fluid, which is how momentum gets dispersed. To see this, and that the stationarity assumption amounts to the idealization of an infinitely fast diffusion of the vorticity $\nabla \times \mathbf{u}$, simply take the curl of Eq. (1).

**Passive Transport or Drag.** Consider a colloidal sphere of radius $a$ moving in a liquid solvent of shear viscosity $\eta$. The law relating the particle’s velocity $\mathbf{v}$ to the applied external force $\mathbf{F}$ (the analog of Newton’s second law for a dissolved particle) is Stokes’ law,

$$\mathbf{v} = \frac{\mathbf{F}}{6\pi \eta a}.$$  

Stokes derived it in the mid-19th century from Eq. (1), which yields the flow field

$$\mathbf{u}(\mathbf{r}) = \frac{3a}{4r}(1 + \hat{\mathbf{r}}) \cdot \mathbf{v} + \frac{a^3}{4\eta r^3}(1 - 3\hat{\mathbf{r}}) \cdot \mathbf{v}$$  

around the sphere, where $\hat{\mathbf{r}} \equiv \mathbf{r}/r$. This important result is illustrated in Fig. 1.

In the far field (or for an infinitely small sphere), the flow velocity in Eq. (3) is dominated by the first term, with the reciprocal dependence on the distance $r \equiv |\mathbf{r}|$. Using Eq. (2), it is immediately recognized as the so-called force monopole or “stokeslet”,

$$\mathbf{u}(\mathbf{r}) = \frac{1}{8\pi \eta r^6} (1 + \hat{\mathbf{r}}) \cdot \mathbf{F}, \quad r \gg a,$$  

which is the fundamental solution of Eq. (1), i.e., the fluid response to a point force $\mathbf{f} = \mathbf{F}(\mathbf{r})$ applied at the origin (Fig. 1a). It is solely responsible for dispersing all the momentum and vorticity supplied to the fluid by the drag force $\mathbf{F}$, as required by Newton’s third law, and apparent from its reciprocal dependence on the distance. (A fluid velocity $\mathbf{u} \propto r^{-1}$ amounts to a total momentum leaking out to infinity at a constant rate — i.e., perfect force balance with the surroundings.) Accordingly, Eq. (4) alone reproduces Stokes’ law, Eq. (2), upon averaging over the surface of the sphere at $r = a$. Indeed, the projector in parentheses in Eq. (4) renders $\mathbf{u}$ divergence-free and averages to $(1 + \hat{\mathbf{r}})^2 = \text{tr}(1 + \hat{\mathbf{r}})/3 = 1(3 + 1)/3 = (4/3)I$, as required. In
summary, stokeslet and external force are inseparable twins that always come and go together, you cannot have one without the other.

The stokeslet does not exactly conform with the no-slip boundary condition, though. In fact, it does not even bear any signature of the particle size, at all. This is where the second term in Eq. (3) comes in, which is known as a source doublet. Its sole purpose is to let the velocity slip along the particle surface, so that it could also very well be called a “sliplet”. By construction, it precisely corrects the “failure” of the stokeslet, Eq. (4), to account for the finite size of the particle. This statement is easily verified by setting \( r = a \) in Eq. (3) and gleaned from Fig. 1, which depicts the streamlines around a dragged particle according to Eq. (3) (c), and the sliplet alone (d), in the particle frame. The figure moreover shows that the dragged particle perturbs the surrounding solvent much more than a particle motion that involves only a sliplet. Note, as an aside, that Figs. 1, 2 also indicate that the visual appearance of the flow field changes drastically with the reference frame, while subtleties related to the character of the flow field may be hard to discern, which complicates experimental studies of the hydrodynamics of low-Reynolds-number swimming [15].

**Phoresis.** According to the above discussion, the second term in Eq. (3), the source doublet or sliplet, does not change the average fluid velocity on the particle surface, nor does it carry vorticity that would give rise to a net force or leak momentum to infinity. It idealizes the complex dynamic processes, which actually take place in a narrow boundary layer around the particle, as a kind of tank-treading motion. With its help, the sphere effectively sneaks through the fluid (Fig. 1d). The normal velocity component of the sliplet on the particle surface is \( \hat{r} \cdot u(a) = (1 - 3) \hat{r} \cdot v/4 = -(v/2) \cos \theta \) in the lab frame. (For the moving frame, subtract \( v \) with normal and tangential components \( v \cos \theta \) and \( v \sin \theta \), respectively.) This means that the sliplet alone, without the stokeslet, fulfills the no-influx boundary condition for a sphere moving with velocity \(-v/2\). Therefore, taking twice the negative sliplet in Eq. (3), namely

\[
\mathbf{u} = \frac{a^3}{2r^3} (3\hat{r} \hat{r} - 1) \cdot \mathbf{v}
\]

allows a spherical particle to move at velocity \( \mathbf{v} \) without violating the no-influx boundary condition. In other words, we have just constructed a perfect swimmer. For such a perfect swimmer (or should we say “slipper”?), the tangential component of the fluid velocity at the surface contributes \( \hat{v} \cdot (1 - \hat{r} \hat{r}) \cdot u(a) = -(v/2) \sin^2 \theta \) along the propulsion direction, i.e., it must be \( -(v/2) \sin \theta \). The relative tangential velocity of the fluid in the particle frame therefore does not vanish but has a slip of magnitude \(-3(v/2) \sin \theta \), corresponding to a maximum tangential slip velocity of magnitude \(-3v/2\) at the equator. If its projection \(-3v/2 \sin^2 \theta \) onto the propulsion direction is averaged over the sphere, one exactly recovers the negative propulsion velocity.

