Mesoscale hydrodynamic simulations
– From a single colloid to the collective dynamics of microswimmers

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Introduction

Animals, such as flocks of birds, mammalian herds, schools of fish or bacteria colonies, show fascinating non-equilibrium collective phenomena [1]. Different models, from continuum descriptions to particle-based simulations have been applied to explore such systems, which are nowadays summarized under the term active matter [1–3]. While we can observe flocks of birds by our own eyes, the fascinating non-equilibrium dynamics of microswimmers, such as spermatozoa, bacteria, protozoa, and algae is invisible until viewed under the microscope. And it turns out that the physics of microswimmers is quite different from our everyday experience. When swimming, we are used to push fluid backwards to propel ourselves, exploiting inertia. For microswimmers, viscous forces dominate over inertial forces, and hence, render such a strategy ineffective. The ratio of viscous to inertial forces is known as Reynolds number. At zero Reynolds number, a scallop, which slowly opens and quickly closes its shell, cannot swim [4]. More generally, every reciprocal motion, i.e., every periodic motion of a swimmer that looks identical when viewed forwards or backwards in a movie, will not result in self-propulsion. This so called scallop theorem [4, 5] is overcome by microswimmers in different ways. Spermatozoa send a bending wave along their flexible tail, while bacteria such as *E. coli* form rotating helices with their flagella, lash-like appendages protruding from their body [3]. Ciliates like *Paramecium* are covered with thousands of flagella – called cilia in this context – which beat in metachronal waves to achieve self-propulsion [3]. Microswimmers, living the “life at low Reynolds number” [4] offer a wide spectrum of interesting and exciting phenomena. In confinement, microswimmers exhibit activity-induced surface accumulation and the bacterium *E. coli* was shown to swim in circles above a surface, as a consequence of hydrodynamic interactions [6, 7]. In channel flows, simplified equations of motion for a microswimmer can be mapped to a Hamiltonian system exhibiting nonlinear dynamics and oscillatory trajectories [8, 9]. For the bacterium *E. coli* and the bloodstream parasite *Trypanosoma brucei* upstream swimming in external flow was observed, which might be relevant for bacterial transport and microbial pathogenesis [9, 10].

Artificial microswimmers, such as Janus particles – named after the two-faced Roman god – and self-propelled liquid droplets, have been designed as generic and easily controllable model systems [11]. Contrary to biological swimmers, they do not swim by performing shape changes, but rather by a self-generation of gradients. Self-diffusiophoretic Janus colloids [12] for example, are dielectric colloids, partially coated with thin layers of catalytic materials. When immersed in a solution of
**Introduction**

H$_2$O$_2$ the chemical reactions on one side of the Janus colloid create a concentration gradient which results in self-diffusiophoresis, i.e., a spontaneous motion of the colloid in the self-generated concentration gradient. A similar mechanism, exploiting a self-generated temperature gradient, is termed self-thermophoresis and induced by absorption of laser light at the metal-coated side of a Janus colloid [13]. Theoretical studies suggest that the swimming of self-phoretic colloids is mainly determined by an induced hydrodynamic slip velocity at the particles’ surface [14]. In a simplified model, we can prescribe a surface velocity on a colloid to describe self-phoresis, independent of the underlying mechanism (e.g., self-diffusiophoresis or self-thermophoresis) [14]. The hydrodynamic model corresponding to a prescribed surface velocity on a spherical colloid is known as the squirmer model and was originally conceived to model self-propulsion of ciliates, such as *Paramecium* [15–17]. In this context, it is argued that the time-averaged beating of cilia on the surface of ciliates generates an effective slip velocity.

Recently, motility-induced phase separation of artificial and biological microswimmers has gained considerable attention [3, 11, 18–20]. In contrast to dilute suspensions of passive colloids, microswimmers or active particles form clusters even when the inter-particle interactions are purely repulsive [21–23]. This motility-induced phase separation is still not fully understood. Several studies are focused on establishing links between the non-equilibrium physics of self-propelled particles and equilibrium statistical physics [24–28]. Analytical investigations typically exclude hydrodynamic interactions, since they are cumbersome to handle, and many interesting collective phenomena already emerge without them. The role of hydrodynamics for cluster formation of microswimmers has been studied in numerical simulations, but contradictory conclusions have been reached [29–31] on whether hydrodynamics enhances or suppresses phase separation. In this thesis we want to establish the role of hydrodynamics for swimmers with isotropic and anisotropic shapes. Thereby, we will apply the squirmer model, as it is a generic model for both, biological, and artificial microswimmers.

To perform such a study, we need a versatile hydrodynamic simulation technique, which includes thermal fluctuations, since the energy scales of microswimmers are typically of the order of the thermal energy scale $k_BT$. We will employ the multi-particle collision dynamics (MPC) method [32–34], a particle-based mesoscale hydrodynamic simulation technique, which – in contrast to various other methods – intrinsically accounts for thermal fluctuations. The MPC fluid consists of point particles, which undergo streaming and subsequent collision steps. While the particles move ballistically in the streaming step, the interactions are accounted for in the collision step. Here, particles are grouped into cells, where a local stochastic process changes the particles’ velocities in a manner that conserves mass and momentum. This implementation of conservation laws gives rise to hydrodynamic behavior on large length and long time scales. In fact, it has been shown by a Chapman-Enskog expansion that the MPC fluid obeys the Navier Stokes equations with an ideal-gas
equation of state [32, 35], and an H-theorem has been derived for the method [32, 36]. Furthermore, the locality of the algorithm allows for an efficient parallel implementation on a computer [37]. Apart from microswimmers, MPC has been employed to study a broad range of soft matter systems including colloids [38–40], polymers [41–43], vesicles [44], and cells [45, 46]. The MPC method has been invented nearly two decades ago [32] and has since been thoroughly studied and extended. Beyond those studies, we found two unresolved issues that needed further investigations which we will discuss in part II of this thesis. The hydrodynamic properties of the MPC fluid are determined by its transport coefficients. While the shear viscosity has been studied extensively, the literature regarding its bulk viscosity is scarce and contradictory. We will study the bulk viscosity of MPC both analytically and by simulations in chapter 5. Another issue which needs to be discussed, is the importance of angular momentum conservation in MPC. The original MPC method does not conserve angular momentum [32], but angular momentum conserving extensions have been developed [47]. Although a study of circular Couette flow [48] found that a lack of angular momentum conservation yields unphysical torques, many studies still apply the original MPC method. We will discuss the consequences of a lack of angular momentum conservation in systems containing hydrodynamic slip boundary conditions in chapter 6.

Further down the road to a simulation study of many squirmers, we enter the area of colloids in part III. The implementation of colloids in MPC has been thoroughly studied. However, as we will find in chapter 7, the existing analysis of the so-called Enskog friction on colloids is flawed, since the lack of angular momentum in the original MPC method was not correctly accounted for. We will present an extensive study on the long-time hydrodynamic friction and short-time Enskog (or local) friction in chapter 7.

Recently, it was put forward that the observation of a colloidal particle’s Brownian motion could be exploited to deduce properties of its environment [49, 50], such as surface elasticity [51] or adsorption layers [49, 52] on a nearby surface. This idea of a colloid as a sensor has been adapted in ref. [53]. The distance of a colloid trapped by optical tweezers above a planar surface was determined by observation of its thermal movement parallel to the surface. However, in ref. [54] it was argued that the theoretical foundation of ref. [53] is incorrect. We will study the experimental setup by MPC simulations to verify this claim in chapter 8.

Finally, in part IV, we study the dynamics of microswimmers. Since we are interested in spherical as well as elongated microswimmers, we introduce a model of a prolate spheroidal squirmer. Since the force dipole singularity is the highest order contribution to the multipole expansion of a microswimmer’s flow field, its inclusion is crucial to model hydrodynamic interactions. However, existing models of spheroidal squirmers either do not account for the force dipole singularity or do not yield an analytically solvable boundary value problem. We will develop a model which solves this problem. Subsequently we implement the model in MPC using a
rigid body Verlet integrator and anisotropic potentials for steric interactions. This enables us to study the behavior of two cooperatively swimming spheroidal squirmers in chapter 10 and more importantly to finally elucidate the role of hydrodynamics in motility-induced phase separation in chapter 11.

But before we dive into our studies on MPC, colloids and microswimmers, we will outline the necessary theoretical and methodological framework for those studies in part I. Since the motion of a microswimmer can be described by a combination of hydrodynamics and rigid body dynamics, we recapitulate the theory of rigid bodies in chapter 1. Thereby we focus on a quaternion-based description, which allows us to formulate an elegant singularity-free numerical integration scheme. We outline the basics of hydrodynamics in chapter 2. In the derivation of the hydrodynamic equations from conservation laws, we explicitly discuss angular momentum conservation, which is often neglected in lectures or text-books. This discussion is particularly important with regard to the study of MPC variants without angular momentum conservation. Furthermore, we include a detailed section on the hydrodynamics of microswimmers, which lays the foundation for the development and study of our model of a spheroidal squirmer in chapter 9. Since thermal fluctuations are intrinsic to MPC and affect the dynamics of colloids and microswimmers, we give an introduction to Brownian motion in chapter 3. Thereby, we discuss colored noise, which we will meet again in our studies of colloids in part III. Moreover, we discuss the numerical integration of the Langevin equation of a rigid body, which we will need for our studies on the collective dynamics of microswimmers in chapter 11. Finally, in chapter 4 we introduce the MPC method, which will accompany us in all subsequent studies.
Part I

Methods
Chapter 1

Rigid body dynamics

In this chapter we briefly summarize the dynamical equations of rigid bodies with an emphasis on a quaternion-based description. Additionally, we present a method for the numerical integration of rigid body dynamics. But before we can discuss the physics of rigid bodies, we need to study the underlying mathematics, namely the algebra of rotations.

1.1 Rotations

1.1.1 Rotation matrices

A rotation is a linear map that fixes the origin, preserves the length of vectors, and preserves the orientation of basis vectors. The first condition tells us that the action of a rotation on a vector \( \mathbf{r} \) can be represented by a matrix multiplication \( \mathbf{r}' = \mathbf{M} \mathbf{r} \).

The second condition demands \( r'^2 = r^2 = \mathbf{r}'^T \mathbf{M}^T \mathbf{M} \mathbf{r} \) and hence \( \mathbf{M}^T = \mathbf{M}^{-1} \), i.e., the matrix needs to be orthogonal. With Cartesian unit vector \( \mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z \), the orientation preserving condition reads
\[
(M \mathbf{e}_x) \cdot [(M \mathbf{e}_y) \times M \mathbf{e}_z] = \mathbf{e}_x \cdot (\mathbf{e}_y \times \mathbf{e}_z),
\]
which is \( \sum_{\alpha \beta \gamma} M_{\alpha x} \varepsilon_{\alpha \beta \gamma} M_{\beta y} M_{\gamma z} = 1 \) in index notation, or \( \det \mathbf{M} = +1 \). Here, \( \varepsilon_{\alpha \beta \gamma} \) is the third order permutation tensor. One can easily see that these orthogonal matrices of determinant 1 form a group. This group is known as \( SO(3) \), the special orthogonal group of order 3.

We will now work out an explicit expression for a rotation matrix representing the rotation about an axis \( \mathbf{n} = (n_x, n_y, n_z)^T, |\mathbf{n}| = 1 \), by an angle \( \psi \). First, we consider the rotation by an infinitesimal angle \( d\psi \) [56]. The infinitesimal displacement \( d\mathbf{r} \) of \( \mathbf{r} \) due to the rotation is (see fig. 1.1)
\[
d\mathbf{r} = d\psi \mathbf{n} \times \mathbf{r}.
\]

To integrate this equation, we exploit the fact that the cross product can be repre-
Chapter 1 Rigid body dynamics

Figure 1.1: Sketch of a small rotation about an axis \( n \) by an angle \( d\psi \). Here \( r' \) is the image of \( r \) under rotation. The small displacement is \( dr = r' - r \) (blue). When \( dr \) becomes infinitesimal, it is tangent to the dashed circle and \( dr = d\psi n \times r \).

Presented by the action of a skew-symmetric matrix\(^a\), i.e.,

\[
{n \times r} = C_n r, \quad \text{where} \quad C_n = \begin{pmatrix} 0 & -n_z & n_y \\ n_z & 0 & -n_x \\ -n_y & n_x & 0 \end{pmatrix}.
\]

Hence, the solution of eq. (1.2) is given in terms of a matrix exponential \[56\]

\[
r(\psi) = \exp(C_n \psi) r(0).
\]

With some algebra, we can establish a closed formula for the rotation matrix \( M = \exp(C_n \psi) = \sum_{k=0}^{\infty} C_n^k \psi^k / k! \). We note that

\[
C_n^2 = \begin{pmatrix} -n_z^2 - n_y^2 & n_x n_y & n_x n_z \\ n_x n_y & -n_z^2 - n_x^2 & n_y n_z \\ n_x n_z & n_y n_z & -n_y^2 - n_z^2 \end{pmatrix}, \quad C_n^3 = -C_n,
\]

and hence \( C_n^{2k} = (-1)^{k+1} C_n^2 \) and \( C_n^{2k+1} = (-1)^k C_n \). Therefore \[56\],

\[
M = \exp(C_n \psi) = 1 + \sum_{k=1}^{\infty} \frac{C_n^{2k} \psi^{2k}}{(2k)!} + \sum_{k=0}^{\infty} \frac{C_n^{2k+1} \psi^{2k+1}}{(2k+1)!}
\]

\[
= 1 - C_n^2 (\cos \psi - 1) + C_n \sin \psi
\]

\[
= \begin{pmatrix} n_x^2 (1 - c) + c & n_x n_y (1 - c) - n_z s & n_x n_z (1 - c) + n_y s \\ n_y n_x (1 - c) + n_z s & n_y^2 (1 - c) + c & n_y n_z (1 - c) - n_x s \\ n_z n_x (1 - c) - n_y s & n_z n_y (1 - c) + n_x s & n_z^2 (1 - c) + c \end{pmatrix},
\]

\[a\) A matrix \( M \) is called skew symmetric if \( M^T = -M \)
with the abbreviations \( c = \cos \psi \) and \( s = \sin \psi \) in the last equality. This identity is known as Rodrigues’ rotation formula and the derivation given here is based on the fact that the skew symmetric matrices are the Lie algebra of \( SO(3) \) [57].

### 1.1.2 Rotation quaternions

As a set, the quaternions \( \mathbb{H} \) are isomorphic to the vector space \( \mathbb{R}^4 \) [55]. A quaternion \( q \) is represented as

\[
q = q_0 1 + q_1 i + q_2 j + q_3 k
\]  

(1.7)

with the basis elements \( 1, i, j, k \). \( \mathbb{H} \) has three operations. The sum of two quaternions is defined as in \( \mathbb{R}^4 \). Similarly, the product of a real number with a quaternion \( q \) is defined as the product by a scalar in \( \mathbb{R}^4 \). Finally, to define the associative quaternion product, we first define the product of basis elements. All other products are then defined using the distributive law. First of all, \( 1 \) is the identity element. Then, all possible basis element products are determined by the identities

\[
i^2 = j^2 = k^2 = ijk = -1.
\]  

(1.8)

The conjugate of a quaternion \( q = q_0 1 + q_1 i + q_2 j + q_3 k \) is given by \( \bar{q} = q_0 1 - q_1 i - q_2 j - q_3 k \). The square of the norm of \( q \) for \(|q| \neq 0\) is \( q^{-1} = \bar{q}/|q|^2 \).

Quaternions can also be represented as scalar-vector pairs

\[
q_0 1 + q_1 i + q_2 j + q_3 k \leftrightarrow (q_0, \mathbf{v}_q) \quad \text{with} \quad \mathbf{v}_q = q_1 e_x + q_2 e_y + q_3 e_z.
\]  

(1.9)

In this representation, we find for the quaternion product

\[
(q_0, \mathbf{v}_q)(p_0, \mathbf{v}_p) = (q_0 p_0 - \mathbf{v}_q \cdot \mathbf{v}_p, q_0 \mathbf{v}_p + p_0 \mathbf{v}_q + \mathbf{v}_q \times \mathbf{v}_p).
\]  

(1.10)

A pure quaternion \((0, \mathbf{v}_q)\) has zero scalar part and a unit quaternion has unit norm \(|q| = 1\). A rotation can be represented by a unit quaternion as follows. Consider the pure quaternion \( r = (0, \mathbf{r}) \) and the unit quaternion \( q = (q_0, \mathbf{v}_q) \). For \( r' = qr\bar{q} \) we find

\[
r' = qr\bar{q} = (0, [q_0^2 - \mathbf{v}_q \cdot \mathbf{v}_q] \mathbf{r} + 2 \mathbf{r} \cdot \mathbf{v}_q \mathbf{v}_q + 2q_0 \mathbf{v}_q \times \mathbf{r}).
\]  

(1.11)

If we define an angle \( \psi \) and a unit vector \( \mathbf{n} \) by

\[
q_0 = \cos(\psi/2) \quad \text{and} \quad \mathbf{v}_q = (q_1, q_2, q_3)^T = \mathbf{n} \sin(\psi/2),
\]  

(1.12)

then

\[
r' = (0, [1 - \cos \psi] \mathbf{n} \cdot \mathbf{r} + \cos \psi \mathbf{r} + \sin \psi \mathbf{n} \times \mathbf{r}) = (0, M\mathbf{r}),
\]  

(1.13)
where $M$ is the rotation matrix of eq. (1.6). Hence, we found that a unit quaternion represents a rotation. By means of eq. (1.12) we can find the representation of $M$ in terms of $(q_0, q_1, q_2, q_3)$,

$$M = \begin{pmatrix} q_0^2 + q_1^2 - q_2^2 - q_3^2 & 2(q_1 q_2 - q_0 q_3) & 2(q_1 q_3 + q_0 q_2) \\ 2(q_2 q_1 + q_0 q_3) & q_0^2 - q_1^2 + q_2^2 - q_3^2 & 2(q_2 q_3 - q_0 q_1) \\ 2(q_2 q_1 - q_0 q_2) & 2(q_3 q_2 + q_0 q_1) & q_0^2 - q_1^2 - q_2^2 + q_3^2 \end{pmatrix}. \quad (1.14)$$

1.2 Dynamical equations of a rigid body

A rigid body is an idealization of a solid body, for which the distance between any given two points inside the body remains constant in time.

1.2.1 Angular velocity

**Angular velocity from the rotation matrix**

The time dependent motion of a point $\mathbf{r}(t)$ in the rigid body can be composed of a translation of a reference point $\mathbf{r}_0$ and a rotation about that reference point [55] (see fig. 1.2)

$$\mathbf{r}(t) = \mathbf{r}_0(t) + \mathbf{D}^T(t)(\mathbf{r}(0) - \mathbf{r}_0(0)). \quad (1.15)$$

Here, $\mathbf{D}^T$ denotes a time-dependent rotation matrix, which is independent of the choice of the reference point [55]

$$\mathbf{r}(t) = \mathbf{r}_0(t) + \mathbf{D}^T(t)(\mathbf{r}(0) - \mathbf{r}_0(0)) + \mathbf{r}_0(0) - \mathbf{r}_0(0) \quad (1.16)$$

$$= \mathbf{r}_0(t) + \mathbf{D}^T(t)(\mathbf{r}(0) - \mathbf{r}_0(0)). \quad (1.17)$$
1.2 Dynamical equations of a rigid body

We chose to include the transpose in the definition of $D$ (eq. (1.15)) for consistency with ref. [58]. The rotation matrix transforms between an axis system fixed in space, which we denote as the laboratory or space-fixed frame, and one fixed with the body [58]. We associate $D^T$ with a rotation quaternion $q$, i.e., $D^T = M$ from eq. (1.14) or

$$
D = \begin{pmatrix}
q_0^2 + q_1^2 - q_2^2 - q_3^2 & 2(q_1q_2 + q_0q_3) & 2(q_1q_3 - q_0q_2) \\
2(q_2q_1 - q_0q_3) & q_0^2 - q_1^2 + q_2^2 - q_3^2 & 2(q_2q_3 + q_0q_1) \\
2(q_3q_1 + q_0q_2) & 2(q_3q_2 - q_0q_1) & q_0^2 - q_1^2 - q_2^2 + q_3^2 
\end{pmatrix}.
$$

(1.18)

Vectors in the space-fixed frame and body-fixed frame are distinguished by a superscript: $v^s$ is a vector in the laboratory (or space-fixed) frame, while $v^b = Dv^s$ (1.19) is the corresponding vector in the body-fixed frame. For vectors in the laboratory frame, we will frequently omit the superscript. We differentiate eq. (1.15) to find

$$
\dot{r}(t) = \dot{r}_0 + \dot{D}^T(r(0) - r_0(0)) = \dot{r}_0 + \dot{D}^T D(r(t) - r_0(t)).
$$

(1.20)

For the second equality we applied eq. (1.15) and $DD^T = 1$. It turns out that $\dot{D}^T D$ is skew-symmetric, since

$$
0 = \frac{d}{dt} 1 = \frac{d}{dt} (D^T D) = \dot{D}^T D + D^T \dot{D} = \dot{D}^T D + (\dot{D}^T D)^T.
$$

(1.21)

Hence, we can represent the action of $\dot{D}^T D$ by a cross product from the left with some vector $\Omega$ (see eq. (1.3)). We call this vector the angular velocity and obtain

$$
\dot{r}(t) = \dot{r}_0 + \Omega \times (r(t) - r_0(t)).
$$

(1.22)

A shortcut to arrive at this equation for a pure rotation around the origin ($r_0(t) = 0$) is to divide eq. (1.2) by $dt$ and to define $\Omega = n \dot{\psi}$.

