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Stochastic low Reynolds number swimmers

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Abstract
As technological advances allow us to fabricate smaller autonomous self-propelled devices, it is clear that at some point directed propulsion could not come from pre-specified deterministic periodic deformation of the swimmer's body and we need to develop strategies for extracting a net directed motion from a series of random transitions in the conformation space of the swimmer. We present a theoretical formulation for describing the 'stochastic motor' that drives the motion of low Reynolds number swimmers based on this concept, and use it to study the propulsion of a simple low Reynolds number swimmer, namely, the three-sphere swimmer model. When the detailed balanced is broken and the motor is driven out of equilibrium, it can propel the swimmer in the required direction. The formulation can be used to study optimal design strategies for molecular scale low Reynolds number swimmers.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Biological molecular motors [1] are ingenious nanoscale machines that convert chemical energy into directed mechanical work amid strong thermal fluctuations. With the current miniaturization trend in technology, one naturally wonders whether it is possible to synthesize devices with similar functionalities [2]. In particular, it is desirable as a first step to design autonomous small scale swimmers, which could later on be steered by coupling to a guiding network or system. These swimmers could be used in carrying cargos or stirring up fluids at small scales.

There is a significant complication in designing swimmers at small scale as they have to undergo non-reciprocal deformations to break the time-reversal symmetry and achieve propulsion at low Reynolds number [3]. While it is not so difficult to imagine constructing motion cycles with the desired property when we have a large number of degrees of freedom at hand—like Nature does, for example—this will prove nontrivial when we want to design something with only a few degrees of freedom and strike a balance between simplicity and functionality, like most human-engineered devices [4]. Recently, there has been an increased interest in such designs [5–23] and two interesting examples of such robotic microswimmers have been realized experimentally using magnetic colloids attached by DNA linkers [24, 25]. Among others, a simple swimmer model based on spheres connected by arms that do not interact with the fluid [5] has been recently used for a number of studies including scattering of two swimmers [17, 19], collective hydrodynamic coupling of swimmers [18, 20], general feasibility of various design properties of swimmers [21], and the effect of large cargos on the performance of swimmers [22]. While constructing small swimmers that generate surface distortions is a natural choice, it is also possible to take advantage of the general class of phoretic phenomena to achieve locomotion—as they become predominant at small scales—as recent experimental [26–29] and theoretical [30–32] works have demonstrated.

Here we construct a general statistical mechanical formulation for studying low Reynolds number swimmers that undergo conformational changes in a stochastic manner pertinent to systems of molecular scale. We attribute transition rates to each deformation move or swimming stroke, and calculate the propulsion velocity as a function of these rates. Our formulation provides a general prescription as regards how to construct the relevant portions of the configurational space of swimmers, and how to take advantage of the complexities in this space to maximize the efficiency of the swimmer. We apply the formulation to the specific example of the three-sphere swimmer model, which yields interesting results.

The rest of the paper is organized as follows. Section 2 describes the general formulation of hydrodynamics of low Reynolds number swimmers, and it is followed by section 3 that is devoted to the statistical mechanics of the conformational changes in swimmers. The formulation is applied to the example of the three-sphere swimmer model in section 4, which is followed by concluding remarks in section 5.

2. Hydrodynamics of low Reynolds number swimming

We consider a deformable extended body as a system composed of N point-like solid components described in terms of their position vectors $\mathbf{r}^\alpha(t)$. The deformation is related to internal forces exerted between these solid components, so that on component α there is exerted a net force $\mathbf{f}^\alpha(t)$, that is in turn applied onto the fluid at $\mathbf{r}^\alpha(t)$. In our description of point-like objects, the hydrodynamic interactions between the objects relate these forces to the velocities of the components $\mathbf{v}^\alpha(t) = \dot{\mathbf{r}}^\alpha(t)$ via the Oseen tensor $\mathcal{H}_{ij}(\mathbf{r}, \mathbf{r}')$ [33] (italic indices describe spatial components), namely