Now, observe that a sinusoidal variation of the tangential slip velocity along the circumference of the particle is precisely what one would expect if the slip mechanism was caused by the tangential component of some linear external gradient along the propulsion direction. The sliplet is thus not only a theoretician’s dream of a swimmer; it is at the same time the simplest coarse-grained hydrodynamic description of a homogeneous sphere moving by any kind of phoretic mechanism, whenever the microscopic processes responsible for the phoretic slip are confined to a narrow boundary layer at the sphere’s surface and proportional to an undisturbed linear external gradient. As an obvious consequence, the swimming velocity of a force-free phoretic particle must be independent of its size if all other parameters are held constant. (The actual size dependence of identically designed hot Janus swimmers exposed to the same laser power is a more subtle issue, since it depends on the size dependence of the scattering cross section [5]).
Fig. 2. Lab-frame streamlines for an imperfect squirmer, consisting of a “sliplet” \( u \propto r^{-3} \) (the perfect engine) and a stokeslet dipole \( u \propto r^{-2} \) (the dominant far-field signature of imperfections) in force-free swimming mode (a) and in pumping mode (b), which creates a stokeslet that counterbalances the external stalling force keeping the pump in place. The forth-back/left-right symmetric dipole field alone (c) corresponds to the far field of a puller/pusher moving vertically/horizontally, respectively, whereas the analytically calculated streamlines around a more realistically modeled force-free Janus particle with a heated gold cap (d, adapted from [16]) exhibits pronounced near-field structure.

**Self-Phoresis** Self-phoresis is just like phoresis, the only difference being that the gradient of the relevant field (the electrical potential in electrophoresis, the concentration of a solute in diffusiophoresis, the temperature in thermophoresis, etc.) is created by the particle itself rather than being externally imposed. Depending on the design of the self-phoretic particle, the self-generated gradient field varies more or less nonlinearly along the particle axis, so that hydrodynamic perturbations to the pure sliplet are usually induced. The leading term, dominating the far field, is a force (or stokeslet) dipole. It can be imagined as consisting of two stokeslets \( u \propto r^{-1} \) of equal strength that are slightly symmetrically displaced from the center along the particle axis and pointing in opposite directions along this axis. The force dipole thus excites a forth-back and left-right symmetric flow field that decays like \( u \propto 1/r^2 \) (Fig. 2). It does not contribute to the propulsion, but, as the slowest radially decaying component in the flow field, it dominates the interactions of most imperfect swimmers with walls and other particles or swimmers. Neglecting all higher-order (faster decaying) corrections amounts to the simplest “squirmer” approximation, in which self-phoretic swimmers can be classified according to the strength and orientation (in or out) of their stokeslet-dipole term. They are called “pushers” or “pullers” if the flow created by the stokeslet dipole is oriented outwards or inwards along the particle axis, respectively, and “neutral squirmers”, if the stokeslet dipole is absent. Pullers are naturally attracted to each other and to walls head-on, while pushers (which actually pull sideways) tend to align with walls and other particles or swimmers [17]. In any case, because of the linearity of the flow, the hydrodynamic effect of walls is very much like the effect of other swimmers. In simple geometries it can be simulated by image swimmers, as familiar from the image-charge method in electrostatics. The pertinence of the simple squirmer description with only a sliplet and a dipole term (Fig. 2a) can be gauged by comparison with the plot in Fig. 2d, which depicts the exact solution for the thermoosmotic flow around a Janus swimmer with a hot isothermal gold cap, assuming (identical) finite thermal conductivities of bulk and solvent and, of course, an infinitely thin thermoosmotically active boundary layer [16]. While some overall resemblance can be detected if the different propulsion directions (diagonal/upwards) in the two plots are taken into account, the more realistic flow field is obviously strongly affected by additional near-field contributions. Recent attempts to classify the collective behavior in dense suspensions by simplified hydrodynamic models might therefore have to be taken with a grain of salt.
Fig. 3. An externally applied temperature gradient along a liquid-solid interface excites a thermoosmotic creep flow. Hydrodynamically, i.e., if seen from a distance much larger than the interaction length $\lambda$ determined by the physics and chemistry at the solid-liquid interface, it can be described by a local surface slip velocity $u_s(r)$. Lateral variations can arise from lateral variations in the solid-liquid interactions or in the temperature gradient. At a fixed solid boundary, the solvent slip acts as an osmotic pump, while it turns a dissolved mobile particle into a phoretic swimmer. Its thermophoretic drift velocity $v_{tp}$ is given by the surface-average of the negative slip velocity $-u_s(r)$.