**Angular velocity in quaternions**

If the position $r(0)$ at time $t = 0$ is the vector part of the pure quaternion $r(0) = (0, r(0))$ and $r(t) = q(t)r(0)\bar{q}(t) = (0, r(t))$, then the velocity $v$ is the vector part of

$$
\dot{r}(t) = \dot{q}r(0)\bar{q} + qr(0)\dot{q} = \dot{q}\bar{q}r(0)\bar{q} + qr(0)(-\bar{q}\dot{q}q) = [\dot{q}\bar{q}, r(t)].
$$

(1.23)

Here, we exploited that the product rule also holds for the non-commutative quaternion algebra and that $\bar{q}\dot{q} = 1$. Furthermore, we introduced the commutator $[,]$ as $[a, b] = ab - ba$. We find

$$
\dot{q}\bar{q} = (0, q_0\dot{v}_q - \dot{q}_0v_q + v_q \times \dot{v}_q) \equiv (0, \omega/2),
$$

(1.24)
and \( \dot{r}(t) = [\dot{q}, \dot{r}(t)] = (0, \omega \times r) \). Comparison with eq. (1.22) shows that \( \omega \) is the angular velocity \( \Omega \). The linear relation \((0, \Omega) = 2\dot{q}\bar{q}\) can be written in matrix form as

\[
\left( \begin{array}{c} 0 \\ \Omega \end{array} \right) = 2 \left( \begin{array}{cccc} q_0 & q_1 & q_2 & q_3 \\ -q_1 & q_0 & -q_3 & q_2 \\ -q_2 & q_3 & q_0 & -q_1 \\ -q_3 & -q_2 & q_1 & q_0 \end{array} \right) \left( \begin{array}{c} \dot{q}_0 \\ \dot{q}_1 \\ \dot{q}_2 \\ \dot{q}_3 \end{array} \right). \quad (1.25)
\]

We can invert this relation to solve for \( \dot{q} \equiv (\dot{q}_0, \dot{q}_1, \dot{q}_2, \dot{q}_3)^T \) and subsequently substitute \((0, \Omega)^T = (1 \oplus D^T)(0, \Omega^b)^T\) to find the important identity

\[
\dot{q} = \frac{1}{2} Q(q) \left( \begin{array}{c} 0 \\ \Omega^b \end{array} \right), \quad \text{where} \quad Q(q) = \left( \begin{array}{cccc} q_0 & -q_1 & -q_2 & -q_3 \\ q_1 & q_0 & -q_3 & q_2 \\ q_2 & q_3 & q_0 & -q_1 \\ q_3 & -q_2 & q_1 & q_0 \end{array} \right). \quad (1.26)
\]

Here we introduced the notation \( q \equiv (q_0, q_1, q_2, q_3)^T \) which identifies the quaternion components with a vector in \( \mathbb{R}^4 \).

### 1.2.2 Equations of motion

#### Translational dynamics

As a model for a rigid body, consider a collection of point particles \( i = 1, \ldots, N \) with \( |\mathbf{r}_i - \mathbf{r}_j| = \text{const.} \) for all \( i, j \). Newton’s second law for a point particle \( i \) states

\[
m_i \ddot{\mathbf{r}}_i = \mathbf{f}_i, \quad (1.27)
\]

where \( \mathbf{f}_i \) is the force on particle \( i \) and \( m_i \) is its mass. Summing this equation over all particles \( i \), yields the total force on the rigid body \( \mathbf{F} \). In the continuum limit \( \sum_i m_i \to \int dV \rho(\mathbf{r}) = \int dm \) where \( \rho(\mathbf{r}) \) is the mass density and \( dV \) is the volume element, we find

\[
\int dm \, \ddot{\mathbf{r}} = \int dV \, \mathbf{f} = \mathbf{F}. \quad (1.28)
\]

From eq. (1.22), we obtain

\[
\ddot{\mathbf{r}} = \dot{\mathbf{r}}_0 + \dot{\Omega} \times (\mathbf{r} - \mathbf{r}_0) + \Omega \times (\dot{\mathbf{r}} - \dot{\mathbf{r}}_0) \quad (1.29)
\]

\[
= \dot{\mathbf{r}}_0 + \dot{\Omega} \times (\mathbf{r} - \mathbf{r}_0) + \Omega \times \Omega \times (\mathbf{r} - \mathbf{r}_0) \quad (1.30)
\]

If we choose \( \mathbf{r}_0 \) as the center-of-mass \( \mathbf{C} = (1/M) \int dm \, \mathbf{r} \), where \( M = \int dm \) is the total mass of the rigid body, eqs. (1.28) and (1.30) yield

\[
M \ddot{\mathbf{C}} = \mathbf{F}. \quad (1.31)
\]
Rotational dynamics

We apply a cross product with \((r_i - r_0)\) from the left to Newton’s second law (1.27)
\[
(r_i - r_0) \times f_i = (r_i - r_0) \times m_i \ddot{r}_i
\]
\[= m_i \frac{d}{dt} \left( (r_i - r_0) \times \frac{d}{dt}(r_i - r_0) \right) + m_i (r_i - r_0) \times \dot{r}_0 \tag{1.32}
\]
Summation over \(i\), or equivalently integration over the the rigid body’s volume yields
\[
\dot{l} + M(C - r_0) \times \dot{r}_0 = \int dV (r - r_0) \times f \equiv T, \tag{1.34}
\]
where we defined the angular momentum \(l = \int dm \ (r - r_0) \times (\dot{r} - \dot{r}_0)\) and the torque \(T\). If we choose \(r_0 = C\), we find
\[
\dot{l} = T. \tag{1.35}
\]
The angular momentum \(l\) depends on the angular velocity \(\Omega\) via (we continue to choose \(r_0 = C\) in the following)
\[
l = \int dm \ (r - C) \times (\dot{r} - \dot{C}) = \int dm \ (r - C) \times [\Omega \times (r - C)]
\]
\[= - \int dm (r - C) \times [(r - C) \times \Omega] \tag{1.36}
\]
\[= \int dm \ [(r - C)^2 - (r - C)(r - C)^T] \Omega \equiv I \Omega, \tag{1.38}
\]
where we used the identity \(a \times (b \times c) = b(a \cdot c) - c(a \cdot b)\), and defined the moment of inertia tensor of the rigid body \(I\), which depends on time in general. In the body-fixed frame however, \(I^b\) is a constant real symmetric matrix, and hence, it can be brought to diagonal form by means of a rotation. Thus, we can and will always choose the body-fixed frame in such a way that \(I^b\) is diagonal. The rotational equation of motion (1.35) yields
\[
T = \frac{d}{dt} \langle I \Omega \rangle = \frac{d}{dt} (D^T I^b D \Omega) = \dot{D}^T I^b D \Omega + D^T I^b (\dot{D} \Omega + D \dot{\Omega}) \tag{1.39}
\]
\[= \dot{D}^T D D^T I^b D \Omega + D^T I^b D \dot{D}^T (D \Omega + D \dot{\Omega}) \tag{1.40}
\]
\[= \Omega \times [D^T I^b D \Omega] + D^T I^b D \dot{\Omega} = \dot{I} \Omega + \Omega \times (I \Omega). \tag{1.41}
\]
This equation holds in the body-fixed frame as well
\[
T^b = D \frac{d}{dt} (D^T I^b D \Omega) = D D^T I^b \dot{\Omega}^b + D \dot{D}^T I^b \Omega^b \tag{1.42}
\]
\[= I^b \dot{\Omega}^b + D \dot{D}^T D D^T I^b \Omega^b = I^b \dot{\Omega}^b + D C^T D^T I^b \Omega \tag{1.43}
\]
\[= I^b \dot{\Omega}^b + \Omega^b \times (I^b \Omega^b). \tag{1.44}
\]
Writing the equation in Cartesian components yields Euler’s rotation equations
\[
\frac{d\Omega^b}{dt} = I^{-1} \left[ T^b + (I_\beta - I_\gamma)\Omega^b\Omega^\gamma \right],
\] (1.45)
where \((\alpha, \beta, \gamma) = (x, y, z), (y, z, x), \) or \((z, x, y)\). The moment of inertia \(I_\alpha\) for \(\alpha \in \{x, y, z\}\) is the eigenvalue of \(I^b\) to the eigenvector \(e_\alpha\).

**Forces and torques from potentials**

We regard the rigid body as a collection of point particles and assume that the forces are given by a potential \(f_i = -\partial U(r_1, \ldots, r_N)/\partial r_i\). Consider an infinitesimal translation \(\delta r\) of the rigid body. The resulting change in potential energy \(U\) is the negative of the work corresponding to the translation
\[
\delta U = -\sum_i f_i \cdot \delta r_i = -\delta C \cdot \sum_i f_i,
\] (1.46)
where we used that \(\delta r_i\) is equal for all \(i\) by the definition of a rigid body. For the continuum rigid body this means \(\delta U = -\delta C \cdot F\) and hence
\[
F = -\frac{\partial U}{\partial C}.
\] (1.47)

Now consider an infinitesimal rotation such that the displacement of particle \(i\) is \(\delta r_i = \delta \psi n \times r_i\) (see eq. (1.2)). The change of potential energy is
\[
\delta U = -\sum_i f_i \cdot \delta r_i = -\sum_i f_i \cdot [\delta \psi n \times r_i] = -\delta \psi n \cdot \sum_i r_i \times f_i.
\] (1.48)

For the continuum rigid body this means \(\delta U = -\delta \psi n \cdot T\) and hence
\[
T_\alpha = -\frac{\partial U}{\partial \psi_\alpha} \quad \text{for } \alpha \in \{x, y, z\}.
\] (1.49)

For the continuum rigid body the potential energy can typically written as \(U(C, D)\), where \(D\) is the rotation matrix. Hence, it is noteworthy that [59]
\[
\frac{\partial}{\partial \psi_\alpha} D = e_\alpha \times D.
\] (1.50)

**1.3 Numerical integration of rigid body dynamics**

We can construct a numerical integration scheme for a time dependent variable \(x\) by Taylor expansion
\[
x(t + h) = x(t) + h \dot{x}(t) + \frac{h^2}{2} \ddot{x}(t) + O(h^3).
\] (1.51)
1.3 Numerical integration of rigid body dynamics

If we know \( \mathbf{x} \), \( \dot{\mathbf{x}} \) and \( \ddot{\mathbf{x}} \) at time \( t \), we can thus predict \( \mathbf{x}(t+h) \). Denoting the time derivative of \( \mathbf{x} \) by \( \mathbf{y} \), we can find an alternative update for \( \mathbf{x} \)

\[
\mathbf{x}(t+h) = \mathbf{x}(t) + \left[ \mathbf{y}(t) + \frac{h}{2} \ddot{\mathbf{y}}(t) \right] h + O(h^3) \\
= \mathbf{x}(t) + \left[ \mathbf{y}(t) + \frac{1}{2}(\mathbf{y}(t+h) - \mathbf{y}(t)) + O(h^2) \right] h + O(h^3) \\
= \mathbf{x}(t) + \frac{h}{2}(\mathbf{y}(t) + \mathbf{y}(t+h)) + O(h^3)
\] (1.52)

This integration scheme requires that \( \mathbf{y}(t+h) \) is already known. With the above two schemes, we construct the integration algorithm presented in ref. [60]. We apply scheme (1.51) to the position \( \mathbf{C}(t) \) and the orientation quaternion \( \mathbf{q}(t) \)

\[
\mathbf{C}(t+h) = \mathbf{C}(t) + \mathbf{U}(t)h + \frac{h^2}{2M} \mathbf{F}^s(t),
\] (1.53)

\[
\mathbf{q}(t+h) = (1 - \tilde{\lambda})\mathbf{q}(t) + \dot{\mathbf{q}}h + \frac{h^2}{2} \ddot{\mathbf{q}},
\] (1.54)

\[
\tilde{\lambda} = 1 - \mathbf{q}^2 h^2 / 2 - \sqrt{1 - \mathbf{q}^2 h^2} - \mathbf{q} \cdot \dot{\mathbf{q}} h^3 - (\mathbf{q}^2 - \mathbf{q}^4) h^4 / 4.
\] (1.55)

For the quaternion update, we added the term \( -\tilde{\lambda}\mathbf{q}(t) \) with the Lagrangian multiplier \( \tilde{\lambda} \) to guarantee normalization, i.e., \( \mathbf{q}(t+h) = 1 \). This normalization condition yields eq. (1.55), which can be easily shown using the relations \( \mathbf{q}^2 = 1 \), \( \mathbf{q} \cdot \dot{\mathbf{q}} = 0 \) and \( \ddot{\mathbf{q}} \cdot \mathbf{q} = -\dot{\mathbf{q}}^2 \). In eqs. (1.54) and (1.55), \( \dot{\mathbf{q}}(t) \) is determined according to eq. (1.26), and an expression for \( \ddot{\mathbf{q}} \) is found by differentiation of eq. (1.26)

\[
\dot{\mathbf{q}} = \frac{1}{2} Q(\dot{\mathbf{q}}) \left( \begin{array}{c} 0 \\ \Omega^b \end{array} \right) + \frac{1}{2} Q(\mathbf{q}) \left( \begin{array}{c} 0 \\ \Omega^b \end{array} \right).
\] (1.56)

The derivative of the body-fixed angular velocity \( \dot{\Omega}^b \) is determined by Euler’s rotation equations (1.45).

Once \( \mathbf{q} \) and \( \mathbf{C} \) have been updated, we apply the integration scheme (1.52) for the velocity \( \mathbf{U} \) and the angular momentum \( \mathbf{l} \)

\[
\mathbf{U}(t+h) = \mathbf{U}(t) + \frac{h}{2M} \left[ \mathbf{F}^s(t) + \mathbf{F}^s(t+h) \right],
\] (1.57)

\[
\mathbf{l}^s(t+h) = \mathbf{l}^s(t) + \frac{h}{2} \left[ \mathbf{T}^s(t) + \mathbf{T}^s(t+h) \right].
\] (1.58)

Here, we assume that the force \( \mathbf{F} \) and the torque \( \mathbf{T} \) only depend on the position \( \mathbf{C} \) and orientation \( \mathbf{q} \), and not on \( \mathbf{l} \) or \( \mathbf{U} \). Once the angular momentum has been updated, the angular velocity is found by \( \dot{\Omega} = \mathbf{D}^T (\mathbf{I}^b)^{-1} \mathbf{D} \mathbf{l}^s \). The rotation matrix \( \mathbf{D}(t) \) is given by eq. (1.18).
Chapter 2
Hydrodynamics

Consider a gas or fluid, characterized by its mass density $\rho(r, t)$, pressure $p(r, t)$, and velocity $v(r, t)$. Each quantity is given at a specific position $r$ and time $t$. In this chapter, we discuss the partial differential equations governing the time evolution of those quantities, which is termed hydrodynamics. Furthermore, we show how to describe the dynamics of an immersed object like a passive colloid or a swimmer in the framework of hydrodynamics.

2.1 Stress tensor of a Newtonian fluid

The stress $\sigma$ in a fluid measures the infinitesimal internal force $dF$ exerted on an infinitesimal surface or fluid layer $dA$. This internal force is due to intermolecular interactions, which we will not resolve, since we are working on continuum length and time scales [61]. Denoting the surface normal vector by $n$ we may write

$$dF_\alpha = \sum_\beta \sigma_{\alpha\beta} n_\beta \, dA,$$

where $\sigma_{\alpha\beta}$ is called the stress tensor. This linear relation is justified by the fact that it is invariant under rotation of the reference frame [61]. The internal force $\mathbf{F}$ and torque $\mathbf{T}$ on a fluid volume $V$ with boundary $\partial V$ is then given by

$$\mathbf{F} = \int_{\partial V} dA \, \mathbf{\sigma} n,$$

$$\mathbf{T} = \int_{\partial V} dA \, \mathbf{r} \times \mathbf{\sigma} n.$$

The total stress tensor $\sigma_{\alpha\beta}$ for a Newtonian fluid is given by the sum of pressure and viscous drag stresses

$$\sigma_{\alpha\beta} = \sigma_{\alpha\beta}^p + \sigma_{\alpha\beta}^v.$$

Stresses due to pressure are always parallel to the surface normal $n$ [61], i.e.,

$$\sigma_{\alpha\beta}^p = -p \delta_{\alpha\beta}.$$
Chapter 2 Hydrodynamics

Figure 2.1: Origin of viscous drag in fluid flow. The slow fluid layer on the right is pulled along by the fast left layer. The corresponding drag force is approximately linear in the velocity gradient: \( \sigma_{xz} = \eta \frac{\partial}{\partial x} v_z \).