$$v_i^\alpha = \sum_\beta M_{ij}^{\alpha\beta} f_j^\beta, \tag{1}$$

where $M_{ij}^{\alpha\beta} = \mathcal{H}_{ij}(\mathbf{r}^\alpha, \mathbf{r}^\beta)$ and summation over repeated indices that define the vector components is understood. The Oseen tensor is the Green function for the Stokes equation with the appropriate boundary conditions and its explicit form depends on the problem that we are considering. For example, in the simplest case we can treat the solid particles as point-like and use the $1/r$-type expressions for the off-diagonal components of the Oseen tensor, while putting in $1/(6\pi\eta a)$ for the diagonal components where a is the radius of the particles and η is the viscosity of water. If necessary, one could also incorporate finite size corrections and the effect of confining boundaries by using the appropriate form of the Green function.

We can now invert equation (1) as

$$f_i^\alpha = \sum_\beta N_{ij}^{\alpha\beta} v_j^\beta, \tag{2}$$

where $N_{ij}^{\alpha\beta}$ is the resistance (friction) tensor that satisfies $\sum_\beta M_{ij}^{\alpha\beta} N_{jk}^{\beta\gamma} = \delta_{\alpha\gamma}\delta_{ik}$.

For a swimmer that is not subjected to external forces, the local and instantaneous forces in the body are subject to the constraint

$$\sum_\alpha \mathbf{f}^\alpha = 0, \tag{3}$$

which yields

$$\sum_{\alpha,\beta} N_{ij}^{\alpha\beta} v_j^\beta = 0. \tag{4}$$

Similarly, if the swimmer is not subjected to a net external torque, an additional constraint applies:

$$\sum_\alpha (\mathbf{r}^\alpha - \mathbf{r}^{\mathrm{CM}}) \times \mathbf{f}^\alpha = 0, \tag{5}$$

where the center of mass (CM) position is defined as $\mathbf{r}^{\mathrm{CM}} = \frac{1}{N}\sum_\alpha \mathbf{r}^\alpha$. We note that this condition might not in general be satisfied, as in the case of a recent experiment on magnetic doublets [25]. When it does hold, however, it will introduce additional constraints on the type of motion and conformations that we can prescribe for the system. Finally, for sufficiently symmetric systems the torque-free constraint might automatically be satisfied [21].

We now assume that the relative positioning of the body components is prescribed, in a reference frame that moves with the average position and orientation of the body. This reference frame, which we call the 'body frame' hereafter, will be constant during one cycle of the deformation in the body. As a result of the deformation, over the period of one cycle the object is expected to be displaced by a small amount due to a non-vanishing translational swimming velocity and rotated slightly if there is a non-vanishing rotational velocity as well. The combination of the displacement and rotation will determine the new position and orientation of the body frame, which will be used in the calculation of the next step of the motion and so on. Therefore, in this picture the motions are grouped into separate slow and fast degrees of freedom, in the sense that what is happening over one deformation in a cycle (fast degrees of freedom) will be averaged to determine a net change in the slow degrees of freedom that determine the overall average translation and rotation of the swimmer through the liquid along its trajectory.

We now assume that the relative positioning of the body components $R_i^{\alpha\beta} \equiv r_i^\alpha - r_i^\beta$ is known in the body frame, which means that the relative velocities $v_i^\alpha - v_i^\beta = \dot{R}_i^{\alpha\beta}$ are also known[3]. These relative positions and relative velocities need to be prescribed in a such a way that all the necessary geometrical constraints are satisfied, as for example, one cannot expect to have arbitrary distances between a number of points that form a body of a given shape.