3 Propulsion: Thermoosmosis and Thermophoresis

As stated in the previous section, a particle with a tangential slip component of the fluid velocity at its surface would allow to construct an ideal swimmer. To achieve this tangential surface slip one may tune the interfacial interactions between the liquid and the particle. In the colloidal domain, the interaction range at the liquid-solid interface usually decays within a certain interaction distance $\lambda$ from the interface (see Fig. 3). Within this boundary layer, the interfacial excess enthalpy $h(r_\perp)$ decays to zero, where $r_\perp$ is the distance from the interface [18]. Depending on the surface chemistry and on the complexity of the solvent, the interfacial excess enthalpy may comprise a number of different contributions. Simple microscopic model descriptions are available for some special cases such as interfacial electrostatic interactions arising from a charged solid surface and a counter ion cloud bound in a double layer [10].

If a thermodynamic field gradient, e.g. of an electric field [10], or of a solute concentration [19], or temperature [18], with a tangential component is externally applied, this modifies the thermodynamic balance in the interfacial layer and causes a tangential interfacial creep flow. In particular, for a temperature gradient $\nabla || T$ along a container wall, this creep flow is a so-called thermoosmotic flow. The wall acts as an osmotic pump. The flow velocity vanishes at the solid boundary and saturates at a distance around the interaction length $\lambda$ (see Fig. 3). Hydrodynamically, i.e., seen from a distance much larger than $\lambda$, the effect can be described by a local slip velocity $u_s(r_\parallel)$ of the solvent at the surface. The thermophoretic mobility $\mu(r_\parallel)$ has been expressed as an integral over the sheared boundary layer [13, 18]

$$\mu(r_\parallel) = -\frac{1}{\eta T} \int_0^\infty dr_\perp r_\perp h(r_\perp, r_\parallel) .$$

(6)

It depends on the interfacial properties and will generally change laterally if the liquid-solid interactions vary along the liquid solid interface, as typically the case for artificial microswimmers. For example, in the case of a Janus particle, the value of $\mu$ will generally differ between the two hemispheres. A thermoosmotic slip flow along the surface of a mobile particle will set the particle into thermophoretic motion [20, 21]. As discussed in the context of Eq. (5), the acquired thermophoretic drift velocity $v_{tp}$ follows by averaging the negative surface creep flow $-u_s$ over the particles surface $S$,
namely,
\[ v_{tp} = -\frac{1}{S} \int u_\parallel(r_\parallel) \, dS = -\frac{1}{S} \int \mu(r_\parallel) \nabla_\parallel T \, dS \]  
\[ (7) \]

If the temperature gradient is generated by the phoretic particle itself, the ensuing motion can justly be called “self-thermophoretic”. However, whenever the particle swims close to a container wall or other boundaries, it will generally induce some thermoosmosis there and, in return, pick up additional flow contributions, so that the overall particle velocity \( v \) will differ from the nominal thermophoretic drift velocity \( v_{tp} \). The same holds for mutual encounters of swimmers. The effect can loosely be thought of as a “catalysis” mechanism for the swimmer’s propulsion engine.

4 Fluctuations: Hot Brownian Motion

The study of microswimmers has a long history dating back to the 17th century, when they were first observed under the microscope, most notably by the Dutch draper Antoni van Leeuwenhoek. Only much later, starting with systematic investigations by scientists like Robert Brown and Adolphe Brogniard in the early 19th century, researchers slowly became aware of the interference of Brownian motion with microscale swimming, and much of the pioneering work was devoted to disentangling both effects. So the study of animalcules predated that of molecules, and what started as an investigation of the former eventually furnished proof of the existence of the latter [22]. Today we are retracing this path backwards, from bottom-up. Brownian motion, which is due to thermal fluctuations of the molecules of the swimmers’ medium and cannot be switched off, is well understood for isothermal solvents. One might think that it gets easily outpaced by the directed ballistic motion of swimmers, but it reappears through the back door of rotational Brownian motion that randomizes the swimming direction. Moreover, since we are particularly interested in non-isothermal swimmers, driven by thermophoresis and thermoosmosis, we have to consider non-isothermal or “hot” Brownian motion [23], which is an interesting subject by itself, and has to be understood if one wants perfect control over hot swimmers.

The heat emanating from a hot micro- or nano-swimmer has two main effects. It reduces the friction and increases the thermal fluctuations around the swimmer and thereby enhances the translational and rotational Brownian motion of the swimmer. It turns out that a major simplification occurs for a coarse-grained description that holds on long times, where the stationary Stokes approximation in Eq. (1) holds for the deterministic solvent flow. This is called the Markovian limit, since it neglects memory effects due to the slow dynamics of vorticity diffusion, which are already present in an accurate description of equilibrium Brownian motion [24, 25], and which considerably complicate the theory of hot Brownian motion [26]. In the Markovian description of hot Brownian motion, the non-equilibrium effects can be subsumed into a small number of effective transport coefficients that can analytically and explicitly be calculated for sufficiently symmetric swimmer designs: chiefly, an effective reduced friction coefficient \( \zeta_{HBM} \) and an effective Brownian temperature \( T_{HBM} \). The two quantities determine the effective diffusivity \( D_{HBM} \) via a generalized Sutherland–Einstein relation [27],

\[ D_{HBM} = \frac{k_B T_{HBM}}{\zeta_{HBM}}. \]  
\[ (8) \]

To estimate the effective friction coefficient \( \zeta_{HBM} \), the equation of state \( \eta(T) \) of the solvent needs to be known. It can often (e.g. for water) accurately be represented by a Vogel Fulcher law, \( \eta(T) = \eta_\infty \exp[A/(T - T_{VF})] \), from which explicit predictions
for the effective translational and rotational friction coefficients of a hot sphere can be calculated (see e.g. Refs. [28, 29] and the supporting online materials in Ref. [27]).