Viscous drag forces arise between fluid layers moving with different velocities (see fig. 2.1). Hence, the corresponding stress will depend on the velocity gradient \( \partial v_\alpha / \partial r_\beta \). In a first order expansion, we assume that only linear combinations of first order derivatives occur in the stress tensor [62]

\[
\sigma_{\alpha\beta}^v = \sum_{\alpha'\beta'} \eta_{\alpha\beta\alpha'\beta'} \frac{\partial v_{\alpha'}}{\partial r_{\beta'}}. \tag{2.6}
\]

For an isotropic fluid we may assume that the tensor \( \eta_{\alpha\beta\alpha'\beta'} \) is isotropic, i.e., that it has the same components in all rotated coordinate systems. The most general isotropic Cartesian tensors of fourth order is [61]

\[
\eta_{\alpha\beta\alpha'\beta'} = \eta_1 \delta_{\alpha\beta} \delta_{\alpha'\beta'} + \eta_2 \delta_{\alpha\alpha'} \delta_{\beta\beta'} + \eta_3 \delta_{\alpha\beta} \delta_{\alpha'\beta'}. \tag{2.7}
\]

This corresponds to the viscous stress tensor

\[
\sigma_{\alpha\beta}^v = \eta_1 \frac{\partial v_\beta}{\partial r_\alpha} + \eta_2 \frac{\partial v_\alpha}{\partial r_\beta} + \eta_3 \delta_{\alpha\beta} \sum_{\gamma} \frac{\partial v_\gamma}{\partial r_\gamma}. \tag{2.8}
\]

No drag forces should occur when the fluid is rotating uniformly, since this corresponds to rotation of the vessel containing the fluid [63]. Calculating the stress tensor for the rotating flow field \( v_\gamma = \sum_{\mu\lambda} \varepsilon_{\gamma\mu\lambda} \Omega_\mu r_\lambda \), where \( \varepsilon_{\gamma\mu\lambda} \) is the third order permutation tensor and \( \Omega \) the angular velocity, we find \( \sigma_{\alpha\beta} = (\eta_1 - \eta_2) \sum_{\mu} \varepsilon_{\beta\mu\alpha} \Omega_\mu \) and hence \( \eta_1 = \eta_2 \equiv \eta \). The parameter \( \eta \) is known as shear viscosity and measures the resistance of the fluid against shearing. Finally, we can decompose the stress tensor into a term with vanishing trace and a term proportional to the identity tensor

\[
\sigma_{\alpha\beta}^v = \eta \left( \frac{\partial v_\beta}{\partial r_\alpha} + \frac{\partial v_\alpha}{\partial r_\beta} - \frac{2}{3} \delta_{\alpha\beta} \sum_{\gamma} \frac{\partial v_\gamma}{\partial r_\gamma} \right) + \eta V \delta_{\alpha\beta} \sum_{\gamma} \frac{\partial v_\gamma}{\partial r_\gamma}, \tag{2.9}
\]
where we defined the bulk viscosity
\[ \eta^V = \eta_3 + (\eta_1 + \eta_2)/3. \]  
(2.10)
The stress associated with the bulk viscosity can be imagined as a kind of pressure, as it is proportional to \( \delta_{\alpha\beta} \), like \( \sigma^p_{\alpha\beta} \). The bulk stress appears, when the fluid is being compressed, but in contrast to the hydrostatic pressure \( p \), it is proportional to the rate of change\(^a\) rather than the extent of compression. The separation of a traceless part from the stress tensor associated with shear stresses can be motivated by the example \( v(r, t = 0) = -Ar, A > 0 \). According to eq. (2.9) the induced viscous stress \( \sigma_{xx} \) is only proportional to the bulk, but not to the shear viscosity, as we would expect it. In summary
\[ \sigma_{\alpha\beta} = -p\delta_{\alpha\beta} + \eta \left( \frac{\partial v_\beta}{\partial r_\alpha} + \frac{\partial v_\alpha}{\partial r_\beta} - \frac{2}{3} \delta_{\alpha\beta} \sum_\gamma \frac{\partial v_\gamma}{\partial r_\gamma} \right) + \eta^V \delta_{\alpha\beta} \sum_\gamma \frac{\partial v_\gamma}{\partial r_\gamma}. \]  
(2.11)

### 2.2 Conservation laws and Navier-Stokes equations

#### 2.2.1 Material volume and material derivative

The volume \( V(t) \) occupied by a specific collection of fluid particles is called a material volume [65]. Such a volume moves and deforms within a fluid flow so that it always contains the same fluid particles. A property \( g(r, t) \) of a material volume changes over time \( \Delta t \) by
\[ g + \Delta g = g(r + v\Delta t, t + \Delta t) = g(r, t) + \Delta t \left( \frac{\partial g}{\partial t} + v \cdot \nabla g \right) + O(\Delta t^2) \]  
(2.12)
In this equation, \( g \) can be subtracted from both sides, to define the material derivative
\[ \frac{Dg}{Dt} = \frac{\partial g}{\partial t} + v \cdot \nabla g. \]  
(2.13)

#### 2.2.2 Mass and momentum conservation

An example for \( g \) would be the mass density \( \rho \). The mass of a material volume \( M = \int_{V(t)} \rho dV \) does not change over time, since a material volume does not lose or gain fluid particles by definition. In order to formulate mass conservation, i.e., \( dM/dt = 0 \), we have to find the time derivative of an integral \( \int_{V(t)} g dV \), with time dependent

\(^a\)Note that \( \nabla \cdot v \) can be interpreted as the relative rate of change of volume of the fluid due to the flow [64].
integration range. This can be done by the three dimensional generalization of the
Leibniz integral rule, known as Reynolds transport theorem [65]
\[
\frac{d}{dt} \int_{V(t)} g(\mathbf{r}, t) \, dV = \int_{\partial V(t)} \frac{\partial g(\mathbf{r}, t)}{\partial t} \, dV + \int_{\partial V(t)} g(\mathbf{r}, t) \mathbf{v}(\mathbf{r}, t) \cdot \mathbf{n} \, dA.
\] (2.14)
The velocity appearing in the surface integral is the velocity of the surface. But
since \( V(t) \) is a material volume, the velocity of its surface is just the fluid velocity \( \mathbf{v}(\mathbf{r}, t) \) [65]. Applying eq. (2.14) to the mass density \( \rho \), and applying Gauss’ theorem, we find [65]
\[
0 = \frac{d}{dt} \int_{V(t)} \rho \, dV = \int_{\partial V(t)} \left\{ \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) \right\} \, dV
\] (2.15)
Since the volume \( V(t) \) is arbitrary, the integrand must be zero, and we obtain the
continuity equation
\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0.
\] (2.16)
In an incompressible flow, the material derivative of the mass density vanishes. In
this case the continuity equation simplifies to \( \nabla \cdot \mathbf{v} = 0 \). Most liquids, such as water
can be considered incompressible [65].

Combining Reynolds transport theorem (eq. (2.14)) and the continuity equation
(2.16), we obtain the useful identity
\[
\frac{d}{dt} \int_{V(t)} \rho \mathbf{v} \, dV = \int_{\partial V(t)} \rho \frac{D g}{Dt} \, dA,
\] (2.17)
which enables us to derive the Navier-Stokes equations from momentum conserva-
tion. Momentum conservation means that the change in momentum over time is
given by the applied force. The momentum of a material volume changes due to the
applied stresses on its surface and volume forces \( \mathbf{f} \) such as gravity
\[
\frac{d}{dt} \int_{V(t)} \rho \mathbf{v} \, dV = \int_{\partial V(t)} \sigma \mathbf{n} \, dA + \int_{V(t)} \mathbf{f} \, dV.
\] (2.18)
We apply eq. (2.17) on the left hand side, and Gauss’ theorem on the right hand
side to find
\[
\int_{V(t)} \rho \frac{D \mathbf{v}}{Dt} \, dV = \int_{V(t)} \left( \nabla \cdot \sigma + \mathbf{f} \right) \, dV.
\] (2.19)
Again, the volume is arbitrary, and we obtain the Navier-Stokes equations
\[
\rho \frac{D \mathbf{v}}{Dt} = \nabla \cdot \sigma + \mathbf{f}.
\] (2.20)
2.2 Conservation laws and Navier-Stokes equations

With the stress tensor of eq. (2.9), the Navier-Stokes equations reads

$$\rho \left( \frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) = -\nabla p + \eta \Delta \mathbf{v} + \left( \frac{\eta}{3} \right) \nabla(\nabla \cdot \mathbf{v}) + \mathbf{f}. \quad (2.21)$$

For an isothermal fluid, eqs. (2.16) and (2.21), together with an equation of state connecting $p$ and $\rho$, form a closed set of equations for $p(\mathbf{r}, t)$, $\rho(\mathbf{r}, t)$, and $\mathbf{v}(\mathbf{r}, t)$. Typically, the equation of state is expanded up to first order to $p = \rho c^2$, with the isothermal velocity of sound $c$.

For an isoenergetic fluid, a temperature field $T(\mathbf{r}, t)$ and thermal energy field $e(\mathbf{r}, t)$ are introduced, for which an additional thermodynamic equation and a partial differential equation expressing energy conservation can be derived [65]. For a compressible fluid, these equations are coupled to eqs. (2.16) and (2.21), while for an incompressible fluid, the Navier-Stokes equations (2.21) are decoupled from the evolution of $e(\mathbf{r}, t)$ and $T(\mathbf{r}, t)$ if one assumes $\rho$ to be constant (sufficient for incompressibility).

### 2.2.3 Angular momentum conservation

Angular momentum conservation means that the change in angular momentum is equal to the applied torque\(^b)\)

$$\frac{d}{dt} \int_{V(t)} \rho \sum_{\beta \gamma} \epsilon_{\alpha \beta \gamma} r_\beta v_\gamma \, dV = \int_{\partial V(t)} \sum_{\beta \gamma} \epsilon_{\alpha \beta \gamma} r_\beta \sigma_{\gamma \zeta} n_\zeta \, dA \quad (2.22)$$

We apply eq. (2.17) on the left hand side, and Gauss’ theorem on the right hand side to find

$$\sum_{\beta \gamma} \epsilon_{\alpha \beta \gamma} \int_{V(t)} \rho \frac{D}{Dt}(r_\beta v_\gamma) \, dV = \sum_{\beta \gamma} \epsilon_{\alpha \beta \gamma} \int_{V(t)} \left\{ r_\beta \frac{\partial}{\partial r_\zeta} \sigma_{\gamma \zeta} + \delta_{\beta \zeta} \sigma_{\gamma \gamma} \right\} dV \quad (2.23)$$

By definition (2.13), it is easy to see that $D(r_\beta v_\gamma)/Dt = 2v_\beta v_\gamma + r_\beta Dv_\gamma/Dt$ and hence

$$\sum_{\beta \gamma} \epsilon_{\alpha \beta \gamma} \int_{V(t)} r_\beta \left( \rho \frac{Dv_\gamma}{Dt} - \frac{\partial}{\partial r_\zeta} \sigma_{\gamma \zeta} \right) \, dV = \int_{V(t)} \sum_{\beta \gamma} \epsilon_{\alpha \beta \gamma} \sigma_{\gamma \beta} \, dV. \quad (2.24)$$

The left hand side vanishes due to the Navier-Stokes equations. Hence, angular momentum conservation is equivalent to a symmetric stress tensor, which means, $\sum_{\beta \gamma} \epsilon_{\alpha \beta \gamma} \sigma_{\gamma \beta} = 0$ or $\sigma_{\gamma \beta} = \sigma_{\beta \gamma}$.

\(^b)\) We neglect body forces and the resulting torques for simplicity. It is easy to show that this does not modify the results of the following considerations.
2.2.4 Boundary conditions

The hydrodynamic equations have to be supplemented by boundary conditions. Usually a vanishing flow field at infinity or periodic boundary conditions are adopted. For the contact between the fluid and an embedded object, the slip or no-slip boundary conditions are commonly employed. We denote the surface of the embedded object by $\partial V$ and the velocity of that surface as $v_S(r)$. For a rigid body translating with velocity $U$ and rotating with angular velocity $\Omega$ around $r_0$ the surface velocity is

\[ v_S(r) = U + \Omega \times (r - r_0). \]  
(2.25)

The no-slip boundary condition states that the fluid sticks to the object

\[ v(r) = v_S(r) \quad \forall r \in \partial V. \]  
(2.26)

This is well established for macroscopic embedded objects and has been confirmed by various experiments [66]. The slip boundary condition can be expressed as

\[ v \cdot n = v_S(r) \cdot n \quad \forall r \in \partial V, \]  
(2.27)

\[ t \cdot \sigma n = 0 \quad \forall r \in \partial V, \]  
(2.28)

where $n$ is the surface normal vector of $\partial V$ at position $r$, and $t$ is a vector tangential to $\partial V$ at $r$. Equation (2.27) expresses that no fluid can enter the embedded object, while eq. (2.28) states that no tangential stresses are exerted on the object. Equation (2.28) can be generalized to yield the partial-slip boundary condition [67]

\[ t \cdot \sigma n = (\eta/\lambda)t \cdot (v(r) - v_S(r)) \quad \forall r \in \partial V; \]  
(2.29)

where $\lambda$ is the slip length. Slip boundary conditions are recovered for $\lambda = \infty$ and no-slip boundary conditions for $\lambda = 0$.

2.3 Stokes equations

2.3.1 Reynolds number and the creeping flow limit

In order to analyze the contributions of the various terms in the Navier-Stokes equations, we rescale according to [63]

\[ v' = v/V_0, \quad r' = r/L_0, \quad t' = t/T_0, \]  
(2.30)

where $V_0$ is a typical velocity, $L_0$ is a typical length and $T_0$ is the timescale of the problem at hand. In those new variables and after multiplying by $L_0^2/((\eta V_0))$, the Navier-Stokes equations read

\[ \text{Re}_T \frac{\partial v'}{\partial t'} + \text{Re} v' \cdot (\nabla' v') = -\nabla' p' + \Delta' v' + f', \]  
(2.31)
where we defined the dimensionless pressure \( p' = L_0 \eta^{-1} V_0^{-1} p \), the dimensionless force \( f' = L_0^2 \eta^{-1} V_0^{-1} f \), and \( \nabla' = \partial / \partial r' \). Furthermore, we introduced two dimensionless numbers, the Reynolds number \( \text{Re} \) and the oscillatory Reynolds number \( \text{Re}_T \) \[68\]

\[
\text{Re} = \frac{\rho V_0 L_0}{\eta},
\]
\[
\text{Re}_T = \frac{\rho L_0^2}{\eta T_0}.
\]

For \( \text{Re} \ll 1 \) we can neglect the nonlinear advective term in eq. (2.31), yielding the linearized Navier-Stokes equations

\[
\rho \frac{\partial v}{\partial t} = -\nabla p + \eta \Delta v + f.
\]

If additionally \( \text{Re}_T \ll 1 \), we can assume an instantaneous relaxation of hydrodynamics on our timescale of interest, yielding the Stokes equations

\[
0 = -\nabla p + \eta \Delta v + f,
\]
\[
0 = \nabla \cdot v.
\]

Here, we additionally assumed incompressibility, which is justified in most applications. The Stokes equations are also known as creeping flow equations \[63\]. They are time-independent and linear. The latter allows for superposition of solutions, which greatly simplifies the theory.

### 2.3.2 Singularities of Stokes flow

The Stokes equations (2.35) and (2.36) for an unbounded fluid can be solved via Fourier transform \[69\], yielding the flow field and pressure

\[
v(r) = \int d^3 r' \ G(r - r') f(r'),
\]
\[
p(r) = \int d^3 r' \ \frac{1}{4\pi |r - r'|^3} \cdot f(r'),
\]

with the Green’s function

\[
G(r) = \frac{1}{8\pi \eta r} \left( 1 + \frac{rr^T}{r^2} \right),
\]

which is known as Oseen tensor \[69\]. Hence, the solution for a point force \( F = Fe \) at position \( r_0 \), i.e., \( f(r) = \delta(r - r_0)eF \), is given by

\[
v(r) = \frac{F}{8\pi \eta |r - r_0|} \left( e + \frac{r - r_0}{|r - r_0|^2} \cdot e \right) \equiv \frac{F}{8\pi \eta} v^F(r - r_0; e),
\]
\[
p(r) = \frac{F}{4\pi |r - r_0|^3} \cdot eF.
\]
where we defined the Stokeslet (or force singularity) \( \mathbf{v}^F(\mathbf{r} - \mathbf{r}_0; \mathbf{e}) \) in direction of the unit vector \( \mathbf{e} \). The Stokeslet is a singularity solution of the Stokes equations. This means that eqs. (2.40) and (2.41) are a solution of the homogeneous Stokes equations 
\[
\eta \Delta \mathbf{v} - \nabla p = 0, \quad \nabla \cdot \mathbf{v} = 0,
\]
everywhere except for \( \mathbf{r} = \mathbf{r}_0 \), where they result in a delta singularity. Consequently, a derivative of the solution eqs. (2.40) and (2.41) also solves the homogeneous Stokes equations everywhere, except for \( \mathbf{r} = \mathbf{r}_0 \), where it results in a derivative of the delta function. Hence, derivatives of the Stokeslet generate higher order singularities. The force dipole \( \mathbf{v}^{FD} \) and force quadrupole \( \mathbf{v}^{FQ} \) are given by [7]
\[
\mathbf{v}^{FD}(\mathbf{r} - \mathbf{r}_0; \mathbf{e}_2, \mathbf{e}_1) = \mathbf{e}_2 \cdot \frac{\partial}{\partial \mathbf{r}_0} \mathbf{v}^F(\mathbf{r} - \mathbf{r}_0; \mathbf{e}_1), \tag{2.42}
\]
\[
\mathbf{v}^{FQ}(\mathbf{r} - \mathbf{r}_0; \mathbf{e}_3, \mathbf{e}_2, \mathbf{e}_1) = \mathbf{e}_3 \cdot \frac{\partial}{\partial \mathbf{r}_0} \mathbf{v}^{FD}(\mathbf{r} - \mathbf{r}_0; \mathbf{e}_2, \mathbf{e}_1). \tag{2.43}
\]
This hierarchy of singularities can be continued ad infinitum. The Stokeslet, force dipole, and force quadrupole decay proportional to \( 1/\mathbf{r} \), \( 1/\mathbf{r}^2 \), and \( 1/\mathbf{r}^3 \) for \( \mathbf{r} \to \infty \), respectively. All these singularities are useful for solving the homogeneous Stokes equations on a domain which excludes \( \mathbf{r}_0 \), for example when an immersed object like a colloid or a swimmer is located at \( \mathbf{r}_0 \).

Additionally to the Stokeslet, we can generate solutions from another singular solution of the Stokes equations, the source singularity, which is associated with the Laplace equation \( (p = \text{const.}, \mathbf{f} = 0 \Rightarrow \Delta \mathbf{v} = 0) \). The source and source dipole singularities are [7]
\[
\mathbf{v}^S(\mathbf{r} - \mathbf{r}_0) = \frac{\mathbf{r} - \mathbf{r}_0}{|\mathbf{r} - \mathbf{r}_0|^3}, \tag{2.44}
\]
\[
\mathbf{v}^{SD}(\mathbf{r} - \mathbf{r}_0; \mathbf{e}) = \mathbf{e} \cdot \frac{\partial}{\partial \mathbf{r}_0} \mathbf{v}^S(\mathbf{r} - \mathbf{r}_0), \tag{2.45}
\]
and decay as \( 1/\mathbf{r}^2 \) and \( 1/\mathbf{r}^3 \) for \( \mathbf{r} \to \infty \), respectively. Note that the source singularities are related to the singularities derived from a Stokeslet by [7]
\[
\mathbf{v}^{SD}(\mathbf{r} - \mathbf{r}_0; \mathbf{e}) = -\frac{1}{2} \left( \frac{\partial}{\partial \mathbf{r}_0} \right)^2 \mathbf{v}^F(\mathbf{r} - \mathbf{r}_0; \mathbf{e}). \tag{2.46}
\]
An important combination of the above singularities is the so called rotlet
\[
\mathbf{v}^R(\mathbf{r} - \mathbf{r}_0; \mathbf{e}) = \frac{1}{2} \left( \mathbf{v}^{FD}(\mathbf{r} - \mathbf{r}_0; \mathbf{e}^\perp, \mathbf{e}^{\perp\perp}) - \mathbf{v}^{FD}(\mathbf{r} - \mathbf{r}_0; \mathbf{e}^{\perp\perp}, \mathbf{e}^\perp) \right) \tag{2.47}
\]
\[
= \frac{\mathbf{e} \times (\mathbf{r} - \mathbf{r}_0)}{|\mathbf{r} - \mathbf{r}_0|^3}, \tag{2.48}
\]
where \( (\mathbf{e}, \mathbf{e}^\perp, \mathbf{e}^{\perp\perp}) \) form an orthonormal basis and \( \mathbf{e} \times \mathbf{e}^\perp = \mathbf{e}^{\perp\perp} \). While the Stokeslet represents a point force on the fluid, the rotlet can be interpreted as a point torque.
The rotlet dipole is constructed as
\[ v^{RD}(r - r_0; e_2, e_1) = e_2 \cdot \frac{\partial}{\partial r_0} v^R(r - r_0; e_1) \] (2.49)
and decays proportional to \(1/r^3\) for \(r \to \infty\).

## 2.3 General theorems for Stokes flow

### Reciprocal theorem

A very important theorem for Stokes flow is the reciprocal theorem, which states that for two solutions of the Stokes equations \(v_1\) and \(v_2\), with corresponding stress tensors \(\sigma_1\) and \(\sigma_2\),
\[ \int_{\partial V} dA \, v_1 \cdot \sigma_2 n = \int_{\partial V} dA \, v_2 \cdot \sigma_1 n. \] (2.50)
Here \(\partial V\) is the bounding surface of an arbitrary volume \(V\) and \(n\) its normal vector [70]. Although quite abstract at first glance, the reciprocal theorem has many applications, one of which we will explore in the context of microswimmers in section 2.5.

### Faxén’s Law

Faxén’s law determines the force \(F\) and torque \(T\) on a spherical colloid with no-slip boundary condition in the external flow \(v^*(r)\) [70]
\[ F = 6\pi \eta R \left( 1 + \frac{R^2}{6} \Delta \right) v^*(r) \big|_{r=0} - 6\pi \eta RU, \] (2.51)
\[ T = 8\pi \eta R^3 \left( \frac{1}{2} [\nabla \times v^*(r)] \big|_{r=0} - \Omega \right), \] (2.52)
where \(U\) and \(\Omega\) are the colloid’s velocity and angular velocity, respectively. Faxén’s law has been generalized to colloids with ellipsoidal shapes [71].