If the shape of the object and the conformational changes are sufficiently symmetric that the object swims on average in a rectilinear fashion, averaging the velocity of any tagged component α over a complete cycle yields the total average translational velocity of the body,

$$\langle \mathbf{v}^\alpha \rangle = \mathbf{V}^{\mathrm{trans}}, \tag{6}$$

as the difference between the velocity of the α component and that of the whole body will be in the form of relative deformations that average out to zero. For a more general case the object will have a rotational component superimposed with the translational one, and the average velocity of the tagged body component in the body frame will have the following form:

$$\langle \mathbf{v}^\alpha \rangle = \mathbf{V}^{\mathrm{trans}} + \boldsymbol{\Omega}^{\mathrm{rot}} \times \langle (\mathbf{r}^\alpha - \mathbf{r}^{\mathrm{CM}}) \rangle, \tag{7}$$

where $\boldsymbol{\Omega}^{\mathrm{rot}}$ is the angular velocity vector of the body about the center of mass. We can extract the translational velocity as

$$V_i^{\mathrm{trans}} = \frac{1}{N}\sum_\alpha \langle v_i^\alpha \rangle, \tag{8}$$

[3] Note that getting the actual form of $R_i^{\alpha\beta}(t)$ from the internal motion of the object may require calculations that involve the force-free and the torque-free relations.

and the rotational component of the velocity as

$$\Omega_i^{\rm rot} = I_{ij}^{-1} \sum_\alpha \epsilon_{jkl} \langle (r_k^\alpha - r_k^{\rm CM}) \rangle \langle v_l^\alpha \rangle, \tag{9}$$

where

$$I_{ij} = \sum_\alpha \delta_{ij} \langle (r_k^\alpha - r_k^{\rm CM}) \rangle \langle (r_k^\alpha - r_k^{\rm CM}) \rangle - \langle (r_i^\alpha - r_i^{\rm CM}) \rangle \langle (r_j^\alpha - r_j^{\rm CM}) \rangle \tag{10}$$

is the average moment of inertia tensor for the object.

We can single out the velocity of one of the components, say $\alpha = 1$, and describe all of the velocities in terms of this and the prescribed relative velocities, namely $v_i^\alpha = v_i^1 + \dot{R}_i^{\alpha 1}$. Putting this back in equation (4), we find

$$v_i^1 = -L_{ij}^{-1} \sum_{\alpha,\beta} N_{jk}^{\alpha\beta} \dot{R}_k^{\beta 1}, \tag{11}$$

where $L_{ij} = \sum_{\alpha,\beta} N_{ij}^{\alpha\beta}$. Note that one can also choose to specify the forces/tensions in the links instead of the relative velocities. In this case it will be straightforward to modify the formulation and calculate the velocities. A more general framework would encompass prescriptions relating stresses and deformations.

We can write the relative positioning of the components in the body frame as $R_i^{\alpha\beta}(t) = R_{0\,i}^{\alpha\beta} + u_i^{\alpha\beta}(t)$, where the $u_i^{\alpha\beta}$ denote the deformations of the body about the average shape described by $R_{0\,i}^{\alpha\beta}$. If we assume that the deformations of the body are relatively small, we can expand equation (11) in powers of the deformations and obtain an expression for the instantaneous velocity of the tagged ($\alpha = 1$) component of the body as

$$v_i^1(t) = \sum_{\alpha,\beta} A_{ij}^{(1)\alpha\beta} \dot{u}_j^{\alpha\beta} + \sum_{\alpha,\beta,\gamma,\delta} B_{ijk}^{(1)\alpha\beta\gamma\delta} \dot{u}_j^{\alpha\beta} u_k^{\gamma\delta} + \sum_{\alpha,\beta,\gamma,\delta,\mu,\nu} C_{ijkl}^{(1)\alpha\beta\gamma\delta\mu\nu} \dot{u}_j^{\alpha\beta} u_k^{\gamma\delta} u_l^{\mu\nu} + \cdots, \tag{12}$$