Practically and conceptually it is more interesting to understand the effective temperature that characterizes the thermal agitation of non-isothermal Brownian particles. For pedagogic reasons, it is best to first consider a homogeneous spherical particle that is constantly maintained at a temperature above the ambient temperature, e.g., because it diffuses in a laser focus and absorbs the laser light much more efficiently than its surrounding solvent. The heating creates in the solvent a radial temperature field $T(r)$, co-moving with the particle (since heat diffuses via molecular collisions and therefore much faster than a colloidal particle). It can then be shown that the Brownian motion of the hot particle is described by the usual overdamped Langevin equations of motion with the following effective temperature for the noise strength [26, 27]

$$T_{\text{HBM}} = \frac{\int \phi(r)T(r)\,dr}{\int \phi(r)\,dr}. \quad (9)$$

This noise temperature is determined from the condition that it characterizes the Brownian motion of the heated diffusing particle as if it were an equivalent isothermal particle in a fluid of constant temperature $T_{\text{HBM}}$. Here $\phi(r)$ is the so-called dissipation function which depends on the viscosity and the solvent velocity gradient and weights the importance of fluctuations at the diverse local temperatures $T(r)$ according to their relevance for the agitation of the Brownian particle. Due to the different flow fields for translational (t) and rotational (r) motion of the particle, this prescription leads to different effective temperatures for the translational and rotational Brownian motion, namely [26–28]

$$T_{\text{HBM}}^t \approx T_0 \left(1 + \frac{5}{12} \Delta T\right), \quad T_{\text{HBM}}^r \approx T_0 \left(1 + \frac{3}{4} \Delta T\right), \quad (10)$$

where $\Delta T$ is the difference between the solvent temperature at the particle surface and the ambient temperature $T_0$. Higher order terms in $\Delta T$, which involve the effective viscosity $\eta_{\text{HBM}}(\Delta T)$, can be calculated but are usually small in actual applications. Note that, in contrast to certain effective temperatures that were recently hotly debated in other areas of non-equilibrium statistical mechanics, the effective temperatures of hot Brownian motion are not merely postulated but can systematically be calculated from an underlying non-isothermal fluctuating hydrodynamic theory [30], namely Eq. (1) with $f(r)$ representing a non-isothermal noise force. The latter can be expressed as the divergence of a fluctuating Gaussian shear stress field with a covariance proportional to $T(r)$.

While the effective translational temperature is usually the only quantity that matters for a homogeneous hot sphere and is most easily experimentally inferred from the translational Brownian fluctuations, its effect is easily outpaced by active propulsion at late times (see Ref. [31] for a detailed analysis of the velocity fluctuations of a hot Brownian swimmer). Then, it is actually the effective rotational temperature $T_{\text{HBM}}^r$ that matters more, because it limits the persistence of the directed propulsion. The observation of trajectories of a hot swimmer could thus be used to experimentally infer its rotational hot Brownian temperature $T_{\text{HBM}}^r$. For a very precise comparison with theory, one would then have to account for deviations from Eq. (10) due to the heterogeneous temperature field around the hot swimmer, depicted in Fig. 2d. To a good approximation, it is sufficient to evaluate Eq. (10) at the average temperature of the Janus particle, though.

So far, several of the predictions of the theory of hot Brownian motion could be validated experimentally and in numerical simulations [8, 23, 27, 31]. We specifically mention the experimental verification of the translational effective temperature
Fig. 4. Parameter-free experimental and numerical tests of the predicted hot Brownian diffusivities of a (homogeneous) hot Brownian particle, Eq. (10). Left: In the experiments, the Twin-PhoCS method (Sec. 6) is employed to measure the average time $\tau_D$ for crossing the laser focus [8]. Right: In the non-equilibrium molecular dynamics simulations (Sec. 5), the effective temperature $T_{HBM}$ is deduced via Eq. (8). Lines indicate the solvent temperature at the particle surface (dotted) and the theoretical predictions for the rotational (solid) and translational (dot-dashed) effective Brownian temperatures. Theoretical predictions [26] for the effective kinetic temperatures $T_{t,r}^k$ (open symbols) for the translational (t) and rotational (r) velocities are not yet available, since the theory has not yet been generalized to compressible solvents.

from Eq. (10). Interestingly, the experiment can exploit the solvent heating due to the hot particle for a highly accurate detection of its Brownian motion, as described in Section 6. Figure 4 (left panel) provides a parameter-free comparison of the average diffusion time $\tau_D$ of a heated Brownian particle in a laser focus, which has been obtained by this method, with the prediction in Eq. (10). An even more direct comparison of the various temperatures, i.e., the conventional local molecular solvent temperature and the effective temperatures characterizing the Brownian dynamics of various degrees of freedom (rotational, translational positions and velocities) of the particle is possible in our atomistic simulations, see Fig. 4 (right panel).