## 2.3.4 Axisymmetrical flow: Stokes stream function

We consider a flow past a body of revolution. For such a body, the surface is described by an equation \(f(\bar{r}, z) = 0\) in cylindrical coordinates \((\bar{r}, \varphi, z)\). We assume the flow to be axisymmetric, i.e., \(\partial v/\partial \varphi = 0\) and \(e_\varphi \cdot v = 0\).

Consider an arbitrary point \((\bar{r}, z)\) in the meridian plane and an arbitrary curve \(C_0\) connecting \((\bar{r}, z)\) with a point on the z-axis \((0, z_0)\) (solid line in fig. 2.2). Revolution of this curve around the z-axis defines a surface \(S_0\). We define the instantaneous flow rate through this surface as \(Q = \int_{S_0} dA \, v \cdot n\). The sign of \(Q\) is chosen such that
it is positive, if the net flow through the surface is in negative $z$-direction. Next, consider an arbitrary curve $C_1$ connecting $(\bar{r}, z)$ with another arbitrary point $(0, z_1)$ on the $z$-axis (dashed line in fig. 2.2). The corresponding surface of revolution is denoted as $S_1$. We can show that the flow rate through $S_1$ equals the flow rate through $S_0$

$$\int_{S_0} dA \, \mathbf{v} \cdot \mathbf{n} - \int_{S_1} dA \, \mathbf{v} \cdot \mathbf{n} = \int_V \nabla \cdot \mathbf{v} = 0.$$  (2.53)

Here, $V$ is the volume enclosed by $S_0$ and $S_1$. Moreover, we used Gauss theorem for the first equality, and incompressibility for the second. Hence, the value of $Q$ is uniquely determined by the position $(\bar{r}, z)$. In particular, it is independent of the curve connecting $(\bar{r}, z)$ with the $z$-axis. Consequently, we can define the stream function $\Psi(\bar{r}, z)$ as

$$\Psi = \Psi(\bar{r}, z) = \frac{Q}{2\pi}. \quad (2.54)$$

$\Psi$ can be defined for any coordinate system that contains the azimuthal angle $\varphi$, defined in a plane perpendicular to the body’s axis of revolution. For example, substituting $\bar{r} = r \sin \theta$, $z = r \cos \theta$ into eq. (2.54), yields the stream function in spherical coordinates.

It can be shown [70] that the velocity field can be constructed from the stream function $\Psi$ via

$$\mathbf{v} = -\text{curl} \left( \frac{1}{h_\varphi} \Psi \mathbf{e}_\varphi \right). \quad (2.55)$$

Here, $h_\varphi$ is the Lamé metric coefficient $h_\varphi = |\partial \mathbf{r} / \partial \varphi|^{-1}$. In cylindrical coordinates, eq. (2.55) yields

$$v_\varphi = \frac{1}{\bar{r}} \frac{\partial \Psi}{\partial z}, \quad v_z = -\frac{1}{\bar{r}} \frac{\partial \Psi}{\partial \bar{r}}. \quad (2.56)$$

Figure 2.2: Sketch for the definition of the Stokes stream function.
In general, if \((q_1, q_2, \varphi)\) form a right-handed curvilinear coordinate system, the velocity components are given by [70]

\[
v_1 = -\frac{1}{h_2 h_\varphi} \frac{\partial \Psi}{\partial q_2}, \quad v_2 = \frac{1}{h_1 h_\varphi} \frac{\partial \Psi}{\partial q_1},
\]

(2.57)

where \(h_i\) is the Lamé metric coefficient corresponding to \(q_i\). The dynamical equation satisfied by the stream function is [70]

\[
E^4 \Psi = 0.
\]

(2.58)

Here, \(E^4 = E^2 \circ E^2\) and the differential operator \(E^2\) is given by [70]

\[
E^2 = \frac{\tilde{r}}{h_1 h_2} \left( \frac{\partial}{\partial q_1} \left( \frac{h_2}{h_1} \frac{1}{\tilde{r}} \frac{\partial}{\partial q_1} \right) \right) + \frac{\partial}{\partial q_2} \left( \frac{h_1}{h_2} \frac{1}{\tilde{r}} \frac{\partial}{\partial q_2} \right),
\]

(2.59)

where \(\tilde{r} = \tilde{r}(q_1, q_2) = h_\varphi\). Note that eq. (2.58) is a partial differential equation for a scalar function dependent on two variables. This is a major simplification of the Stokes equations, for which the three dimensional velocity \(\mathbf{v}\) and the pressure \(p\) depend on the three dimensional position \(\mathbf{r}\).

### 2.4 Drag on a colloid

**Instantaneous drag** Consider a rigid colloid, e.g., an ellipsoid, translating with velocity \(\mathbf{U}\) and angular velocity \(\Omega\) in an unbounded fluid described by the Stokes equations. Due to the interaction with the fluid, it experiences a force \(\mathbf{F} = \int_{\partial V} dA \mathbf{\sigma n}\) and torque \(\mathbf{T} = \int_{\partial V} dA \mathbf{r} \times \mathbf{\sigma n}\) (see eqs. (2.2) and (2.3)). Due to linearity of the Stokes equations we can expect the linear response

\[
\begin{pmatrix}
\mathbf{F} \\
\mathbf{T}
\end{pmatrix} = -\begin{pmatrix}
\gamma & \mathbf{c}_{TR} \\
\mathbf{c}_{RT} & \xi
\end{pmatrix} \begin{pmatrix}
\mathbf{U} \\
\Omega
\end{pmatrix},
\]

(2.60)

where the friction matrices \(\gamma, \xi, \mathbf{c}_{TR},\) and \(\mathbf{c}_{RT}\) need to be determined. By application of the reciprocal theorem, it can be shown that \(\mathbf{c}_{TR} = \mathbf{c}_{RT}^T\). Throughout this thesis, we will restrict ourselves to bodies of revolution, for which a symmetry argument [70] yields that the rotation-translation coupling is zero, i.e., \(\mathbf{c}_{TR} = \mathbf{c}_{RT} = 0\). For the symmetry argument, the assumption of an unbounded fluid is highly relevant. If the fluid is bounded or other objects are immersed in the fluid, the rotation-translation coupling is non-zero in general. For example, a sphere sedimenting next to a planar wall will experience a torque and start rotating [70]. However, for a single body of revolution in an unbounded fluid

\[
\mathbf{F} = -\gamma \mathbf{U} \quad \text{and} \quad \mathbf{T} = -\xi \Omega.
\]

(2.61)
Expressed in the body-fixed frame, which is spanned by the axes of symmetry of the body, $\gamma$ and $\xi$ are diagonal matrices [70].

Let the axes of the body-fixed frame be denoted by $x, y, z$. We can find $\gamma_\alpha \equiv \gamma_{\alpha\alpha}$ for $\alpha \in \{x, y, z\}$ by solving the Stokes equations with the no-slip boundary condition $v(r \in \partial V) = U e_\alpha$, where $\partial V$ is the surface of the colloid and $e_\alpha$ is the unit vector in $\alpha$-direction. For each $\alpha$ this will yield a velocity and pressure field and the corresponding force can be calculated according to $F = \int_{\partial V} dA (\sigma n)$, where the stress tensor is a function of the velocity and pressure field (see eq. (2.11)).

Comparison with eq. (2.61) finally yields $\gamma_\alpha$. For $\xi_\alpha \equiv \xi_{\alpha\alpha}$ we proceed analogously with the no-slip boundary condition $v(r \in \partial V) = \Omega e_\alpha \times r$ and determine the torque $T = \int_{\partial V} dA r \times (\sigma n)$.

In our applications we will regard spheroids, i.e., ellipsoids of revolution (see section 9.2.1), for which $\gamma_x = \gamma_y = \gamma^\perp$, $\gamma_z = \gamma^\parallel$ and $\xi_x = \xi_y = \xi^\perp$, $\xi_z = \xi^\parallel$, where the spheroid’s major axis is along the $z$-direction, and

$$\gamma^\parallel = 6\pi \eta b_z \frac{8}{3} e^3 \left( -2e + (1 + e^2) \log \left( \frac{1 + e}{1 - e} \right) \right)^{-1},$$

$$\gamma^\perp = 6\pi \eta b_z \frac{16}{3} e^3 \left( 2e + (3e^2 - 1) \log \left( \frac{1 + e}{1 - e} \right) \right)^{-1},$$

$$\xi^\parallel = 8\pi \eta b_z^3 \frac{4}{3} e^3 (1 - e^2) \left( 2e - (1 - e^2) \log \left( \frac{1 + e}{1 - e} \right) \right)^{-1},$$

$$\xi^\perp = 8\pi \eta b_z^3 \frac{4}{3} e^3 (2 - e^2) \left( -2e + (1 + e^2) \log \left( \frac{1 + e}{1 - e} \right) \right)^{-1}.$$  

(2.62) \hspace{1cm} (2.63) \hspace{1cm} (2.64) \hspace{1cm} (2.65)

Here, $e = \sqrt{b_z^2 - b_x^2} / b_z$ is the spheroid’s eccentricity and $b_z$ and $b_x$ are the spheroid’s semi-major and semi-minor axis, respectively. For spheres, the simple results

$$\gamma_{\alpha\beta} = 6\pi \eta R \delta_{\alpha\beta},$$

$$\xi_{\alpha\beta} = 8\pi \eta R^2 \delta_{\alpha\beta},$$

(2.66) \hspace{1cm} (2.67)

are obtained, where $R$ is the radius.

**Retarded drag** For the linearized Navier-Stokes equations (see eq. (2.34)), a very similar argument to the one above holds. However, force and torque do not follow velocity and angular velocity instantaneously and eq. (2.61) becomes

$$F(t) = - \int_{-\infty}^t dt' \gamma(t - t') U(t') \quad \text{and} \quad T(t) = - \int_{-\infty}^t dt' \xi(t - t') \Omega(t').$$

(2.68)

Typically, the Fourier transformed equations are regarded for which the above convolution corresponds to a simple product, i.e.,

$$\hat{F}(\omega) = -\hat{\gamma}(\omega) \hat{U}(\omega) \quad \text{and} \quad \hat{T}(\omega) = -\hat{\xi}(\omega) \hat{\Omega}(\omega).$$

(2.69)
In Fourier-space, we can then involve the same strategy as outlined above to find $\gamma(\omega)$ and $\xi(\omega)$.

## 2.5 Hydrodynamics of microswimmers

### 2.5.1 The swimming problem

Microorganisms, such as bacteria, protozoa, and algae inhabit a world where viscous forces dominate over inertial forces. Both the Reynolds, as well as the oscillatory Reynolds number are typically very small for microswimmers, so that their dynamics can be described by the Stokes equations, which are linear and independent of time [5]. In this subsection we want to explore how “swimming in Stokes flow” can be formulated mathematically and which strategies microswimmers can employ to propel themselves.

**Mathematical formulation** A body immersed in a fluid is a swimmer, if it propels itself by changing its shape autonomously and periodically. In the swimmer’s body-fixed frame, we define the surface velocity $u_S(t)$ with which the swimmer deforms its surface and hence shape. Mathematically, the swimming problem is to solve the Stokes equations with the no-slip boundary condition

$$\mathbf{v} = \mathbf{U}(t) + \Omega(t) \times \mathbf{r} + u_S(t)$$

on the swimmer’s surface. The unknown translational velocity $\mathbf{U}(t)$ and angular velocity $\Omega(t)$ are determined by the condition that the swimmer experiences no external force and torque [5]. Only if the resulting translational velocity $\mathbf{U}$ averaged over one period is nonzero, the body actually propels itself and can be regarded as a swimmer. If the swimmer’s shape remains constant, i.e., the surface velocity $u_S$ is purely tangential to the swimmer’s surface, the swimmer is called a squirmer.

In ref. [72], it was shown how to find $\mathbf{U}(t)$ and $\Omega(t)$ using the reciprocal theorem. At every instant in time, we pick $\mathbf{v}_1$ and $\sigma_1$ as solutions to the swimming problem, and $\mathbf{v}_2$ and $\sigma_2$ as the solution to the corresponding drag problem, i.e., a colloid which has the same shape as the swimmer and is moving with the same velocities $\mathbf{U}$ and $\Omega$, resulting in a drag force $\mathbf{F}$ and torque $\mathbf{T}$ exerted by the fluid on the colloid. The reciprocal theorem eq. (2.50) then states

$$\int_{\partial V} dA \ (\mathbf{U} + \Omega \times \mathbf{r} + u_S) \cdot \sigma_2 n = \int_{\partial V} dA \ (\mathbf{U} + \Omega \times \mathbf{r}) \cdot \sigma_1 n. \quad (2.71)$$

By means of the relation

$$\int_{\partial V} dA \ (\mathbf{U} + \Omega \times \mathbf{r}) \cdot \sigma n = \mathbf{U} \cdot \int_{\partial V} dA \ \sigma n + \Omega \cdot \int_{\partial V} dA \ \mathbf{r} \times \sigma n, \quad (2.72)$$

"
we find that the right hand side of eq. (2.71) is zero due to the assumption that the swimmer is force and torque free. The left hand side of eq. (2.71) simplifies similarly and we find

\[ U \cdot F + \Omega \cdot T = - \int_{\partial V} dA \, u_S \cdot \sigma n. \]  

(2.73)

Hence, if the surface velocity \( u_S \) is known, we can find the velocities \( U \) and \( \Omega \) of a swimmer by solving the drag problem for each of its shapes. This result is particularly interesting for squirmers: Since they do not change shape, only one drag problem has to be solved.

**Scallop theorem and biological microswimmers** Consider a body that changes its initial shape \( A \) to a shape \( B \) and then performs exactly the same shape changes backwards to recover its initial shape \( A \). The times taken for these two shape changes \( A \to B \) and \( B \to A \) need not be equal. The motion of such a body is called reciprocal, and the scallop theorem states that a body performing reciprocal motion will not swim in Stokes flow [4]. The theorem is based on the linearity and time independence of the Stokes equations [5]. A scallop may propel itself when it opens its shell, but it reverses its translation when closing the shell, ending up at its original position. Hence, a scallop could not swim at zero Reynolds number. One can easily see that a swimmer needs at least two degrees of freedoms to perform a non-reciprocal motion [4].

Biological microswimmers employ different strategies to change shape in a non-reciprocal manner and hence escape the scallop theorem [4]. Sperm propagate a bending wave along their tail, which is similar to the movement of a snake [3]. The bacterium *E. coli* uses its flagella, whip-like appendages protruding from its body, to swim. The flagella bundle together and form a rotating helix that propels the cell body [73]. *Chlamydomonas*, a green alga, has two flexible flagella, which perform a breast-stroke-like motion [74]. Each cilium on the surface of ciliated microswimmers such as *Paramecia* or *Opalina* perform a power- and a subsequent recovery-stroke [75]. Thereby, the cilium is bent significantly stronger during the recovery stroke which results in non-reciprocity. Note that a non-reciprocal shape change is a necessary but not a sufficient condition for swimming [5]. The review article [5] demonstrates the physics behind the swimming mechanisms of several biological microswimmers.

### 2.5.2 Far field of a microswimmer

Similar to electrostatics, there is a multipole expansion for objects in Stokes flow. We start from the single layer representation of the flow around a particle derived
2.5 Hydrodynamics of microswimmers

where we integrate over the surface of the particle, which is a microswimmer in our case. Here, we abbreviated the product of the stress tensor \( \sigma \) and the surface normal vector \( n \) as \( f \). We assume this force density \( f \) to be cylindrically symmetric, i.e., \( f = f(\vec{r}, z) \) does not depend on \( \varphi \) [76]. The multipole expansion is [76]

\[
v_\alpha(r) = \int dA' \left( G_{\alpha\beta}(r) - \frac{\partial G_{\alpha\beta}(r)}{\partial r_\gamma} r'_\gamma + \frac{1}{2} \frac{\partial^2 G_{\alpha\beta}(r)}{\partial r_\gamma \partial r_\delta} r'_\gamma r'_\delta + \ldots \right) f_\beta(r')
\]

(2.75)

\[
= G_{\alpha\beta} f_\beta - G_{\alpha\beta,\gamma} D_{\beta\gamma} + \frac{1}{2} G_{\alpha\beta,\gamma\delta} Q_{\beta\gamma\delta} + \ldots,
\]

(2.76)

where we applied the Einstein summation convention, as in the remainder of this subsection, and defined the multipole moments \( F_\beta, D_{\beta\gamma}, \) and \( Q_{\beta\gamma\delta} \) as well as the dipole singularity tensor \( G_{\alpha\beta,\gamma} \) and the quadrupole singularity tensor \( G_{\alpha\beta,\gamma\delta} \). The successive terms in eq. (2.77) decay as \( r^{-1}, r^{-2}, r^{-3} \), etc. The multipole moment \( F_\beta \) is the force on the swimmer, which is zero.

Since \( \nabla \cdot v = 0 \), the Oseen tensor satisfies \( \partial_\alpha G_{\alpha\beta} = 0 \) and due to its symmetry it follows that \( \partial_\beta G_{\alpha\beta} = 0 \), i.e.,

\[
\delta_{\beta\gamma} G_{\alpha\beta,\gamma} = 0.
\]

(2.78)

Hence, the tensor \( D_{\beta\gamma} \) can be replaced by the traceless tensor \( \tilde{D}_{\beta\gamma} = D_{\beta\gamma} - \frac{1}{3} D_{\alpha\alpha} \delta_{\beta\gamma} \), yielding the same flow field, i.e., \( \tilde{D}_{\beta\gamma} G_{\alpha\beta,\gamma} = D_{\beta\gamma} G_{\alpha\beta,\gamma} \). Furthermore, \( D_{\beta\gamma} \) can be separated into a symmetric and an antisymmetric part \( \tilde{D}_{\beta\gamma} = S_{\beta\gamma} + T_{\beta\gamma} \), with

\[
S_{\beta\gamma} = \frac{1}{2} \int dA (f_\beta r_\gamma + f_\gamma r_\beta) - \frac{1}{3} \delta_{\beta\gamma} \int dA f_\alpha r_\alpha,
\]

(2.79)

\[
T_{\beta\gamma} = \frac{1}{2} \int dA (f_\beta r_\gamma - f_\gamma r_\beta).
\]

(2.80)

Evaluating these integrals under the assumption of a cylindrically symmetric force density \( f = f(\vec{r}, z) \), we find [76]

\[
S = \begin{pmatrix} -a & 0 & 0 \\ 0 & -a & 0 \\ 0 & 0 & 2a \end{pmatrix}, \quad T = \begin{pmatrix} 0 & -b & 0 \\ b & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},
\]

(2.81)
with constants $a$ and $b$. The torque on a particle is $T = \int dA \varepsilon_{\alpha\beta\gamma} r_\beta f_\gamma$, which is $\varepsilon_{\alpha\beta\gamma} T_{\beta\gamma}$ due to eq. (2.80). In the case of of our cylindrically symmetric swimmer, we obtain a torque in $z$-direction of $2b$. Since a swimmer is torque-free, $b = 0$ and the dipole contribution to the flow field is solely characterized by the parameter $a$. Due to eq. (2.78), we can replace $S_{\beta\gamma}$ by $\tilde{S}_{\beta\gamma} = S_{\beta\gamma} + \delta_{\beta\gamma} a = 3a \delta_{\beta z} \delta_{\gamma z}$, and hence, the resulting flow field is

$$v_\alpha = -\tilde{S}_{\beta\gamma} G_{\alpha\beta\gamma} = -8\pi \kappa F D \partial_z G_{\alpha z} = \kappa F D v_\alpha^{FD}(r; e_z, e_z) \equiv \kappa^{FD} v_\alpha^{FD}(r). \quad (2.82)$$

Here, we identified $v_\alpha^{FD}(r; e_z, e_z)$ from section 2.3.2 and introduced the convention that omitting the direction vectors after the semicolon corresponds to choosing all direction vectors as $e_z$. A similar analysis [76] reveals that the Stokeslet quadrupole tensor $Q_{\beta\gamma\delta}$ contains three independent parameters for a cylindrically symmetric swimmer, which couple to the source dipole, force quadrupole, and rotlet dipole, respectively. In summary, the far field of a cylindrically symmetric microswimmer up to $O(|r|^4)$ is given by

$$v(r) = \kappa^{FD} v^{FD}(r) + \kappa^{SD} v^{SD}(r) + \kappa^{FQ} v^{FQ}(r) + \kappa^{RD} v^{RD}(r) + O(|r|^4). \quad (2.83)$$

Note that all higher order contributions (octupole, hexadecapole, etc.) also have three independent coefficients each [76]. The flow fields in eq. (2.83) are given as [7]

$$v^{FD}(r) = \frac{r}{r^3} \left( \frac{3z^2}{r^2} - 1 \right), \quad (2.84)$$

$$v^{SD}(r) = \frac{1}{r^3} \left( -e_z + \frac{3zr}{r^2} \right), \quad (2.85)$$

$$v^{FQ}(r) = \frac{1}{r^3} \left[ \left( 1 - \frac{3z^2}{r^2} \right) e_z + \left( \frac{15z^3}{r^4} - \frac{9z}{r^2} \right) r \right], \quad (2.86)$$

$$v^{RD}(r) = \frac{3ze_z \times r}{r^5} = \frac{3\vec{r}z}{r^5} e_\phi. \quad (2.87)$$

The force dipole, source dipole, and force quadrupole are depicted in fig. 2.3. The rotlet dipole is easily visualized since it is proportional to $e_\phi$ in cylindrical coordinates. The rotlet dipole corresponds to a counter-clockwise rotation of fluid for $z > 0$ and to a clockwise rotation of fluid for $z < 0$.