where the coefficients $A_{ij}^{(1)\alpha\beta}$, $B_{ijk}^{(1)\alpha\beta\gamma\delta}$, $C_{ijkl}^{(1)\alpha\beta\gamma\delta\mu\nu}$, etc are purely geometrical pre-factors (i.e. involving only the characteristic length scales describing the shape of the body). Averaging over a full cycle, the contribution due to the linear terms $\dot{u}_j^{\alpha\beta}$ and the symmetric combinations $\dot{u}_j^{\alpha\beta} u_k^{\gamma\delta} + \dot{u}_k^{\gamma\delta} u_j^{\alpha\beta} = {\rm d}(u_j^{\alpha\beta} u_k^{\gamma\delta})/{\rm d}t$ vanish. Therefore, to the leading order, we find the average swimming velocity as

$$V_i^{\rm trans} = \frac{1}{N} \sum_\mu \langle v_i^\mu \rangle = \frac{1}{2} \sum_{\alpha,\beta,\gamma,\delta} B_{ijk}^{\alpha\beta\gamma\delta} \langle \dot{u}_j^{\alpha\beta} u_k^{\gamma\delta} - \dot{u}_k^{\gamma\delta} u_j^{\alpha\beta} \rangle = \sum_{\alpha,\beta,\gamma,\delta} B_{ijk}^{\alpha\beta\gamma\delta} \left\langle \frac{\Delta \mathcal{A}_{jk}^{\alpha\beta\gamma\delta}}{\Delta t} \right\rangle, \tag{13}$$

where $\Delta \mathcal{A}_{jk}^{\alpha\beta\gamma\delta}$ is the area element enveloped by the trajectory of the system in the $(u_j^{\alpha\beta}, u_k^{\gamma\delta})$ space, and $B_{ijk}^{\alpha\beta\gamma\delta} = \frac{1}{N} \sum_\mu B_{ijk}^{(\mu)\alpha\beta\gamma\delta}$. Note that $\Delta \mathcal{A}_{jk}^{\alpha\beta\gamma\delta}/\Delta t$ is not a complete time derivative, and its average over a full cycle does not vanish. A similar expression can be written for the angular velocity. The averaging here denotes time averaging if the conformation of the system is prescribed. If, however, the system undergoes stochastic conformational changes, the averaging needs to be performed over the distribution of the various conformations. The formulation needed to carry out this step of the calculation is developed in the next section.

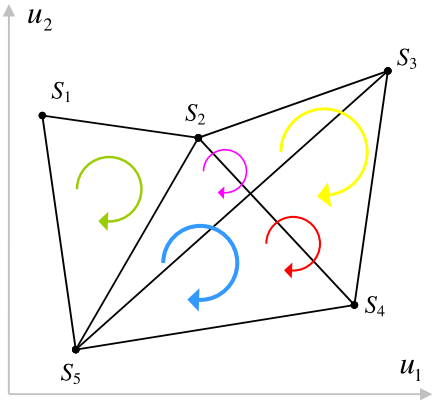

Figure 1. A typical conformation subspace describing the shape of the swimmer. Distinct conformational states are identified and connected to one another when transitions are permissible, making a graph. The swimming velocity will be determined by the sum of the currents in each loop of the graph (denoted by different arrows here), weighted by the area of each loop, correspondingly (see equation (16)).

3. Kinetics in the conformation space

Let us now consider a conformation subspace of the system corresponding to two representative deformations u_1 and u_2 (see figure 1). Since we aim to model molecular systems, we should take into account the stochastic nature of the conformational changes and not prescribe a deterministic trajectory for the deformation of the system. We identify distinct conformational states of the system, denoted as S_n, and construct a kinetic description where the deformations of the system are described as transitions between these states with given rates, assuming that they occur one at a time and do not overlap with each other. We denote the probability of finding the system in S_n as P_n and the rate for transition $m \to n$ as k_{nm}. These probabilities are normalized as $\sum_n P_n = 1$. Connecting the states that have permissible transitions between them with links, we find a graph that characterizes the conformational kinetics of the system in each subspace, as seen in figure 1. To every link, we can attribute a probability current