As a useful and instructive application of these concepts to the motion a hot Brownian swimmer, we also want to mention the validation [31] of a recently discovered spatial fluctuation relation [32]. In this work, the fluctuating velocity of a hot Brownian swimmer was recorded both in experiment and in numerical simulations. The corresponding histograms were shown to be in good accord with the predicted fluctuation theorem for the probabilities $P(J)$ to observe particle currents $J, J'$ of equal strength $J$ but in different directions, namely,

$$P(J) = P(J') e^{F(J - J')}.$$  \hspace{1cm} (11)

Here, the strength $|F| = \nu \zeta_{\text{eff}}/(2T_{\text{eff}})$ of the dissipative driving is proportional to the propulsion speed $\nu$ of the particle and $\zeta_{\text{eff}}$ and $T_{\text{eff}}$ denote the appropriate effective hot Brownian friction and temperature parameters for the Janus particle. The exponent in Eq. (11) thus has the interpretation of a non-equilibrium (hot Brownian) entropy production due to dissipation to a virtual bath at the effective temperature $T_{\text{eff}}$. The exact symmetry in Eq. (11) is found to hold far from thermal equilibrium, even though the solvent is not in thermal equilibrium and the driving is not due to a deterministic external force, as usually assumed in the derivation of Eq. (11). Instead, the hot Brownian swimmer is surrounded by a temperature gradient and the driving is due to its thermophoretic (force-free) self-propulsion.
5 Molecular Dynamics Simulations

Numerical simulations of hot microswimmers can be performed on various levels of coarse graining. One has to decide whether one can content oneself with a numerical modeling of the swimming engine on the phenomenological level, where it is subsumed into an effective slip boundary condition as in the above theoretical discussion, or whether higher resolution is required. In the former case, one can use efficient strategies to solve the hydrodynamic flow patterns around swimmers and between swimmers and other immersed bodies [33–36]. In the second case, classical atomistic molecular dynamics simulations (e.g. with Lennard–Jones particles) are more suitable. Here, we pursue the second route, since we want to be able to resolve some microscopic details, such as the interfacial thermal resistance (“Kapitza resistance”) and the mechanism of phoresis on an atomic scale, which cannot be captured by effective thermodynamic or hydrodynamic theories.

We simulate a spherical nanoparticle made of Lennard–Jones atoms that are tightly bound together by a FENE potential \( U(r) = -0.5\kappa_{\alpha\beta}R_0^2 \ln(1 - (r/R_0)^2) \), with \( R_0 = 1.5\sigma \), which is immersed in a Lennard–Jones solvent. A simple strategy to atomistically realize the double-faced structure of the experimentally employed Janus particles is to give the atoms on the two hemispheres different thermal resistances to the solvent [37]. (This is easier to achieve than maintaining a strong temperature gradient inside the particle by asymmetric heating or making the thermophoretic mobility of its hemispheres strongly heterogeneous. It is only the temperature gradient in the solvent that matters, after all.) The Kapitza resistances, in turn, are very sensitive to the wetting properties of the particle surface that can, in a simple way, be encoded in the atomic particle-solvent interaction potentials, given by the modified Lennard–Jones \( 12 - 6 \) potential,

\[
U_{\alpha\beta}(r) = 4\epsilon \left[ (\sigma/r)^{12} - c_{\alpha\beta} (\sigma/r)^6 \right],
\]

with an interaction cutoff at \( r = 2.5\sigma \). Here \( c_{\alpha\beta} \) play the role of wetting parameters for the various atom types \( \alpha \) and \( \beta \) [38, 39]. The value \( c_{ss} = 1 \) corresponds to the standard Lennard–Jones interaction, which we choose for the mutual interactions between the solvent particles. The atoms in a boundary layer of thickness \( \approx 1\sigma \) on one hemisphere of the nanoparticle represent the gold cap and are characterized by \( c_{gs} \) while the bulk atoms represent the polystyrene core of the particle and are characterized by \( c_{ps} \). For the FENE spring constants, we use \( \kappa_{gg} = 30\epsilon/\sigma^2 \) and \( \kappa_{sp} = \kappa_{gp} = 35\epsilon/\sigma^2 \). Choosing different wetting parameters has the effect of varying the minimum position \( (2\sigma^6/c_{\alpha\beta})^{1/6} \) of the pair potential. For \( c_{\alpha\beta} = 2 \), the equilibrium distance between the centers of a particle in the colloid and the solvent is \( \sigma \), for \( c_{\alpha\beta} \to 0 \) the attractive part and the local minimum of the potential are absent.

A typical simulation run consists of an equilibration phase in the NPT ensemble, with a Nosé–Hoover thermostat and barostat, at a temperature of \( T_0 = 0.75\epsilon/k_B \) and a thermodynamic pressure of \( p = 0.01\sigma^3/\epsilon \). In the ensuing heating phase, the global thermostat is then switched off and a non-equilibrium steady state is created: (a) by thermostating a vertical domain of solvent particles at the center of the simulation box to establish a tent-shaped temperature field (phoresis), or (b) by thermostating the “gold cap” at the temperature \( T_p \) by a momentum conserving velocity rescaling procedure (self-phoresis). The fluid at the boundary of the simulation box is always kept at the ambient temperature \( T_0 \), by a similar rescaling procedure. Data acquisition starts once the system has reached a steady state.