A swimmer with positive $\kappa^{FD}$ as depicted in fig. 2.3 (a) is termed a pusher, since it pushes fluid away at the bottom and the top, and pulls fluid from the sides. The flow field of a swimmer with negative $\kappa^{FD}$, which is termed a puller, is obtained from fig. 2.3 (a) if all arrows are inverted. Biological examples for a pusher, a puller, and a neutral ($\kappa^{FD} \approx 0$) are *E. coli*, *Chlamydomonas*, and *Volvox*, respectively [74, 77]. Since the force dipole decays as $\propto 1/r^2$, it is the leading order singularity in the far field of a microswimmer. The third order singularities are the source dipole, force quadrupole, and rotlet dipole. The source dipole is attributed to the finite size of a
2.5 Hydrodynamics of microswimmers

2.5.3 Hydrodynamics of swimmers near surfaces

Hydrodynamic interactions with surfaces strongly affect a microswimmer’s dynamics. In experiments, microswimmers in confinement were found to be attracted by surfaces [79–81]. In ref. [81], it has been argued that this surface attraction is due to the force dipole flow field. A detailed theoretical study is presented in ref. [7]. Here, we summarize the procedure to find the dynamics of a microswimmer near a wall due to its hydrodynamic far field [76] and discuss the resulting dynamics [7].

First, the Green’s function of the Stokes equations for the geometry at hand is determined. For an unbounded fluid, the Green’s function of the Stokes equations is the Oseen tensor \( G(r) \) (see section 2.3.2), which is to say that the flow field generated by a point force \( f = F \delta(r - r_0) \) in an unbounded fluid is given by \( v = G(r - r_0)F \).

However, for a fluid in a semi-infinite space, bounded by an infinite planar wall with no-slip boundary condition, the flow field generated by \( f(r) = F \delta(r - r_0) \) is \( v = B(r, r_0)F \), where \( B \) is the Blake tensor [76, 82, 83]. It can be derived by a method similar to the image charge technique of electrostatics. The far field of a microswimmer can then be determined as outlined in section 2.5.2, with the exception that the Blake tensor is Taylor expanded instead of the Oseen tensor.

Again, for a cylindrically symmetric swimmer, we obtain an expansion in terms of a force dipole, source dipole, force quadrupole, rotlet dipole etc. [76]. Naturally, the corresponding flow fields will differ from their versions in an unbounded fluid. We denote the flow field of the swimmer’s far field expansion in an unbounded fluid by \( v^\infty \) and the corresponding flow field in the bounded system by \( v \). The so-called reflected flow field is defined as \( v^* = v - v^\infty \) and corresponds to a reflection of the microswimmer’s flow field from the boundary. We can interpret the solution in
the bounded system $\mathbf{v}$ as the solution for a swimmer in an unbounded fluid with background flow $\mathbf{v}^\ast$, since $\mathbf{v} = \mathbf{v}^\infty + \mathbf{v}^\ast$. Hence, the reflected flow field $\mathbf{v}^\ast$ can be substituted into Faxén’s law (eqs. (2.51) and (2.52)) as a background flow, to determine the resulting force and torque on the swimmer near a wall. Setting force and torque to zero, we obtain the swimmer’s induced velocity and angular velocity due to the interaction with the no-slip wall

$$\mathbf{U} = \left(1 + \frac{R^2}{6} \Delta\right) \mathbf{v}^\ast(r)|_{r=C},$$

$$\mathbf{\Omega} = \frac{1}{2} \nabla \times \mathbf{v}^\ast(r)|_{r=C}. \quad (2.88)$$

Here, $C$ is the swimmer’s center position and we assume the swimmer to be spherical. The total velocity of the swimmer is $\mathbf{U} = U_0 \mathbf{e} + \mathbf{\tilde{U}}$, where $\mathbf{e}$ is the swimmer’s orientation vector ($|\mathbf{e}| = 1$), and $U_0$ is its self-propulsion velocity in an unbounded fluid. In ref. [7], the analogue of eqs. (2.88) and (2.89) for a spheroidal swimmer is analyzed, which is

$$\mathbf{\tilde{U}} = \mathbf{v}^\ast + \mathcal{O} \left(\frac{b^2_2 \Delta \mathbf{v}^\ast}{z} \right) \quad (2.90)$$

$$\mathbf{\tilde{\Omega}} = \frac{1}{2} \nabla \times \mathbf{v}^\ast + \Upsilon \mathbf{e} \times \left[ (\nabla \mathbf{v}^\ast)^T + (\nabla \mathbf{v}^\ast)^T \mathbf{e} \right] + \mathcal{O} \left(\frac{b^2_2 \Delta \nabla \times \mathbf{v}^\ast}{z} \right). \quad (2.91)$$

Here, all fields are evaluated at $r = C$ and $\Upsilon = (1 - b^2_x/b^2_y)/(1 + b^2_x/b^2_y)$, where $b_x$ and $b_z$ are the semi-minor and semi-major axis of the spheroidal swimmer. Denoting the surface normal vector as $\mathbf{e}_y$, the authors of ref. [7] find the approximate result

$$\mathbf{e}_y \cdot \mathbf{\tilde{U}} = -\frac{3 \kappa_{FD}}{8 h^2} (1 - 3 \sin^2 \theta), \quad (2.92)$$

where $\theta$ is the angle between the surface and the swimmer’s orientation and $h$ is the distance between the surface and the swimmer’s center $C$. The angle $\theta$ is positive if $\mathbf{e}$ points away from the surface. Hence, a pusher ($\kappa_{FD} > 0$) is attracted to the bottom wall for $|\sin \theta| < 1/\sqrt{3}$. An approximately parallel pusher ($\kappa_{FD} > 0$, $\theta \approx 0$) is attracted to the wall, while an approximately parallel puller is repelled. On the other hand, a perpendicular pusher ($\kappa_{FD} > 0$, $\theta \approx \pi/2$) is repelled, while a perpendicular puller is attracted. These results can be intuited by the flow field of fig. 2.3 (a), imagining the wall to follow the streamlines [7]. The surface induced angular velocity is approximately [7]

$$\dot{\theta} = -\frac{3 \kappa_{FD}}{8 h^3} \left(1 + \frac{1}{2} \Upsilon\right) + \mathcal{O}(\theta^2). \quad (2.93)$$

Hence, for a pusher, we obtain a fixed point at $\theta = 0$, which corresponds to parallel alignment. The puller’s orientation on the other hand, is rotated away from the wall.
A stable alignment perpendicular to the wall has been predicted in ref. [81] for a spherical puller. These results can again be intuited by the flow field of fig. 2.3 (a), imagining the wall to follow the streamlines [7]. The same intuition can be applied to the source dipole and force quadrupole, which are discussed in detail in ref. [7].

Finally we want to discuss the dynamics of a swimmer with a rotlet dipole near a boundary. The rotlet dipole has no influence on wall-attraction and orientation ($\ddot{U} = U_0 e_\theta, \theta = 0$). However, for $\theta = 0$, it leads to a rotation of the swimmer with angular velocity parallel to the surface normal vector

$$e_y \cdot \tilde{\Omega} = -\frac{3\kappa_{RD}}{32h^4} (1 - \U).$$  \hspace{1cm} (2.94)

The radius $R_{rot}$ of this rotation can be found via $|e_y \cdot \tilde{\Omega}|R_{rot} = U_0$, as

$$R_{rot} = \frac{32h^4 U_0}{3\kappa_{RD}(1 - \U)}. \hspace{1cm} (2.95)$$

Such rotations have been observed experimentally for *E. coli* [6].

Although the analysis of surface-induced microswimmer dynamics by far-field hydrodynamics has been successful in explaining different experimental observations [6, 81], one has to keep in mind that as soon as the swimmer is very close to the surface, the hydrodynamic near field as well as steric interactions begin to dominate [77]. It has been argued that the direct contact of cilia or flagella dominate surface scattering of biological microswimmers [84]. In ref. [85] the influence of near-field hydrodynamics and steric interactions on the dynamics of a squirmer (see section 2.5.4) near a wall have been investigated. It was found that if the range of repulsive steric interactions is large enough, the qualitative picture of the far field analysis remains unchanged, i.e., pullers are trapped with an orientation perpendicular to the surface, while pushers swim nearly parallel close to the wall, pointing slightly away from it. Additionally, small amplitude oscillations of the swimmer height were observed. For very short-ranged repulsive interactions, where near-field hydrodynamics dominates, pullers change their dynamics and swim nearly parallel close to the wall, pointing slightly towards it. The latter results where also reported in ref. [86], where a fixed-point analysis was performed without taking steric interactions into account.

### 2.5.4 Squirmer model

So-called ciliated microswimmers, such as *Paramecia* (see fig. 2.4), are covered with hair-like organelles, known as cilia, beating in metachronal waves, which results in self-propulsion of the whole body. As a model for ciliated microswimmers, Lighthill proposed the squirmer model in 1952 [15], which was later revised by Blake in 1971 [16]. Though originally intended as a model for ciliated microswimmers, nowadays
it is considered as a generic model for a broad class of microswimmers, including self-phoretic particles [11–14, 87, 88]. The squirmer model has been applied to study collective effects in bulk [89–95], at surfaces [86, 89, 96], and in a narrow slit [30]. The idea is to coarse-grain the time-averaged cilia beating pattern into a mean slip velocity on an effective surface. Similarly, for self-phoretic particles an effective slip velocity arises due to self-generated gradients [14]. In fig. 2.5, this effective surface is represented by dashed lines. The actual model consists of a hard colloid, on which the coarse-grained surface velocity is prescribed, as if the surface of the colloid was tread-milling. Note that this squirming velocity corresponds to the surface velocity \( u_S \), we discussed earlier, with the simplification that the prescribed surface velocity does not depend on time, since it has already been averaged over a period. The original work [15, 16] was not restricted to squirming, i.e., it allowed \( u_S \) to have a component normal to the swimmer’s surface. However, compliant with modern literature [17], we will focus our discussion on squirming.

The original model considered by Lighthill and Blake is restricted to a spherical swimmer shape. The prescribed tangential surface velocity in spherical coordinates \((r, \theta, \varphi)\) is given by

\[
\mathbf{u}_{sq}(\theta) = \sum_{n=1}^{\infty} B_n \frac{2 \sin \theta}{n(n+1)} \frac{dP_n(\cos \theta)}{d \cos \theta} \mathbf{e}_\theta. \tag{2.96}
\]

Here, \( B_n \) are the surface velocity modes and \( P_n(x) \) the Legendre polynomials. Without solving the Stokes equations for this boundary problem, we can find the squirmer’s swimming velocity by eq. (2.73), since the drag problem for a sphere is well known. The surface stress is \( \sigma_2 \mathbf{n} = -3\eta \mathbf{U}/(2R) \) and the force is \( \mathbf{F} = -6\pi \eta R \mathbf{U} \),
which yields the swimming velocity \[ U_0 = -\frac{1}{4\pi R^2} \int_{\partial V} dA \, \mathbf{u}_S \cdot \mathbf{e}_z = \frac{2}{3} B_1, \] (2.97)

where we used that the swimmer can only translate in \( z \)-direction due to the cylindrical symmetry of the squirmer velocity. Hence, the first surface velocity mode \( B_1 \) determines the propulsion.

The flow field of a squirmer is easily found, since eq. (2.96) is a restriction of Lamb’s general solution of the Stokes equations \[71\] to the surface of a sphere. The force-free condition yields the flow field \[97\]

\[ \mathbf{v}(r, \theta, \phi) = \frac{2 \cos \theta}{3} R^3 B_1 \mathbf{e}_r + \frac{\sin \theta}{3} R^3 B_1 \mathbf{e}_\theta + \sum_{n=2}^{\infty} B_n \left( \frac{R^{n+2}}{r^{n+2}} - \frac{R^n}{r^n} \right) P_n(\cos \theta) \mathbf{e}_r 
+ \sum_{n=2}^{\infty} B_n \left( \frac{R^{n+2}}{r^{n+2}} - \frac{n - 2 R^n}{n} \right) \frac{\sin \theta}{n+1} \frac{dP_n(\cos \theta)}{d \cos \theta} \mathbf{e}_\theta. \] (2.98)

The flow field due to the first mode is

\[ \mathbf{v}_1 = \frac{B_1 R^3}{3} (2 \cos \theta \mathbf{e}_r + \sin \theta \mathbf{e}_\theta) = \frac{B_1 R^3}{3} \mathbf{v}^{SD}(r), \] (2.99)

where the last equality follows by comparison with eq. (2.85) in spherical coordinates and with the identity \( \mathbf{e}_z = \cos \theta \mathbf{e}_r - \sin \theta \mathbf{e}_\theta \). Hence, the first mode \( B_1 \) generates the flow field of a source dipole \[97\] with coefficient \( \kappa^{SD} = B_1 R^3/3 \). The flow field due to the second mode is

\[ \mathbf{v}_2 = -\frac{B_2 R^2}{2r^2} (3 \cos^2 \theta - 1) \mathbf{e}_r + \frac{B_2 R^4}{2r^4} ((3 \cos^2 \theta - 1) \mathbf{e}_r + \sin(2\theta) \mathbf{e}_\theta) \] (2.100)

\[ = -\frac{B_2 R^2}{2} \mathbf{v}^{FD}(r) + \frac{B_2 R^4}{6} \mathbf{v}^{SQ}(r). \] (2.101)

Hence, it consists of a force dipole with coefficient \( \kappa^{FD} = -B_2 R^2/2 \) (see eq. (2.84)) and a source quadrupole with coefficient \( \kappa^{SQ} = B_2 R^4/6 \). The source quadrupole was not discussed in section 2.5.2. It is an octupolar contribution decaying like \( r^{-4} \) for \( r \to \infty \) \[7\]

\[ \mathbf{v}^{SQ}(r) = \frac{3}{r^4} \left( \frac{5z^2 r}{r^3} - \frac{2z e_z + r}{r} \right). \] (2.102)

In current studies, the modes \( B_n \) for \( n > 2 \) are often neglected \[17\] and the squirmer is characterized by the ratio \( \beta = B_2/B_1 \). A positive \( \beta \) corresponds to a negative \( \kappa^{FD} \) and hence to a puller, while a negative \( \beta \) corresponds to a pusher.

An extension of the squirmer model to spheroidal shapes has been put forward in refs. \[98, 99\] with \( B_n = 0 \) for \( n > 1 \). A spheroidal squirmer with modes \( B_n \in \mathbb{R} \)
for all $n \in \mathbb{N}$ was introduced in ref. [86]. However, to the best of our knowledge, it is not possible to determine the flow field of the spheroidal squirmer of ref. [86] analytically. We will introduce an alternative model for a spheroidal squirmer in chapter 9.
Chapter 3

Brownian motion

In section 2.4 we discussed that a colloid experiences a hydrodynamic drag exerted by the surrounding fluid. Additionally, thermal fluctuations of the microscopic fluid particles will lead to random collisions with the colloid, which can be summed up into a random force and torque. The resulting random motion of the colloid is termed Brownian motion.

Drag and random forces are not independent, but rather linked, which is expressed by the so-called fluctuation dissipation relations, as will be illustrated in section 3.1 for an isotropic colloid. Note that an abbreviated version of section 3.1 was previously published in publication [C] (see List of publications on page 203). Subsequently, we introduce the concept of Gaussian white noise in section 3.2, and extend the discussion to anisotropic bodies in section 3.3. We briefly discuss the concept of active Brownian particles in section 3.4, before we illustrate the discretization and computer simulation of Brownian motion in section 3.5.