$$J_{\langle nm \rangle} = k_{mn} P_n - k_{nm} P_m, \tag{14}$$

and at stationary state we can impose the continuity of current at every node, namely

$$\sum_m J_{\langle nm \rangle} = 0. \tag{15}$$

Solving the system of equations, we can find all probabilities and currents, and in particular the currents $J(\alpha)$ running through all the loops in the graph (see figure 1). We can then write

$$\left\langle \frac{\Delta \mathcal{A}}{\Delta t} \right\rangle = \sum_\alpha \mathcal{A}(\alpha) J(\alpha), \tag{16}$$

where $\mathcal{A}(\alpha)$ is the area enclosed by loop α in the conformation subspace. Equation (16) shows that the contributions from the different loops act together analogously to circuits *in parallel*, and therefore, it will be the fastest route that will determine the effective swimming velocity. In each loop, however, the different legs are connected *in series*, and the slowest kinetic leg will control the contribution to the effective swimming velocity from each loop (see the example below).

4. Example: three-sphere swimmer model

We now focus on the specific example of a three-sphere swimmer model [5]. We define the conformation space of the swimmer using the two variables (u_1, u_2) that describe the longitudinal deformation of the two arms of the swimmer. We assume that the two arms can be in the two states corresponding to $u_i = 0$ and $u_i = \delta_i$, and transit from one to the other in an almost instantaneous fashion. This means that the configuration space of the swimmer will be made up of four distinct states as shown in figure 2, defined by different values of the pair (u_1, u_2), namely: state A for (δ_1, δ_2), state B for $(\delta_1, 0)$, state C for $(0, 0)$, and state D for $(0, \delta_2)$. We then assign transition rates to the system, corresponding to the average rate of opening and closing of the arms along the cycle

$$\mathrm{A} \underset{k_{AB}}{\overset{k_{BA}}{\rightleftharpoons}} \mathrm{B} \underset{k_{BC}}{\overset{k_{CB}}{\rightleftharpoons}} \mathrm{C} \underset{k_{CD}}{\overset{k_{DC}}{\rightleftharpoons}} \mathrm{D} \underset{k_{DA}}{\overset{k_{AD}}{\rightleftharpoons}} \mathrm{A}. \tag{17}$$

Note that in this simple example there is only one loop in the conformation space graph of the system (figure 2).

We can now calculate the swimming velocity as a function of the transition rates. Using the general formulation described in sections 2 and 3, we find

$$V = K\delta_1\delta_2 J, \tag{18}$$

where $K = \frac{a}{3}[\frac{1}{\ell_1^2} + \frac{1}{\ell_2^2} - \frac{1}{(\ell_1+\ell_2)^2}]$ with ℓ_1 and ℓ_2 being the undeformed lengths of the two arms and a being the radius of the spheres [21]. The probability current J is a function of the transition rates, which can be obtained from the following straightforward algebra. At steady state, the current conservation equations can be written as $J = k_{BA}P_A - k_{AB}P_B = k_{CB}P_B - k_{BC}P_C = k_{DC}P_C - k_{CD}P_D = k_{AD}P_D - k_{DA}P_A$, which provide us with four equations for the current and the four probabilities, which are also normalized as $P_A + P_B + P_C + P_D = 1$. Solving the system of linear equations, we find

$$J = (k_{AD}k_{DC}k_{CB}k_{BA} - k_{AB}k_{BC}k_{CD}k_{DA}) \times \left\{ \sum_{\text{replace A by B, C, D}} (k_{AD}k_{DC}k_{CB} + k_{AB}k_{BC}k_{CD} + k_{AB}k_{AD}k_{DC} + k_{AD}k_{AB}k_{BC}) \right\}^{-1}. \tag{19}$$

Equations (18) and (19) give the swimming velocity of the three-sphere swimmer [23].