The temperature profile at the surface of the Janus particle can directly be inferred from the average kinetic energy of nearby solvent particles. We parameterize the ensuing angular temperature variation in the particle frame by a series of Legendre
Fig. 5. Molecular Dynamics Simulations (adapted from Ref. [37]). (a) Snapshot of a heated Janus particle with wetting parameters $c_{gs} = 2$, $c_{ps} = 1$ on the two hemispheres; coloring indicates the measured kinetic energy from which the continuum temperature field $T(r, \theta)$ depicted in (b) is deduced. (c) Lab frame $(x, y, z)$ and (co-rotating) body frame $(\pi, \theta, z)$. (d) Solvent temperature $T(a, \theta)$ in a thin shell of thickness $0.5\sigma$ around the heated Janus particle, with its cap maintained at temperatures $T_p = 1.20\epsilon/k_B$ ( ), $1.50\epsilon/k_B$ ( ), and $2.00\epsilon/k_B$ ( ). For wetting parameters $c_{gs} = 2$, $c_{ps} = 1$ and $c_{gs} = 1$, $c_{ps} = 2$ (inset). Solid lines represent fits by the series expansion from Eq. (13) and dashed lines solutions of the heat equation with the appropriate temperature-dependent thermal conductivity [27], both truncated after $n = 3$ (which causes spurious oscillations).

polynomials $P_n$,\n
$$T(a, \theta) = \bar{T} \sum_n B_n P_n(\cos \theta),$$

(13)

where $\bar{T}$ is the average temperature of the shell. Truncating the series at $n = 3$ gives rise to some spurious oscillations, but provides a decent description of the data (Fig. 5) and is also close to the theoretical prediction from Fourier’s heat equation for a previously established temperature-dependence of the thermal conductivity of the Lennard–Jones solvent [27] (truncated to the same order).

To obtain the data shown in the following figures, the velocity of the Janus particle was measured both in the lab frame and in the co-moving body frame. The displacements and the velocities in the body frame were obtained by projecting the corresponding quantities from the lab frame at every time step of the simulations. At late times, the mean-square displacements along and perpendicular to the instantaneous propulsion direction of the self-thermophoretic Janus particle deviate from each other, as expected from the superposition of diffusion and ballistic self propulsion (Fig. 6). In the body frame, the propulsion velocity can thus be read off from the asymptotic slope of the mean-square displacement along the propulsion direction (or from the corresponding velocity distribution). In the lab frame, the randomization of the propulsion direction due to the rotational diffusion on the characteristic rotational diffusion time scale $\tau_R$ ultimately renders the particle motion diffusive. (A quantitative prediction for $\tau_R$ follows from the theory of hot Brownian motion, as discussed above.) The mean-squared displacement as function of time (Fig. 6)

$$\langle \Delta r(t)^2 \rangle = 4Dt + 2v^2\tau_R^2 \left[ t/\tau_R + e^{-t/\tau_R} - 1 \right],$$

(14)

thus exhibits a characteristic crossover from ballistic motion at short times $t \ll \tau_R$ to diffusive motion with an effective diffusion coefficient of $D_{eff} = D + v^2\tau_R/2$ at late times $t \gg \tau_R$. 
It is interesting to compare the propulsion velocities of Janus particles moving in their self-generated temperature gradient to those in an external temperature gradient. In the passive phoretic setup, the Janus particle was exposed to a constant temperature gradient along the z-direction of the lab frame. In order to avoid a temperature discontinuity across the periodic simulation boundaries, the temperature profile was actually chosen to be tent-shaped around a central maximum, and particle velocities to the left and right were recorded and correlated separately. Moreover, to quantitatively compare active and passive phoresis, the symmetry axis of the Janus particle was subjected to an angular confinement during passive thermophoresis, so that it remained parallel to the direction of the external gradient. Altogether, four cases were investigated: a homogeneous particle with homogeneous wetting parameters $c_{ps} = c_{ps} = 1$ and $c_{gs} = 2$ and a Janus particle with $c_{gs} = 1$, $c_{ps} = 2$ and $c_{gs} = 2$, $c_{ps} = 1$, respectively. In all four scenarios we observed that the particle moved towards the cold, indicating a positive phoretic mobility (Fig. 7a). Stronger potential attractions correlate with weaker thermophoresis for the homogeneous particles. The measured passive phoretic mobilities of the Janus particle are intermediate between those of the hemispheres and corroborate this trend.