3.1 Fluctuation dissipation relations for isotropic colloids

Translational motion The translational equation of motion of a single isotropic, colloidal Brownian particle of mass $M$ and velocity $U(t)$ in a viscous unbounded fluid, experiencing a hydrodynamic drag force (see section 2.4, eq. (2.61)) together with a random force $K(t)$ due to thermal fluctuations, is given by the generalized Langevin equation [100]

$$M \frac{dU}{dt} = - \int_{-\infty}^{t} \! \! \! \! \! \! \! \gamma(t-t')U(t') + K(t).$$

Note that the friction matrix $\gamma(t)$ of eq. (2.61) becomes a scalar $\gamma(t)$ due to the isotropic shape of the colloid. We changed the lower integration time in eq. (2.68) from $t' = -\infty$ to $t' = 0$ [100], since we assume the colloid to rest until $t' = 0$. The random force is a stochastic process [101], i.e., a time-dependent random variable, for which we assume zero mean, $\langle K(t) \rangle = 0$, and no correlations between its Cartesian
components, i.e., \( \langle K_\alpha(t)K_\beta(t') \rangle = 0 \) for \( \alpha \neq \beta \). Since we assume the system to be in thermal equilibrium, the time origin is arbitrary. Hence, any correlation function \( \langle a(t)b(t') \rangle \) for observables \( a(t), b(t) \) only depends on the time difference \( |t - t'| \) [100]. By multiplying eq. (3.1) by \( U(0) \), averaging over different realizations of \( K \), and assuming \( \langle K(t) \cdot U(0) \rangle = 0 \), we find the governing equation [100]

\[
M \frac{dC_U(t)}{dt} = - \int_0^t dt' \gamma(t - t')C_U(t'),
\]

for the velocity autocorrelation function (VACF)

\[
C_U(t) = \frac{1}{3} \langle U(t) \cdot U(0) \rangle.
\]

We can solve eq. (3.2) by performing a Laplace transformation [100], with \( \tilde{f}(z) = \int_0^\infty dt \, e^{-zt} f(t) \), and obtain

\[
\hat{C}_U(z) = \frac{C_U(0)}{\tilde{\gamma}(z)/M + z} = \frac{k_B T}{\tilde{\gamma}(z) + Mz},
\]

with the initial condition \( C_U(0) = k_B T/M \), i.e., equipartition of energy, where \( z \) is the Laplace variable, \( k_B \) is Boltzmann’s constant, and \( T \) the temperature. Since \( C_U(-t) = C_U(t) = C_U^*(t) \), the correlation function \( \hat{C}_U(z) \) is related with the Fourier transform

\[
\hat{C}_U(\omega) = \int_{-\infty}^{\infty} dt \, e^{i\omega t} C_U(t)
\]

of \( C_U(t) \) according to

\[
\hat{C}_U(\omega) = \int_{-\infty}^{\infty} dt \, e^{i\omega t} C_U(t) = \int_0^\infty dt \, e^{i\omega t} C_U(t) + \int_{-\infty}^{0} dt \, e^{i\omega t} C_U(t)
\]

\[
= \int_0^\infty dt \, (e^{-i\omega t} C_U(-t) + e^{i\omega t} C_U(t)) = 2 \text{Re} \left\{ \int_0^\infty dt \, e^{i\omega t} C_U(t) \right\}
\]

\[
= 2 \text{Re} \{ \hat{C}_U(z = -i\omega) \}.
\]

In the following, we will exclusively work in Fourier space for convenience. Combining eq. (3.6) and eq. (3.4), we find

\[
\hat{C}_U(\omega) = 2 \text{Re} \left\{ \frac{k_B T}{\tilde{\gamma}(\omega) - i\omega M} \right\},
\]

where we used \( \tilde{\gamma}(z = -i\omega) = \tilde{\gamma}(\omega) \). The latter relation follows from \( \gamma(t) = 0 \ \forall t < 0 \), which is in turn due to causality. Similarly, the autocorrelation function of the random force (FACF) can be determined

\[
C_K(t) = \frac{1}{3} \langle K(t) \cdot K(0) \rangle.
\]
In order to find $C_K(t)$ we establish a relation between $\langle \hat{U}(\omega_1) \cdot \hat{U}^*(\omega_2) \rangle$ and $\langle \hat{K}(\omega_1) \cdot \hat{K}^*(\omega_2) \rangle$. First we note that for an observable $\hat{a}$ (e.g., $\hat{a} = \hat{U}$ or $\hat{a} = \hat{K}$)

\[
\frac{1}{3} \langle \hat{a}(\omega_1) \cdot \hat{a}^*(\omega_2) \rangle = \int dt_1 \int dt_2 \ e^{i\omega_1 t_1} e^{-i\omega_2 t_2} \frac{1}{3} \langle a(t_1) \cdot a(t_2) \rangle \\
= \int dt_0 \ e^{i(\omega_1 - \omega_2) t_0} \int dt \ e^{i\omega t} \frac{1}{3} \langle a(t + t_0) \cdot a(t_0) \rangle \\
= 2\pi \delta(\omega_1 - \omega_2) \hat{C}_a(\omega_1).
\]

(3.9)

Secondly, the Fourier transformation of eq. (3.1) reads

\[
\hat{K}(\omega) = (\gamma(\omega) - i\omega M) \hat{U}(\omega).
\]

(3.10)

Here, we extended the integration range in eq. (3.1) to $-\infty$ to $\infty$, which is justified as $\gamma(t - t') = 0$ for $t' > t$ and $U(t') = 0$ for $t' < 0$. With eq. (3.10) follows

\[
\frac{1}{3} \langle \hat{K}(\omega_1) \cdot \hat{K}^*(\omega_2) \rangle = (\gamma(\omega_1) - i\omega_1 M)(\gamma^*(\omega_2) + i\omega_2 M) \frac{1}{3} \langle \hat{U}(\omega_1) \cdot \hat{U}^*(\omega_2) \rangle \\
= |\gamma(\omega_1)|^2 2\pi \delta(\omega_1 - \omega_2) 2\text{Re}\left\{ \frac{k_B T}{\gamma(\omega_1) - i\omega_1 M} \right\} \\
= 2k_B T \text{Re}\{\gamma(\omega)\} 2\pi \delta(\omega_1 - \omega_2),
\]

(3.11)

where we used eq. (3.9) and eq. (3.7) for the second equality. Finally, comparing eq. (3.9) for $\hat{a} = \hat{K}$ with eq. (3.11) yields

\[
\hat{C}_K(\omega) = 2k_B T \text{Re}\{\gamma(\omega)\}.
\]

(3.12)

This equation relates the fluctuations of the random force $\hat{K}$ with the dissipative properties of the fluid $\gamma$, and is therefore called a fluctuation-dissipation relation. The time integral of the force autocorrelation function is related to the zero-frequency friction coefficient according to

\[
\int_0^\infty dt \ C_K(t) = \lim_{\omega \rightarrow 0} \frac{1}{2} \int_0^\infty dt \ (e^{i\omega t} C_K(t) + e^{-i\omega t} C_K(t)) = \frac{1}{2} \hat{C}_K(\omega = 0) \\
= k_B T \hat{\gamma}(\omega = 0).
\]

(3.13)

Equation (3.13) is an example of a Green Kubo relation [102], which connects a transport coefficient, such as $\hat{\gamma}(\omega = 0)$, to an integral of a time correlation function. The diffusion coefficient $D$ is defined as the time integral of the velocity autocorrelation function and is related to the friction coefficient via

\[
D = \int_0^\infty dt \ C_U(t) = \frac{1}{2} \hat{C}_U(\omega = 0) = \frac{k_B T}{\hat{\gamma}(\omega = 0)}.
\]

(3.14)
which is known as Einstein-Sutherland relation. The relation between the mean-square displacement and the diffusion constant is \[ \langle (C(t) - C(0))^2 \rangle = 3 \int_0^t dt_1 \int_0^t dt_2 \langle U(t_1) \cdot U(t_2) \rangle = 3 \int_0^t dt_1 \int_0^t dt_2 C_U(|t_1 - t_2|) \]

\[
= 3 \left( \int_{-t}^0 d\tau \int_0^{t+\tau} dt_1 C_U(|\tau|) + \int_0^t d\tau \int_0^t dt_1 C_U(|\tau|) \right) \\
= 6t \int_0^t d\tau C_U(\tau) - 6 \int_0^t d\tau C_U(\tau) \tau \rightarrow 6Dt.
\]

For the change of variables in the integrals see fig. 3.1. The limit \( t \rightarrow \infty \) is performed under the assumption that \( C_U(\tau) \) decays faster than \( \tau^{-1} \) for \( \tau \rightarrow \infty \).

**Rotational motion**  Similarly, the rotational equation of motion of an isotropic colloid is

\[
I d\Omega \over dt = -\int_0^t dt' \xi(t - t')\Omega(t') + N(t),
\]

where \( I \) is the moment of inertia, \( \Omega \) is the angular velocity, \( \xi(t) \) is the rotational friction, and \( N \) is the random torque. This equation is mathematically equivalent to eq. (3.1). Hence, all results for the translational motion apply here as well. In particular, eq. (3.2) to eq. (3.14) hold, when we replace \( M \) by \( I \), \( U \) by \( \Omega \), \( \gamma \) by \( \xi \), \( K \) by \( N \), and \( D \) by \( D_R \), with the rotational diffusion coefficient \( D_R \).

### 3.2 Gaussian white noise

Until now, we characterized the random force by its first and second moment, i.e., its mean \( \langle K(t) \rangle = 0 \) and variance \( \langle K_\alpha(t)K_\beta(t') \rangle = \delta_{\alpha\beta}C_K(|t-t'|) \). Since the random
force is a result of many collisions with fluid particles, it is well justified by the central limit theorem to assume the random force to be Gaussian distributed. Hence, mean and variance already fully characterize the random force [104].

If the friction force follows the velocity \( \mathbf{U}(t) \) instantaneously, i.e., \( \mathbf{F} = -\gamma(t)\mathbf{U} \), we can substitute \( \gamma(t) = \gamma_0 \delta(t) \) in section 3.1. When \( \gamma(t) \) is given by a delta function, its Fourier transform \( \hat{\gamma}(\omega) \) is constant, i.e., \( \hat{\gamma}(\omega) = \gamma_0 \). Hence, the random force autocorrelation in frequency space (see eq. (3.12)) is constant as well

\[
\hat{C}_K(\omega) = 2k_B T \gamma_0.
\]

(3.17)

In this case, the random force is called white. This term originates from a comparison to white light, which contains all visible electromagnetic frequencies equally. If \( \hat{C}_K(\omega) \) depends on \( \omega \), the random force is colored. In the time domain eq. (3.17) translates to \( \dot{C}_K(t) = 2k_B T \gamma_0 \delta(t - t') \), and hence, the first and second moment of the random force are

\[
\langle K(t) \rangle = 0, \quad \langle K_\alpha(t)K_\beta(t') \rangle = 2k_B T \gamma_0 \delta_{\alpha\beta} \delta(t - t').
\]

(3.18)

Equation (3.18) shows that random forces at different times are uncorrelated, hence the white random force describes a Markovian stochastic process [101]. Gauss-Markov stochastic processes have many applications in modeling, even outside of physics (see ref. [101]). Equation (3.7) reveals, that for a Gaussian and Markovian random force, the velocity autocorrelation will decay exponentially

\[
C_U(t) = \frac{k_B T}{M} e^{-\gamma_0/M} t.
\]

(3.19)

For the rotational motion, the same discussion applies, and eq. (3.18) holds, when \( K \) is replaced by \( N \) and \( \gamma_0 \) by \( \xi_0 \).

### 3.3 Anisotropic colloids

For an anisotropic rigid body of revolution, the friction kernels \( \gamma(t) \) and \( \xi(t) \) are matrices \( \mathbf{\gamma}(t) \) and \( \mathbf{\xi}(t) \) (see section 2.4). For the translational dynamics, all results of section 3.1 remain valid, but have to be replaced by corresponding matrix expressions. However, the rotational equations of motion are no longer mathematically equivalent to the translational equations and the derivations of section 3.1 do not apply.

If the friction force and torque follow velocity and torque instantaneously

\[
\mathbf{F} = -\mathbf{\gamma U}, \quad \mathbf{T} = -\mathbf{\xi \Omega},
\]

(3.20)

and the random force \( \mathbf{K} \) and random torque \( \mathbf{N} \) are Gaussian white noise with

\[
\langle K(t) \rangle = 0, \quad \langle K_\alpha(t)K_\beta(t') \rangle = 2k_B T \gamma_{\alpha\beta} \delta(t - t'),
\]

(3.21)

\[
\langle N(t) \rangle = 0, \quad \langle N_\alpha(t)N_\beta(t') \rangle = 2k_B T \xi_{\alpha\beta} \delta(t - t'),
\]

(3.22)
it can be shown \[105\] that the rigid body equations of motion (see chapter 1) supplemented with the above friction and random forces and torques will obey the equipartition theorem, i.e.,

\[
\langle U_\alpha^2 \rangle = \frac{k_B T}{M} \quad \text{and} \quad \langle (\Omega_\alpha^b)^2 \rangle = \frac{k_B T}{I_{b\alpha}^\alpha}.
\] (3.23)

Here, the superscript \(b\) stands for the body-fixed frame as introduced in chapter 1. Note that eqs. (3.21) and (3.22) simplify when transformed to the body-fixed frame, where \(\gamma\) and \(\xi\) are diagonal (see section 2.4)

\[
\langle K^b(t) \rangle = 0, \quad \langle K_\alpha^b(t) K_\beta^b(t') \rangle = 2k_B T \gamma_\alpha \delta_{\alpha \beta} \delta(t - t'), \quad \langle N^b(t) \rangle = 0, \quad \langle N_\alpha^b(t) N_\beta^b(t') \rangle = 2k_B T \xi_\alpha \delta_{\alpha \beta} \delta(t - t').
\] (3.24) (3.25)

### 3.4 Active Brownian particles

The model of an active Brownian particle is a strongly simplified model of a microswimmer and can be derived from the squirmer model introduced in section 2.5. Note that the model of an active Brownian particle we present here, is well established and frequently applied in the literature \[11\], but several slightly different models exist, which are also termed active Brownian particles. For a review, see refs. \[11, 106\].

Consider a single spherical (or spheroidal) squirmer in an unbounded fluid with thermal fluctuations. The prescribed surface velocity on the squirmer yields a force \(F = -\gamma(U - U_0 e)\) \[107\], resulting in the rigid body equations of motion (see section 1.2.2)

\[
M \ddot{C} = -\gamma(U - U_0 e) + K, \quad \dot{i} = -\xi \Omega + N, \quad \dot{q} = \frac{1}{2} Q(q) \left( \begin{array}{c} 0 \\ \Omega^b \end{array} \right).
\] (3.26) (3.27) (3.28)

Here, \(e = D^T e_z\) is the orientation of the swimmer, \(U_0\) is its mean velocity, and \(K\) and \(N\) are the Gaussian and Markovian random force and torque from section 3.3. Note that within the hydrodynamic theory of swimmers, one typically assumes the steady state condition, where the swimmer is force free, i.e., \(U = U_0 e\).

If other squirmers or passive objects like walls are introduced, we have to account for the steric and hydrodynamic interactions with these objects in our equations of motion. In the model of active Brownian particles, however, hydrodynamic interactions are neglected. Only steric interactions are accounted for via forces and torques, which are added to the right hand sides of eqs. (3.26) and (3.27). Typically, the overdamped limit is assumed, which means that the friction forces and torques
3.5 Discretization and Langevin dynamics simulations

are much larger than the inertial terms $M\ddot{C}$ and $\dot{l}$. For a spherical active Brownian particle this yields the equations

\[
\dot{C} = U_0 e + \frac{1}{\gamma}(K + F^{\text{det}}), \tag{3.29}
\]

\[
\dot{e} = (N/\xi) \times e. \tag{3.30}
\]

Here, we set $\ddot{C} = \dot{l} = 0$ (overdamped limit) in eqs. (3.26) and (3.27), which yields eq. (3.29) and the equation $\Omega = \xi^{-1}N$. Furthermore, for the orientation dynamics of a sphere it suffices to track the movement of only one axis. Hence, eq. (3.28) can be substituted by $\dot{e} = \Omega \times e$. Finally, we used $\gamma = 6\pi\eta R$ and $\xi = \frac{8\pi\eta R^3}{\gamma}$ and added a deterministic force $F^{\text{det}}$ for steric interactions and external forces.

The mean-square displacement of a force-free ($F^{\text{det}} = 0$) active Brownian particle is enhanced, since

\[
\langle (C(t) - C(0))^2 \rangle = \int_0^t dt_1 \int_0^t dt_2 \left\langle \dot{C}(t_1) \cdot \dot{C}(t_2) \right\rangle \tag{3.31}
\]

\[
= \int_0^t dt_1 \int_0^t dt_2 \left[ U_0^2 \langle e(t_1) \cdot e(t_2) \rangle + \gamma^{-2} \langle K(t_1) \cdot K(t_2) \rangle \right] \tag{3.32}
\]

\[
= U_0^2 \int_0^t d\tau \frac{2(1 - \exp(-2D_R\tau))}{\gamma} + \frac{6k_BT}{\gamma} t \tag{3.33}
\]

\[
= 6 \left( \frac{U_0^2}{6D_R} + \frac{k_BT}{\gamma} \right) t - \frac{U_0^2}{2D^2_R}(1 - \exp(-2D_Rt)). \tag{3.34}
\]

Here, we assumed $\langle K(t_1) \cdot e(t_2) \rangle = 0$, employed the substitution of variables as in eq. (3.15) (see fig. 3.1), and used $\langle e(t_1) \cdot e(t_2) \rangle = \exp(-2D_R|t_1 - t_2|)$, which is shown in ref. [63] based on eq. (3.30). Hence, the total diffusion coefficient is $D = U_0^2/(6D_R) + k_BT/\gamma$ and is typically dominated by the contribution proportional to $U_0^2$ [12].

3.5 Discretization and Langevin dynamics simulations

In this section we will present how to simulate the Langevin dynamics (LD) of anisotropic rigid bodies. In applications, we will always work in parameter regimes where inertia is negligible. However we will still include it in our simulations, since we found that algorithms neglecting inertia need significantly smaller time steps. Note that in principle, for systems containing several bodies, hydrodynamic interactions need to be accounted for, as outlined, e.g., in ref. [108]. Here, we will neglect those hydrodynamic interactions. Hence, each particle is only subjected to the local fluid drag, thermal fluctuations, and steric interactions. The simulation algorithm works for active as well as passive ($U_0 = 0$) Brownian particles.
3.5.1 Algorithm

In section 1.3, we outlined how to integrate the rigid body equations of motion (eqs. (1.26), (1.31) and (1.35)) numerically. Here, we include friction, and random forces and torques. We assume that friction forces and torques follow velocity and angular velocity instantaneously (see eq. (2.61)) and that the random force and torque are Gaussian white noise (see eqs. (3.24) and (3.25)).

The inclusion of friction forces and torques is straightforward. We determine the friction matrices $\gamma$ and $\xi$ in the body-fixed frame analytically and add the resulting forces and torques to the rigid body integrator. If active Brownian particles are considered, we implement the force $F = -\gamma(U - U_0 e)$ (see section 3.4).

The inclusion of Gaussian white noise is more involved. Here, we present the most simple approach, which relies on a very small time step. For higher order methods and a more detailed and rigorous discussion see refs. [109, 110]. Since time is discretized to $t_i = ih$ with $i \in \mathbb{N}$ and time step $h$, we need to discretize the delta function $\delta(t - t')$ which appears in eqs. (3.21) and (3.22). According to the trapezoidal rule [111], the integral of a function $f(t)$ with values $f_i = f(ih)$ is discretized as

$$\int_0^\infty dt \ f(t) \to \frac{h}{2} f_0 + h \sum_{i=1}^{\infty} f_i. \quad (3.35)$$

We can convince ourselves that the discretized delta function is

$$\delta(t - t') \to \frac{1}{h} \delta_{i,i'}, \quad (3.36)$$

where $t_{i'} = i'h = t'$. This is consistent with the continuum behavior of the delta function, since

$$\int_0^\infty dt \ f(t) \delta(t - t') \to \frac{1}{2} f_0 \delta_{0,i'} + \sum_{i=1}^{\infty} f_i \delta_{i,i'} \quad (3.37)$$

yields $f_0/2$ for $i' = 0$ and $f_{i'}$ for $i' > 0$.

Hence, in the body-fixed frame, the $\alpha$-component of the random force acting on the rigid body at time $t_i$ is a Gaussian random number with mean 0 and variance $2k_B T \gamma_\alpha/h$ (see eqs. (3.24) and (3.36)). A new, uncorrelated random number is drawn for each time step and Cartesian direction. In summary, $K^b(t_i)$ is a Gaussian random number with

$$\langle K^b(t_i) \rangle = 0, \quad (3.38)$$
$$\langle K^b_\alpha(t_i) K^b_\beta(t_j) \rangle = \frac{2k_B T \gamma_\alpha}{h} \delta_{\alpha\beta} \delta_{ij}. \quad (3.39)$$
3.5 Discretization and Langevin dynamics simulations

The same holds for the random torque, but with variance $2k_B T \xi_\alpha / h$, i.e., $N^b(t_i)$ is a Gaussian random number with

$$\langle N^b(t_i) \rangle = 0,$$  \hspace{1cm} (3.40)

$$\langle N^b(t_i) N^b(t_j) \rangle = \frac{2k_B T \xi_\alpha}{h} \delta_{\alpha \beta} \delta_{ij}. \hspace{1cm} (3.41)$$

In a computer simulation, we implement the Gaussian random numbers by pseudo-random numbers using the Box-Muller transform [111]. As mentioned above, the algorithm is only applicable for small time steps $\gamma_\alpha h / M \ll 1$.

3.5.2 Simulation results

We validate the Langevin-dynamics algorithm by simulating the diffusion of a single spheroidal rigid body with semi-major axis $b_z = 2$, semi minor-axis $b_x = 1$ and mass density $\rho = 10$ in a fluid of viscosity $\eta = 20$ at $k_B T = 1$\(^a\). We employ the friction coefficients $\gamma_\alpha$ and $\xi_\alpha$ specified in section 2.4. First, we switch off random forces and torques as well as drag forces and torques and check energy conservation. Then, we switch on drag as well as random forces and torques and check the equipartition theorem. After these basic checks, we determine the orientation autocorrelation function $\langle e(t) \cdot e(0) \rangle$, where $e$ is the unit vector pointing in the direction of the major axis. The theory of rotational Brownian motion [112] predicts

$$\langle e(t) \cdot e(0) \rangle = \exp \left(-2D_R t \right), \hspace{1cm} (3.42)$$

where $D_R = k_B T / \xi^\perp$, and $\xi^\perp$ is given in eq. (2.65). Simulation results for the orientation autocorrelation function\(^b\) are shown in fig. 3.2 for different time steps $h$. Furthermore, we determine the mean-square displacement $\langle (C(t) - C(0))^2 \rangle$, which obeys [63]

$$\langle (C(t) - C(0))^2 \rangle \rightarrow 6Dt, \text{ for } t \rightarrow \infty \hspace{1cm} (3.43)$$

with $D = (2D^\perp + D^\parallel) / 3$, $D^\perp = k_B T / \gamma^\perp$, and $D^\parallel = k_B T / \gamma^\parallel$, where $\gamma^\perp$ and $\gamma^\parallel$ are given in eqs. (2.62) and (2.63). The simulation results\(^b\) are presented in fig. 3.2.