From equation (19) it is clear that if detailed balance holds, then J vanishes as the numerator is zero. Using the average steady state current, we can deduce the average period of completing one full cycle of the motion along the A → B → C → D → A loop as

$$T = J^{-1}. \tag{20}$$

We can gain useful insight by looking at the particular limit where the forward rates are all much higher than the corresponding backward ones ($k_{BA} \gg k_{AB}$, etc). In this limit, we find

$$T = k_{AD}^{-1} + k_{DC}^{-1} + k_{CB}^{-1} + k_{BA}^{-1}, \tag{21}$$

which means that the period for a full cycle is the sum of the time intervals needed to complete each leg of the cycle.

As another example, we can assume that all of the equilibrium rates $k_{\beta\alpha}$ are equal to 1 (for simplicity), and that by external action just one of them is modified to $k_{BA} = 1 + \epsilon$. In this case, one can show that equation (19) yields

$$J = \frac{\epsilon}{16 + 6\epsilon}, \tag{22}$$

which leads to a velocity proportional to the perturbation for small values of ϵ and independent of it if the perturbation is very large. The linear dependence can be easily understood for a system that is only slightly driven out of equilibrium, and the saturation at large perturbations occurs because the cycling will then be limited by the other three unperturbed transitions. In general, one can see that the slowest leg of the reaction controls the average rate of full cyclic motion, which suggests the interpretation that in each loop the different legs are connected in series, by analogy to circuits.

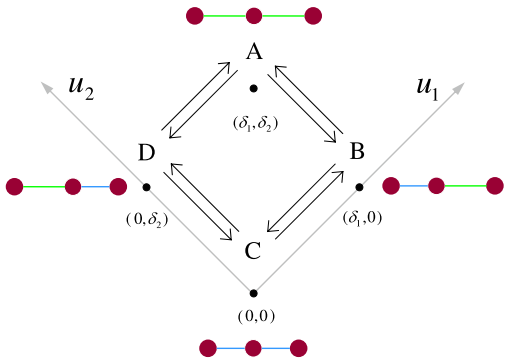

Figure 2. Conformation space of the three-sphere swimmer model. This minimal model involves only one loop. The convention is such that a net swimming to the right requires the system to make more cycles in the clockwise direction than in the counterclockwise direction.

5. Conclusion

We have presented a general formulation that can be used in studying the swimming of a small object that undergoes stochastic deformations. The program to follow to this end has two stages. (1) Treat the deformations as prescribed and follow the hydrodynamic formulation of section 2 to calculate the average swimming velocity in terms of the

relevant deformation variables. (2) Construct the conformation space of the system on the basis of the deformation variables and follow the statistical mechanical description of section 3 to work out the contributions to the net swimming velocity of various *modes* of swimming defined as loops in the conformation space. We found that a useful circuits analogy can be invoked to describe the efficiency of the swimming, with two notable features. (1) The different modes of swimming can be effectively considered to act *in parallel*, which means that their contributions will be independently added to each other to yield the net swimming velocity and therefore the *fastest* route will be the dominant mode of swimming controlling the velocity. (2) In each loop, the different kinetic legs could be considered as acting *in series* with respect to one another, which means that the *slowest* kinetic leg will control the net contribution to the velocity from the loop.

The formulation also allows us to study the effect of an external force or load on the performance of swimmers. External forces both add to the hydrodynamic drag and affect the performance of the swimming strokes as activated moves, as the deformations will involve doing work against or being helped by forces endured by the arms. These forces will modify the transition rates, and their effects can be readily accommodated by using the force-dependent rates in the kinetic formulation. This effect has been studied for the three-sphere swimmer model, which has revealed that the performance of the motor strongly depends on where the force is exerted [23]. This shows that for such small swimmers, the concept of a generic force–velocity response breaks down, which might have interesting implications for designing molecular swimmers.

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