Within the boundary layer approximation, the propulsion velocities for both the passive and the active scenarios is determined by the temperature on the surface of the particle and can be calculated from Eqs. (6), (7). In the simulation, we can discern the finite thickness $s$ of the boundary layer, and find that the temperature changes radially within it. This prohibits a literal application of the boundary layer equations. Instead, we propose here to subsume the two different phoretic mobilities characterizing the two hemispheres of a Janus bead into an effective mobility $\mu_{eff}$, which is taken to be a material parameter of the particle-solvent interface as a whole. The propulsion speeds $v$ in both the active and the passive scenario are then related to the relevant part of the temperature gradient $TB_1/(a + s)$ (which excites the sliplet) by

$$v = -|v_{tp}| = \frac{2}{3} \mu_{eff} TB_1/(a + s).$$

By averaging the temperature field over shells of various thicknesses for an active and a passive Janus particle moving at the same propulsion velocity in the simulation, the effective boundary layer thickness $s$ can thus be estimated by requiring this equation...
(a) Passive phoretic velocities for a homogeneous particle with $c_{ph} = c_{ph} = 1$ (●) and $c_{ph} = c_{ph} = 2$ (○) and a Janus particle with $c_{ph} = 2$, $c_{ph} = 1$ (▲) and $c_{ph} = 2$ (▽) (inset). (b) The component $\overline{T B_1/(a+s)}$ of the surface temperature gradient that causes the propulsion, as measured in fluid shells of various thicknesses $s$ around the Janus bead. The active and passive propulsion velocities are the same for each heating power/temperature gradient, therefore the measured $\overline{T B_1/(a+s)}$ should also coincide for Eq. (15) to hold. Lines guide the eye in inferring the corresponding effective boundary layer thickness $s$ (gray bar).

6 “Hot” Experimental Techniques

**Mechanism and synthesis of hot swimmers:** Hot swimmers can very conveniently be fueled by absorbed light [1] or by an oscillating magnetic field [40]. In principle, also chemical energy could be used to generate heat, but the ensuing diffusiophoretic effects would likely largely overshadow the temperature effects [19, 41]. Note that the heating mechanism itself does not provide a gradient, as in phoresis. It is the asymmetric design of the particle that determines its propulsion direction. In practice, self-thermophoretic swimmers can for example be fabricated by partly covering a small plastic or glass bead by a more strongly absorbing material e.g. gold, or a number of other synthesis procedures, and a variety of more complicated (e.g. chiral) designs are feasible [42], which give rise to more fancy swimming styles [43].

As a simple and common example, Fig. 8 shows an electron microscopy image of a gold-coated polystyrene particle. The gold cap acts as the asymmetric heat source upon homogeneous illumination of the particle. The resulting temperature profile is depicted in Fig. 8 (central panel), as calculated by finite-element simulations for a polystyrene particle with a 50 nm gold cap in water.

**Propulsion velocity:** In a typical experiment, the gold-coated hemisphere of a Janus particle is heated by the absorption of a 532 nm laser. Self-thermophoresis by laser heating exhibits two characteristic properties illustrated by the experimental results in Figure 9: firstly, a linear dependence of the velocity on the incident laser power (i.e. heating power); and secondly a propulsion velocity that is independent of the particle size. Both can be understood from simple arguments. As discussed in the context of Eq. (5) and Eq. (7), the thermophoretic propulsion velocity $v_{tp}$ is the surface average of the negative tangential slip velocity at the particle surface, which
is in turn proportional to the temperature gradient $\nabla_{||} T$ across the particle and its thermal mobility coefficient $\mu(r)$ [1, 44]. The thermophoretic propulsion velocity is thus directly proportional to the temperature gradient. The tangential temperature gradient $\nabla_{||} T$ is proportional to the temperature jump $\delta T$ divided by the particle radius $a$, i.e., $\nabla_{||} T \approx \delta T / a$. The temperature jump $\delta T$ itself is proportional to the power $P_{\text{abs}} = \sigma_{\text{abs}} I_{\text{inc}}$ absorbed in the gold cap, where $\sigma_{\text{abs}}$ is the absorption cross section of the gold cap and $I_{\text{inc}}$ is the incident laser intensity. Altogether, we thus have

$$\nabla_{||} T \approx \frac{\delta T}{a} = \frac{\sigma_{\text{abs}} I_{\text{inc}}}{4\pi\kappa T a^2},$$

with an effective heat conductivity $\kappa T$. One thus arrives at the conclusion that the propulsion velocity is directly proportional to the incident laser intensity $I_{\text{inc}}$ and independent of the particle radius $a$, if the absorption cross section scales with the square of the particle radius, $\sigma_{\text{abs}} \propto a^2$. This is indeed the case for typical microswimmer designs, if the thickness of the gold cap is independent of the particle size, because the absorption cross section of a micron-sized thin gold cap scales with the volume of the cap. For very small particle sizes, additional considerations are required [7].

**Detection methods:** To experimentally study hot swimmers and hot Brownian motion, optical microscopy techniques are very suitable. The paths of individual microswimmers can conveniently be analyzed with the help of optical microscopy (bright field or dark-field microscopy) under constant optical heating of the absorbing gold hemisphere [5, 45]. The tracking concerns two observables: the 3-dimensional orientation of the Janus particle and its position in the lab frame in each exposure. Both are best determined in darkfield microscopy [45] as the light scattering from a metal cap and a polymer particle differ strongly (see Fig. 8). From a series of images one can calculate the displacement vector between images separated by multiples of the inverse framerate [5]. The mean-squared displacement during that time period yields the effective diffusion coefficient of that particle, while the mean displacement vector in the particle frame yields the propulsion velocity.