Both, the orientation correlation function as well as the mean-square displacement show good agreement with theory for $h = 0.01$ and $h = 0.001$. For $h = 0.1$, however, the algorithm diverges. This agrees with the fact that $h$ should be much smaller than $M / \gamma_\alpha$, which is around 0.5 here. In conclusion, the presented Langevin dynamics algorithm for rigid bodies produces adequate results as long as a sufficiently small time step is chosen.

\(^a\)For this section, we define units by setting $k_B T = 1$, $b_x = 1$, $\rho = 10$.

\(^b\)The autocorrelation function and the mean-square displacement of a discrete time series can be efficiently computed numerically using the Fast Fourier Transform (FFT). For the FFT, we refer to ref. [111], while the FFT-based algorithms for the computation of correlation functions and the mean-square displacement are discussed in sections 4.1 and 4.2 of ref. [113].
Figure 3.2: (a) Orientation autocorrelation function and (b) mean-square displacement of a prolate spheroid with $b_z = 2, b_x = 1$ in a fluid of viscosity $\eta = 20$ at $k_B T = 1$. The solid black lines show the theoretical expectation (eq. (3.42) and eq. (3.43)), while the blue dashed and green dotted lines correspond to simulation results with time step $h = 0.01$ and $h = 0.001$, respectively.
Chapter 4

Multiparticle collision dynamics

In this chapter, we present the multiparticle collision dynamics simulation method. The method has been well described in various articles and reviews [33, 34]. Here, we base our presentation on publication [A] (see List of publications on page 203). Verbatim quotes of publication [A] are indicated as “…”[A], can extend over several pages and have been reformatted to fit with the main text. The label of a cited article inside a verbatim quote follows the enumeration in the bibliography (page 186).

4.1 Introduction

“During the last few decades various mesoscale hydrodynamic simulation techniques have been developed and have been applied to study soft matter systems. Prominent examples are Lattice Boltzmann (LB) [114–116], dissipative particle dynamics (DPD) [117, 118], and multiparticle collision dynamics (MPC) [32–34, 119]. Common to these approaches is a simplified, coarse-grained description of the fluid degrees of freedom while maintaining the essential microscopic physics on the length scales of interest.

Multiparticle collision dynamics is a particle-based hydrodynamic simulation method, which incorporates thermal fluctuations, provides hydrodynamic correlations, and is easily coupled with other simulation techniques, such as molecular dynamics simulations for embedded particles [33, 34]. MPC proceeds in a ballistic streaming step and a collision step. Collisions occur at fixed discrete time intervals and establish a local stochastic interaction between particles. Thereby, the particles are sorted into cells to define the multiparticle-collision environment. Various schemes for the collisional interaction have been proposed [32–34, 119–121]. The original method, which employs rotation of relative velocities, is often denoted as stochastic-rotation dynamics (SRD) [32–34, 119–121]. Other methods, adopting an Andersen-thermostat-like idea, denoted as MPC-AT, use Gaussian distributed random numbers for the relative velocities [120, 121][A].

It has been shown by a Chapman-Enskog expansion that the MPC-SRD fluid obeys the Navier-Stokes equations with an ideal-gas equation of state [32, 35]. An H-theorem has been derived [32, 36], which guarantees relaxation of the MPC fluid
to equilibrium and it has been shown that the equilibrium distribution of velocities is Maxwellian [32, 36]. Furthermore the MPC algorithm exhibits unconditional numerical stability [34]. “In the simplest version, angular momentum is not conserved in a collision. However, angular-momentum-conserving extensions have been introduced for both, the stochastic rotation version of MPC (MPC-SRD+a) and the Andersen variant (MPC-AT+a) [68, 121]. The non-angular momentum conserving methods will be denoted by MPC-SRD-a and MPC-AT-a, respectively.

The MPC method has successfully been applied to a broad range of soft matter systems ranging from equilibrium colloid [32–34, 39, 40, 48, 122–127] and polymer [33, 34, 42, 43, 128] solutions and, more importantly, non-equilibrium systems such as colloids [38, 120, 129–132], polymers [41, 42, 133–141], vesicles [44], and cells [45, 46] in flow fields, colloids in viscoelastic fluids [142–144], to self-propelled spheres [90, 145, 146] and rods [33, 147], squirmers [8, 30, 90, 148], and other microswimmers [73, 149–152]”[A]. Moreover, extensions have been proposed to fluids with non-ideal equations of state [153], binary fluids [154, 155], and nematohydrodynamics [156].

4.2 Algorithm

We will introduce the MPC algorithm in three dimensions. A reduction to two dimensions is straightforward. “The MPC fluid consists of \( N \) point particles with mass \( m \), continuous positions \( \mathbf{r}_i \) and velocities \( \mathbf{v}_i \), which undergo streaming and subsequent collision steps”[A] (see fig. 4.1).

**Streaming and collisions** “In the streaming step, the particles move ballistically during the time interval \( h \), denoted as collision time, and their positions are updated according to

\[
\mathbf{r}_i(t + h) = \mathbf{r}_i(t) + h\mathbf{v}_i(t). \tag{4.1}
\]

In the collision step, the rectangular cuboid fluid volume \( V \) of dimensions \( L_x \times L_y \times L_z \) is divided into cubic cells of length \( a \) to define the multiparticle collision environment”[A]. Useful data structures for the sorting of particles into collision cells are linked lists (see the section on “Cell structures and linked lists” in ref. [157]). “Particles sharing a cell exchange momenta in a stochastic process, whereby the total linear momentum is conserved. The velocity \( \mathbf{v}'_i \) of particle \( i \) after collision is given by

\[
\mathbf{v}'_i = \mathbf{v}_{cm} + C [\mathbf{v}_{ic}]. \tag{4.2}
\]

Here, \( \mathbf{v}_{cm} = (1/N_c) \sum_{i=1}^{N_c} \mathbf{v}_i \) is the center-of-mass velocity, \( \mathbf{v}_{ic} = \mathbf{v}_i - \mathbf{v}_{cm} \) is the particle’s relative velocity with respect to the center-of-mass velocity, \( N_c \) is the total
4.2 Algorithm

Streaming step (during interval $h$)  
Collision step (instantaneously)

Figure 4.1: Schematic representation of multiparticle collision dynamics. Black circles represent particle positions, black arrows represent velocities. Green circles (arrows) represent the new positions (velocities) after streaming (collision). The collision cells are indicated by dashed lines.

number of particles in the cell, and $\mathcal{C}$ is the collision operator\textsuperscript{[A]}. Note that the collision step is only performed for cells containing at least two particles.

For MPC-SRD-a, the operator $\mathcal{C}$ corresponds to a rotation around a randomly oriented axis $(n_x, n_y, n_z)$ by the constant angle $\alpha$, i.e.,

$$\mathcal{C}[v_{ic}] = R(\alpha)v_{ic}. \quad (4.3)$$

Thereby, the matrix $R(\alpha)$ can be represented as (see eq. (1.6))

$$R(\alpha) = \begin{pmatrix}
  n_x^2 (1 - c) + c & n_xn_y (1 - c) - n_zs & n_xn_z (1 - c) + n_y s \\
  n_y n_x(1 - c) + n_z s & n_y^2 (1 - c) + c & n_y n_z (1 - c) - n_x s \\
  n_z n_x (1 - c) - n_y s & n_z n_y (1 - c) + n_x s & n_z^2 (1 - c) + c
\end{pmatrix}, \quad (4.4)$$

where $c = \cos \alpha$, $s = \sin \alpha$, and the unit vector $(n_x, n_y, n_z)^T$ is chosen randomly for each time step and collision cell\textsuperscript{[32–34]} according to

$$n_x = \sqrt{1 - u^2} \cos \phi, \ n_y = \sqrt{1 - u^2} \sin \phi, \ n_z = u. \quad (4.5)$$

Here, $\phi \in [0, 2\pi]$ and $u \in [-1, 1]$ are uncorrelated uniformly distributed random numbers. This choice guarantees a uniform distribution of $(n_x, n_y, n_z)^T$ on the unit sphere. The constant angle $\alpha$ is typically chosen as $\alpha = 130^\circ$, to obtain a fluid-like behavior, i.e., a high Schmidt number $\eta/(\rho D)$ (see ref. [158]).

Note that the rotation matrix for SRD in two dimensions is

$$R(\alpha) = \begin{pmatrix}
  \cos(\alpha) & -u \sin(\alpha) \\
  u \sin(\alpha) & \cos(\alpha)
\end{pmatrix}, \quad (4.6)$$

where $u \in \{-1, 1\}$ is chosen with uniform probability and $\alpha$ is typically chosen as $\alpha = 90^\circ$. 
“In MPC-AT-a, a velocity $v_{i}^{\text{ran}}$ is chosen for each particle $i$ with Gaussian distributed Cartesian components of zero mean and variance $k_{B}T/m$ \[120\], where $k_{B}$ is Boltzmann’s constant and $T$ the temperature”\[A\]. The collision operator is then defined by

$$C[v_{ic}] = v_{i}^{\text{ran}} - \frac{1}{N_{c}} \sum_{j=1}^{N_{c}} v_{j}^{\text{ran}}.$$ (4.7)

It can easily be seen that the collision rules conserve mass and linear momentum, which gives rise to dynamics governed by the Navier-Stokes equations on large length and time scales. The collision operator of MPC-SRD additionally conserves energy, since

$$\sum_{i=1}^{N_{c}} v_{i}^{2} = \sum_{i=1}^{N_{c}} (v_{cm}^{2} + R(\alpha)v_{ic}^{2}) = \sum_{i=1}^{N_{c}} (v_{cm}^{2} + 2v_{cm} \cdot R(\alpha)v_{ic} + v_{ic}^{2})$$

$$= \sum_{i=1}^{N_{c}} (v_{cm}^{2} + v_{i}^{2} - 2v_{cm} \cdot v_{i} + v_{cm}^{2}) = \sum_{i=1}^{N_{c}} v_{i}^{2},$$ (4.8)

$$= \sum_{i=1}^{N_{c}} (v_{cm}^{2} + v_{i}^{2}) = \sum_{i=1}^{N_{c}} (v_{cm}^{2} + v_{i}^{2}) = \sum_{i=1}^{N_{c}} v_{i}^{2},$$ (4.9)

where we used the orthogonality of $R(\alpha)$ and the definition of $v_{cm}$. Hence, MPC-SRD and MPC-AT correspond to a microcanonical and a canonical ensemble, respectively.

**Thermostat** By means of the Maxwell-Boltzmann scaling approach (MBS)[42], a canonical ensemble is obtained for MPC-SRD, if desired. The MBS thermostat acts on the cell level. After each time step, the relative velocities $v_{ic}$ of all MPC-particles in a cell are rescaled, i.e.\[a\],

$$v_{i} \leftarrow v_{cm} + \kappa v_{ic},$$ (4.10)

with the scaling factor

$$\kappa = \sqrt{\frac{2E_{k}}{\sum_{i=1}^{N_{c}} m v_{ic}^{2}}}.$$ (4.11)

Here, the kinetic energy $E_{k}$ is a random number, drawn from a Gamma distribution

$$P(E_{k}) = \frac{1}{E_{k}\Gamma(f/2)} \left( \frac{E_{k}}{k_{B}T} \right)^{f/2} \exp \left( -\frac{E_{k}}{k_{B}T} \right),$$ (4.12)

\[a\]Here, “$\leftarrow$” denotes the assignment operator. $x \leftarrow y$ means that the value of $y$ is assigned to the variable $x$. 

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where $\Gamma(x)$ is the gamma function and $f = 3(N_c - 1)$ is the number of degrees of freedom. Note that the relative velocities $v_{i}$ are considered in eqs. (4.10) and (4.11), since the transformation to another inertial frame of reference changes the velocities $v_i$, but not the relative velocities and not the temperature of the fluid. The distribution function of kinetic energies (4.12) follows from the expression [42]

$$P(E_k) = \frac{1}{Z} \int d^{N_c} v_{ic} \delta \left( E_k - \frac{m}{2} \sum_{i=1}^{N_c} v_{ic}^2 \right) \delta \left( \sum_{i=1}^{N_c} v_{ic} \right) \exp \left( -\frac{m}{2k_B T} \sum_{i=1}^{N_c} v_{ic}^2 \right),$$

(4.13)

with the partition function $Z = \int d^{N_c} v_{ic} \delta \left( \sum_{i=1}^{N_c} v_{ic} \right) \exp \left( -\frac{m}{2k_B T} \sum_{i=1}^{N_c} v_{ic}^2 \right)$. Here, we used the probability distribution function of velocities in a canonical ensemble. It has been shown that the MBS thermostat yields the correct hydrodynamic correlations of an isothermal fluid [42, 159], as long as the time step $\Delta t$ is not larger than $0.1 \sqrt{k_B T/(ma^2)}$. For large $\Delta t$ a considerable energy transfer during a streaming step occurs, which destroys isothermality.

Throughout the thesis, we will exclusively work within the canonical ensemble.

**Angular momentum conserving collisions** The collision step in eq. (4.2) does not conserve angular momentum, which can result in unphysical behavior (see chapter 6). To conserve angular momentum locally, we modify the collision, such that all particles in a cell additionally gain a rigid body angular velocity $\omega$ according to [47, 121]

$$v'_{i} = v_{cm} + \mathcal{C}[v_{ic}] + \omega \times r_{ic},$$

(4.14)

where $r_{ic} = r_i - r_{cm}$ and $r_{cm} = (1/N_c) \sum_{i=1}^{N_c} r_i$ is the center-of-mass position of the particles in a collision cell. The requirement of angular momentum conservation (AMC) determines the angular velocity $\omega$ to be

$$\omega = I^{-1} \sum_{j=1}^{N_c} m \left\{ r_{jc} \times (v_{jc} - \mathcal{C}[v_{jc}]) \right\}. \quad (4.15)$$

Here, $I$ is the moment of inertia tensor (see eq. (1.38)) of the respective particles in the center-of-mass reference frame,

$$I_{\alpha\beta} = \sum_{j=1}^{N_c} m \left\{ r_{j,c}^2 \delta_{\alpha,\beta} - (r_{j,c})_{\alpha} (r_{j,c})_{\beta} \right\}. \quad (4.16)$$

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Chapter 4 Multiparticle collision dynamics (MPC)

We can easily confirm that the choice of $\omega$ in eq. (4.15) yields AMC. The change in angular momentum of a cell in a collision step is

$$\Delta L = \sum_{i=1}^{N_c} mr_{ic} \times (v_i' - v_i) = \sum_{i=1}^{N_c} mr_{ic} \times (-v_{ic} + C v_{ic} + \omega \times r_{ic})$$

(4.17)

$$= \sum_{i=1}^{N_c} mr_{ic} \times [(C-1)v_{ic}] - \sum_{i=1}^{N_c} mr_{ic} \times (r_{ic} \times \omega)$$

(4.18)

$$= \sum_{i=1}^{N_c} mr_{ic} \times [(C-1)v_{ic}] + I \omega = 0,$$

(4.19)

where we used the identity $a \times (b \times c) = b (a \cdot c) - c (a \cdot b)$. Note that for angular momentum conserving MPC (MPC+a) the collision step is only performed for cells containing at least three particles, since the moment of inertia tensor for two particles is not invertible. Since $I$ is three dimensional, its inverse can be easily computed using an analytical formula. For a $3 \times 3$ matrix $M = (v_1, v_2, v_3)$ consisting of three column vectors, the matrix inverse is given by

$$M^{-1} = \frac{1}{v_1 \cdot (v_2 \times v_3)} \begin{pmatrix} (v_2 \times v_3)^T \\ (v_3 \times v_1)^T \\ (v_1 \times v_2)^T \end{pmatrix}.$$  

(4.20)

Equation (4.14) violates energy conservation for MPC-SRD. For isothermal systems, to which we restrict ourselves here, this is not problematic and the MBS thermostat is applied after each collision. If angular momentum and energy conservation are desired for MPC-SRD, a rescaling of the velocities after a collision as described in ref. [47], can be performed. We denote the angular momentum conserving versions by MPC-SRD+a and MPC-AT+a, respectively. The addition of angular momentum conservation typically yields an increase in computation time by a factor of roughly two.

**Random shift** Partition of the system into collision cells leads to a violation of Galilean invariance. In order to reestablish Galilean invariance, a random shift of the collision lattice is introduced at every collision step [36, 160]. An equivalent point of view is that the particle positions are shifted for sorting them into collision cells, i.e., the positions for particle sorting are given by $r_i + s$, where the Cartesian components $s_\alpha$ ($\alpha \in \{x, y, z\}$) of the shift vector $s$ are taken from a uniform distribution in the interval $[-a/2, a/2]$. Note, that appropriate boundary conditions (e.g. periodic or Lees-Edwards boundary conditions, see section 4.3) have to applied to the shifted particle positions. If the particle positions stay fixed and the lattice is shifted during the sorting into collision cells, some floating point operations can be saved. However, when calculating correlation functions of fluid properties, consistent results can only be found when the particles are shifted and the lattice is kept fixed.
4.3 Boundary conditions

Units and independent parameters When we implement the MPC algorithm, we choose $m = k_B T = a = 1$. Hence, lengths are measured in units of the collision cell length $a$, masses are measured in units of the fluid particle mass $m$, and times are measured in units of $\sqrt{ma^2/(k_B T)}$. Note that all other mechanical quantities can be expressed in terms of length, mass, and time. Viscosity, for example, is measured in units of $\sqrt{ma^2/(k_B T)}$ and pressure in units of $k_B T/a^3$.

The parameters which define the properties of the MPC fluid, are the average number of particles per cell $\bar{N}_c \equiv \langle N_c \rangle$, the time step $h$, and in case of MPC-SRD the rotation angle $\alpha$. Note however that, as mentioned before, we will always choose $\alpha = 130^\circ$ to obtain a fluid-like behavior (see ref. [158]). A typical value for the average number of particles is $\bar{N}_c = 10$, while the time step is typically chosen as $h \lesssim 0.1 \sqrt{ma^2/(k_B T)}$. These values can be tuned, e.g., to modify viscosity (see section 4.5). However, to guarantee isothermality, the time step should not be much larger than $0.1 \sqrt{ma^2/(k_B T)}$.

4.3 Boundary conditions

4.3.1 Periodic and Lees-Edwards boundary conditions

To model a bulk fluid, we apply periodic boundary conditions. This means, that a particle leaving the box due to streaming, or due to a random shift, will directly re-enter the box on the other side (see algorithm 1). An interpretation of this procedure is that there are infinitely many copies of the simulation box in all Cartesian directions (see fig. 4.2 (a)).