A class of optical microscopy methods that put the heating to good use for detection, and can thereby detect even nanometer-sized particles and single molecules, are so-called photothermal techniques. They build on photothermal single-particle detec-
Fig. 9. Dependence of the propulsion velocity of Janus particles on the absorbed heating power $P_{abs}$ (left) and the particle radius (right). The particles all have a gold cap of 50 nm thickness but vary in size. Figure adapted from Ref. [7]

**tation methods** [46, 47]. In case of a gold-capped Janus particle, the illumination excites the conduction-band electrons in the gold, which transfer their excitation energy to phonons within some 100 femtoseconds. The heat is then released to the surrounding liquid. A steady-state temperature profile in the solvent is quickly (within a few microseconds) established by heat diffusion, for which the solvent acts as an infinite heat bath. The basic idea of photothermal microscopy is then to exploit the “mirage”, i.e., the induced refractive index change, around the heat source. It acts as a lens that can be detected by another laser, called the probe laser, which is focused into the volume illuminated by the heating laser [47, 48]. If a particle diffuses through the focal volume of such a photothermal microscopy setup, the length of the signal bursts gives information on the drift and diffusion of the particle. The method can be turned into a photothermal correlation spectroscopy (PhoCS) technique [49–51], which is largely equivalent to fluorescence correlation spectroscopy [52, 53]. The main difference is that the optical contrast is not caused by the probe fluorescence but by the emanating heat. The experimental results displayed in the left panel of Fig. 4 have been obtained by a slightly more sophisticated, highly quantitative (twin-focus) implementation of this technique [8].

**Steering by photon nudging:** As mentioned above, the orientation of the Janus particles symmetry axis is also subject to Brownian fluctuations, which randomize the direction of propulsion. Therefore, microswimmers, and nanoswimmers only retain their ballistic trajectories for times shorter than the rotational diffusion time. As this rotational diffusion time scales with the particle radius cubed, small particles lose their directionality already after a few 100 microseconds and reveal an enhanced diffusive motion rather than a ballistic propulsion. A certain degree of built-in persistence of the swimming motion is crucial for applications. To this end, two strategies can be pursued. One can slow down rotational diffusion by designing slender swimmer bodies [54]. Or one can employ feedback-based control mechanisms to rectify the orientational fluctuations and yield more directed motion or even perfectly steered motion [5, 45].

For the latter purpose, self-thermophoretic propulsion by laser heating is well suited, as the propulsion can be switched on and off at will. A successful technique that uses the switchable motion and the information about the current orientation of the
Fig. 10. Photon nudging. Left: sketch of the nudging principle; the propulsion is only switched on if the particle orientation is found within an acceptance angle of a desired target direction. Middle: false-color histograms of particle positions around the target location; trapping is limited by diffusion for small nudging velocities and acceptance angles (top), diffusive and ballistic displacements are balanced for optimum localization (middle), and the particle overshoots the target for too rapid nudging or too large acceptance angles. Right: nudging along a path defined by successive targets (black dots).

Janus swimmer to steer and trap swimmers in solution is called “photon nudging” [45]. During phases of rotational Brownian motion without self-propulsion, the orientation is analyzed in real time. If the particle orientation is by chance found within a certain acceptance angle $\theta$ around the desired target direction, the propulsion is switched on. This causes a net motion of the swimmer towards the target location and finally a localization of the swimmer at the target position. Note that, for homogeneous illumination, this type of steering or trapping does not involve any external forcing nor any external torque (as rotational Brownian motion provides the random reorientation mechanism). It is therefore reminiscent of a Maxwell daemon.

The accuracy of localization by photon nudging is again limited by diffusion, mostly during the off periods, when the particle goes off track. Now, the passive rotational diffusion not only limits the persistence of the path, and thus leads to intermittent rest phases of the swimmer, it also achieves the reorientation during these phases that initiates the next active phase. The naively expected growth of the relative localization error (the square root of the mean-square displacement of the particle from the target location divided by the particle radius) with decreasing particle size can therefore be avoided with the nudging method [5, 45].

7 Conclusion

We have introduced some basic notions and techniques relevant for self-thermophoretic microswimmers and gathered some important recent results concerning their hot Brownian fluctuations, their phoretic boundary layers, and their experimental detection and active feedback control. A more detailed investigation of the precise microscopic conditions within the boundary layer would be a worthwhile task for future numerical work. Ongoing work moreover attempts to put these techniques to good use for the analysis of mutually interacting swimmers and swimmers interacting with liquid-solid boundaries [55]. An important goal would be to establish coarse-
grained hydrodynamic or thermodynamic theories or even a statistical mechanics of many-body swimming [56], along these lines. Our discussion has indicated that one may expect several new and interesting aspects to show up in such theories that are not present in conventional many-body theories for passive particles, chiefly due to the complicated non-equilibrium (and not pairwise additive) interactions and to the non-equilibrium character of the Brownian fluctuations of hot swimmers. These unconventional effects, if well understood and controlled, could possibly again inspire new applications and techniques of manipulation, along the directions outlined here.

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