To implement shear flow $v_\alpha(r) = \delta_\alpha x \dot{\gamma} r_z$, Lees-Edwards boundary conditions [157] are applied, which are illustrated in fig. 4.2 (b) and algorithm 2. The key idea is to let the copies of the simulation box move with assigned velocities. The row of copies containing the main simulation box is resting, while the row above moves with velocity $\dot{\gamma} L_y$ and the one above that with velocity $2\dot{\gamma} L_y$ and so forth. Correspondingly the rows below move with velocities $-\dot{\gamma} L_y$, $-2\dot{\gamma} L_y$ and so forth. The displacement between rows is denoted as $\delta x$ and it is updated at each time step according to

$$\delta x(t + h) = \delta x(t) + h\dot{\gamma} L_y$$

and periodically wrapped into the interval $[-L_x/2, L_x/2)$. After $\delta x$ is updated, the MPC particles are streamed and algorithm 2 is applied to them. For the subsequent collision step, the following procedure is applied:

- Shift all particles: $r_i \leftarrow r_i + s$
- Apply Lees-Edwards (or periodic) boundary conditions to all particles
- Perform the collision step (and apply thermostat if desired)
Figure 4.2: Sketch of different boundary conditions. A particle is represented by a black point before streaming, and as a gray point after streaming. (a) For periodic boundary conditions the main simulation box (blue) has infinitely many copies in all Cartesian directions. (b) For Lees-Edwards boundary conditions each row of simulation box copies is moving in $x$-direction with velocity $0, \pm \dot{\gamma}L_y, \pm 2\dot{\gamma}L_y, \ldots$, depending on their position. $\dot{\gamma}$ is the shear rate, $L_\alpha$ the simulation box length in $\alpha$ direction and $\delta x(t+h) = \delta x(t) + \dot{\gamma}L_y$, with $\delta x \in [-L_x/2, L_x/2)$.

```
// Apply periodic boundary conditions to position vector r
for $\alpha \in \{x, y, z\}$ do  // for each Cartesian component
    if $r_\alpha \geq L_\alpha$ then
        $r_\alpha = r_\alpha - L_\alpha$
    if $r_\alpha < 0$ then
        $r_\alpha = r_\alpha + L_\alpha$

Algorithm 1: Periodic boundary conditions
```

```
// Apply Lees-Edwards boundary conditions to position vector r and velocity v
$c_y = 0$
if $r_y \geq L_y$ then
    $c_y = 1$
else if $r_y < 0$ then
    $c_y = -1$
$r_x = r_x - c_y \delta x$
PBC(r) // Apply periodic boundary conditions
$v_x = v_x - c_y \dot{\gamma}L_y$

Algorithm 2: Lees-Edwards boundary conditions
4.3 Boundary conditions

- Shift all particles back: $r_i \leftarrow r_i - s$
- Apply Lees-Edwards (or periodic) boundary conditions to all particles

We mention once again, that some floating point operations can be saved by shifting the lattice instead of the particles. For $\delta x = 0$ and $\dot{\gamma} = 0$, Lees-Edwards boundary conditions are identical to periodic boundary conditions.

4.3.2 Slip and no-slip boundary conditions for immersed objects and boundaries

Consider an immersed rigid body, which can be a colloid or a squirmer, moving in an isothermal MPC fluid. Additionally the fluid can be confined, e.g., by fixed parallel walls, or by a rotating hollow cylinder of infinite length (see chapter 6). In this section, we will show how to couple such immersed objects or boundaries to the MPC algorithm. The notation of chapter 1 is applied. In particular, the immersed body has a center position $C$, linear velocity $U$, and angular velocity $\Omega$. We consider either slip or a no-slip boundary conditions on the object’s surface. For a realization of partial slip boundary conditions in MPC, see refs. [126, 151]. Inside an object there are no fluid particles. Hence, when the MPC fluid is initialized, MPC particles are only distributed outside of the immersed objects. In the following, we will present the coupling of an immersed body during the streaming and collision step. At the end of this section, we will briefly discuss the implementation of a boundary. Note that for a proper resolution an object should extend over several MPC collision cells. A spherical colloid, for example, should have a radius of at least 3 collision cells (see chapter 7).

Streaming step

In the streaming step, the immersed body moves for the time step $h$ according to its equations of motion. However, we have to guarantee that no fluid particle ends up inside the body. For no-slip boundary conditions (see section 2.2.4), a fluid particle reverses its velocity, i.e. $v_i \leftarrow -v_i$, whenever it hits the surface of the body. This is known as the bounce-back rule (see fig. 4.3). For slip boundary conditions, only the velocity in the direction of the surface normal $n$ is reversed, which corresponds to a specular reflection (see fig. 4.3). If the body is fixed in space, no further steps have to be taken.

If the body is not fixed, the momentum and angular momentum lost by a colliding MPC particle is transferred to the body. This changes the body’s velocity and angular velocity. In principle, we would have to determine which particle first collides with the body and resolve this collision. Subsequently, given the body’s new (angular) velocity, we would compute the next collision and so on, until the time $h$ has passed. This corresponds to a so called event-driven simulation, which
is computationally demanding, especially when the body is nonspherical, or several bodies are immersed in the fluid. A coarse grained approach is applicable for a short streaming interval, e.g., \( h \lesssim 0.05 \sqrt{ma^2/(k_B T)} \) for bodies with linear dimension of 2-3 cell lengths or more [161]. Since only a few particles collide with the body during the short time interval \( h \), and since the mass of the body is much larger than a fluid particle’s mass, the body’s velocity and angular velocity do not change significantly. Hence, we can assume the body to move undisturbed by collisions with MPC particles during one streaming interval. Under this assumption, we can just build a list of particles which end up inside the body after the streaming interval and resolve the collisions of these particles independently, and hence, in parallel. At the end of the streaming step, when all particle collisions have been treated, we transfer the sum of all (angular) momentum transfers to the body. This ensures total (angular) momentum conservation, which is essential for a proper hydrodynamic coupling.

Only for certain body shapes, such as spherical shapes, we can easily find analytical expressions for the time at which the collision between a MPC particle and the body occurs, even under the assumption of undisturbed body motion. However, independent of shape, this time will be \( t + h/2 \) on average. Thus, we perform all collisions at time \( t + h/2 \), which is justified, as long as the body’s dimension is large compared to the distance \( |v_i h| \) a particle travels during the streaming step [161].

Our considerations are summarized in the following algorithm [148, 161]:

(i) Stream the MPC particles and let the body move according to its equations of motion for the time \( h \).

(ii) Build a list of all particles that ended up inside the body.

(iii) Move the body and each particle \( i \) in that list back in time by half a time step, i.e., by \( h/2 \). Change the position of each particle \( i \) to its projection onto

---

**Figure 4.3:** Realization of boundary conditions during the streaming step. A fluid particle’s trajectory before (after) the collision is visualized by a solid black (dashed green) line. The no-slip boundary condition is realized by a bounce-back, while the slip boundary condition is realized by a specular reflection.
4.3 Boundary conditions

the body’s surface. If the body is a sphere of radius \( R \) this corresponds to \( r_i \leftarrow C + R(r_i - C)/|r_i - C| \).

(iv) Resolve the collision between each particle \( i \) and the body according to a bounce-back or a specular reflection. The velocity update is

\[
v_i \leftarrow v_i - J_i / m, \tag{4.22}
\]

where the momentum transfer \( J_i \) is either given by

\[
J_i = 2m \left( v_i - U - \Omega \times (r_i - C) \right), \tag{4.23}
\]

for the bounce-back rule, which corresponds to a no-slip boundary condition, or by

\[
J_i = 2m n_i^T \left( v_i - U - \Omega \times (r_i - C) \right), \tag{4.24}
\]

for the specular reflection, which corresponds to a slip boundary condition. Here, the surface normal vector at point \( r_i \) is denoted by \( n_i \). If the immersed body is a squirmer, we perform a bounce-back that takes the squirming velocity into account. The momentum transfer is then given by [90, 148]

\[
J_i = 2m \left( v_i - U - \Omega \times (r_i - C) - D^T u^b_{sq} (D(r_i - C)) \right), \tag{4.25}
\]

where \( u^b_{sq}(r) \) is the squirming velocity (see section 2.5.4), the superscript \( b \) stands for the body-fixed frame (see section 1.2), and \( D \) is the rotation matrix that transforms vectors into the body-fixed frame (see section 1.2). The momentum transfer \( J_i \) and angular momentum transfer \( L_i \) are not directly applied to the body, but rather added to the summation variables \( J \) and \( L \).

(v) The particles are propagated to the time \( t + h \), i.e, they stream for the remaining part of the time step with their new velocities.

(vi) After all collisions have been resolved, the body is propagated to the time \( t + h \) again and subsequently changes its (angular) velocity by

\[
U \leftarrow U + J / M, \tag{4.26}
\]
\[
\Omega \leftarrow \Omega + I^{-1} L, \tag{4.27}
\]

where \( M \) and \( I \) are the mass and moment of inertia of the body, respectively. Finally, set \( J \) and \( L \) to zero.

When exact collision times can be calculated analytically, we can perform the alternative step (iii-a) instead of (iii):
(iii-a) For each particle $i$ in that list, determine the time $t + h_i$ at which it penetrated the body, and move the particle to that point in time. The following collision (step (iv)) is resolved between the particle and a virtual body, which has the same variables, i.e., position, orientation and (angular) velocity, that the real body had at time $t + h_i$.

**Collision step**

In case of slip boundary conditions, no additional measure is taken in the collision step. For no-slip boundary conditions, $N_g$ ghost particles are inserted into the body before the collision step (see fig. 4.4) \[129\]. These ghost particles have the same number density and mass as the fluid particles. Hence, $N_g = \lfloor \rho V/m \rfloor$, where $V$ is the volume of the immersed body and $\lfloor \ldots \rfloor$ is the floor function. A uniform distribution of ghost particles is achieved by enclosing the body with a cuboid, inside which uniformly distributed positions are generated, which are only accepted if they lie inside the body. Ghost particles do not take part in the streaming step. Before each collision step, their positions and velocities are randomized. The positions $\mathbf{r}_i^g$ are uniformly distributed inside the body and the velocities are equal to the mean rigid body velocity plus a random velocity

$$
\mathbf{v}_i^g = \mathbf{U} + \Omega \times (\mathbf{r}_i^g - \mathbf{C}) + \mathbf{v}_{i\text{ran}}.
$$

(4.28)
Here, $v_{i}^{ran}$ is a random velocity with Gaussian distributed Cartesian components of zero mean and variance $k_{B}T/m$, which guarantees a correct thermal coupling. For a squirmer, the ghost particle velocities are

$$v_{i}^{g} = U + \Omega \times (r_{i} - C) + v_{i}^{ran} + D^{T}u_{sq}^{b}[PD(r_{i} - C)].$$  
(4.29)

Since the position $D(r_{i} - C)$ is inside the body, it is projected onto the squirmer’s surface before evaluation of the prescribed surface velocity $u_{sq}$, which is indicated by an operator $P$ in eq. (4.29).

Subsequently, the ghost particles take part in the collision step just like the fluid particles. This includes the random shift and the application of periodic boundary conditions. If the body is not fixed in space, the (angular) momentum gained by the ghost particles during the collision step is now given to the body. Denoting ghost particle velocities after the collision by $v_{i}'$ and before the collision by $v_{i}$, the (angular) momentum transfer is

$$J = \sum_{i} m (v_{i}' - v_{i}),$$  
(4.30)

$$L = \sum_{i} (r_{i} - C) \times m (v_{i}' - v_{i}).$$  
(4.31)

We transfer this (angular) momentum gain to the body according to

$$U \leftarrow U + J/M,$$  
(4.32)

$$\Omega \leftarrow \Omega + I^{-1}L.$$  
(4.33)

**Remarks regarding an energy conserving simulation**

The bounce-back rule and the specular reflection do not conserve energy. For simple body shapes, such as spheres, it is possible to construct similar rules, which do conserve energy. For a spherical colloid, we define the relative velocity as

$$\bar{v} = v - U - \Omega \times (r_{i} - C),$$  
(4.34)

where $C$ is the colloid’s center. The equations for conservation of momentum, angular momentum and energy yield two solutions for $J_{i}$, [126, 162]

$$J_{i} = 2\mu \bar{v}_{i,n} + 2(1 - \Gamma)\mu \frac{M\chi}{\mu + M\chi} \bar{v}_{i,t}. $$  
(4.35)

Here $\chi = 2/5$, $\mu = mM/(m + M)$ is the reduced mass, $\Gamma = 0, 1$ distinguishes the two solutions, and the indices $n$ and $t$ denote the normal and tangential component of $\bar{v}$, i.e. $\bar{v}_{n} = n n^{T} \bar{v}$ and $\bar{v}_{t} = (1 - n n^{T}) \bar{v}$. Since $M \gg m$, the choice $\Gamma = 1$ corresponds to slip and $\Gamma = 0$ to no-slip boundary conditions.
Note, however, that in case of no-slip boundary conditions, where ghost particles are necessary, the conservation of energy is violated in the collision step. This is due to the Maxwell-Boltzmann-distributed ghost particle velocities. Hence, spherical colloids with slip boundary conditions are typically considered in energy conserving MPC simulations, because they do not need ghost particles.

**Boundaries**

The implementation of boundaries is analogous to that of immersed objects. However, they are assumed to be of sufficient mass, such that the momentum transfer of MPC fluid particles is not transferred to the boundary. We briefly discuss the streaming and collision step for parallel bounding walls and for a bounding hollow cylinder.

In the streaming step, exact collision times can be calculated and method (iii-a) is applied. For a wall, the momentum transfer of a colliding fluid particle is $J_i = 2m(v_i - Ue_x)$ for no-slip and $J_i = 2mnT(v_i - Ue_x)$ for slip boundary conditions. Here, the wall’s normal vector is $n = e_y$ and it is moving with velocity $U$ in $x$-direction. In this thesis, however, we will only consider stationary walls, i.e., $U = 0$. For a bounding hollow cylinder, we choose its center as origin, such that the momentum transfer of a colliding MPC particle is $J_i = 2m(v_i - \Omega \times r_i)$ for no-slip and $J_i = 2mnT(v_i - \Omega \times r_i)$ for slip boundary conditions. Here, $\Omega = \Omega e_z$ is the angular velocity of the bounding cylinder and the normal vector $n$ points into the fluid.

For the collision step, ghost particles are only distributed in a layer behind the bounding surface. The width of that layer (cyan in fig. 4.4) is chosen large enough, such that fluid particles will not interact with the empty space behind the layer in the collision step. Hence for parallel walls, the layer width is $a$, i.e., one MPC cell length, and for a bounding cylinder the layer width is $\sqrt{2}a$. The ghost particle velocities are $v_i^g = Ue_x + v_{i^{ran}}$ for a wall, and $v_i^g = \Omega \times r_i^g + v_{i^{ran}}$ for a hollow cylinder.

**4.3.3 Coupling of point particles to MPC**

In many applications, objects build from point particles, like polymers, are immersed in the MPC fluid. The point particles move according to their equations of motion in the streaming step and are treated just like MPC fluid particles in the collision step to achieve coupling with the MPC fluid. However, care has to be taken, since their mass is unequal to the fluid particle mass in general. Hence, the collision step is slightly modified. When calculating the center-of-mass position and center-of-mass velocity, the different masses $m_j$ of particles have to be weighted, i.e., $r_{cm} = (\sum_i m_i r_i) / (\sum_j m_j)$ and $v_{cm} = (\sum_i m_i v_i) / (\sum_j m_j)$. The MPC-AT collision operator
4.4 Stress tensor

(see eq. (4.7)) changes to

$$C[v_{ic}] = v_{i}^{ran} - \frac{1}{\sum_{k=1}^{N_c} m_k} \sum_{j=1}^{N_c} m_j v_j^{ran},$$

(4.36)

and finally \(m\) has to be replaced by \(m_j\) in eqs. (4.15) and (4.16). Typically, the mass of immersed point particles is chosen as \(N_c m\) to yield a good coupling.

In ref. [40] it has been shown that a spherical colloid, represented by a discretized mesh of point particles on its surface, shows the same hydrodynamic correlations as a hard spherical colloid with no-slip boundary conditions, although fluid particles are allowed to enter the discretized colloid.

We discussed the coupling to objects build from point particles here, since it is widely applied. However, we will not consider such objects throughout this thesis.

4.4 Stress tensor

4.4.1 Particle level stress tensor

We consider a fixed control volume \(V\) at position \(r\) inside the fluid, which is so small that macroscopic gradients can be considered negligible, however large enough to contain many particles. We define the quantity [163]

$$\dot{G}_{\alpha\beta} = \left\langle \frac{1}{V} \frac{d}{dt} \sum_{i,r_i \in V} m_i v_{i\alpha} r_{i\beta} \right\rangle,$$

(4.37)

where the average \(\langle \ldots \rangle\) can correspond to a time average or an ensemble average respectively. Performing the time average we find

$$\dot{G}_{\alpha\beta} = \frac{1}{V} \lim_{T \to \infty} \frac{1}{T} \int_0^T \frac{d}{dt} \sum_{i,r_i \in V} m_i v_{i\alpha} r_{i\beta} = \frac{1}{V} \lim_{T \to \infty} \frac{1}{T} \left( \sum_{i,r_i \in V} m_i v_{i\alpha} r_{i\beta} \right) \bigg|_0^T = 0.$$

(4.38)

In the last equality we used that the expression \((\sum_{i,r_i \in V} m_i v_{i\alpha} r_{i\beta})\bigg|_0^T\) is bounded. We explicitly differentiate in eq. (4.37) to find [163]

$$\dot{G}_{\alpha\beta} = \left\langle \frac{1}{V} \sum_{i,r_i \in V} m_i v_{i\alpha} v_{i\beta} \right\rangle + \left\langle \frac{1}{V} \sum_{i,r_i \in V} r_{i\beta} m_i \frac{dv_{i\alpha}}{dt} \right\rangle.$$

(4.39)

For simplicity we assume that no body forces are exerted on the fluid. Hence, the change of \(v_{i\alpha}\) has an internal contribution due to all fluid particles in \(V\) and an
Chapter 4 Multiparticle collision dynamics (MPC)

external contribution resulting from the fluid particles outside of $V$

$$\frac{dv_{i\alpha}}{dt} = \frac{dv_{i\alpha}}{dt} \bigg|_{int} + \frac{dv_{i\alpha}}{dt} \bigg|_{ext}. \quad (4.40)$$

We evaluate the external term first. As we know from continuum theory, the momentum density $\rho v_\alpha$ in a control volume changes due to inflow of fluid carrying momentum, i.e., $-\int dA \rho v_\alpha v_\gamma n_\gamma$, and due to the stresses applied on the surface of $V$, i.e., $\int dA \sigma_{\alpha\gamma} n_\gamma$. Hence [163],

$$\left\langle \frac{1}{V} \sum_{i,r_i \in V} m_i \frac{dv_{i\alpha}}{dt} \bigg|_{ext} \right\rangle = \frac{1}{V} \int_{\partial V} dA r_\beta \left[ \sigma_{\alpha\gamma} - \rho v_\alpha v_\gamma \right] n_\gamma$$

$$= \frac{1}{V} \int dV \frac{\partial}{\partial r_\gamma} \left\{ r_\beta \left[ \sigma_{\alpha\gamma} - \rho v_\alpha v_\gamma \right] \right\}$$

$$= \sigma_{\alpha\beta} - \rho v_\alpha v_\beta$$

$$= \sigma_{\alpha\beta} - \frac{1}{V} \left\langle \sum_{i,r_i \in V} m_i v_\alpha(r_i) v_\beta(r_i) \right\rangle. \quad (4.41)$$

Here, we applied Gauss’ theorem for the second equality and assumed that gradients in $\rho$, $v$, and $\sigma$ are negligible inside $V$ for the third equality. Substituting eqs. (4.38) and (4.41) into eq. (4.39) yields [163]

$$\sigma_{\alpha\beta}(r,t) = -\left\langle \frac{1}{V} \sum_{i,r_i \in V} m_i (v_{i\alpha} - v_\alpha(r_i))(v_{i\beta} - v_\beta(r_i)) \right\rangle$$

$$- \left\langle \frac{1}{V} \sum_{i,r_i \in V} r_{i\beta} m_i \frac{dv_{i\alpha}}{dt} \bigg|_{int} \right\rangle. \quad (4.42)$$

We can read the equation as follows. In a fluid consisting of point particles, the stress tensor at position $r$ is given by the right hand side of eq. (4.42), where the volume $V$ around $r$ is chosen such that many particles are inside, but the gradients of macroscopic quantities are negligible.

We can now apply the general expression (4.42) to the MPC fluid. Thereby we regard MPC as a discrete time random process. Consequently, the stress tensor is defined at times $t = jh$, with $j \in \mathbb{N}$. The internal change of velocity $dv/dt|_{int}$ is due to the change of velocity in the collision step, i.e.,

$$m \frac{dv_{i\alpha}}{dt} \bigg|_{int} \rightarrow \frac{\Delta p_{i\alpha}}{h}, \quad (4.43)$$

where $p_i = mv_i$. With this mapping, we can define the instantaneous particle level stress tensor $\sigma^i$ of MPC as

$$\sigma^i_{\alpha\beta}(r,t) = -\frac{1}{V} \sum_{i,r_i \in V} m(v_{i\alpha} - v_\alpha(r_i))(v_{i\beta} - v_\beta(r_i)) - \frac{1}{Vh} \sum_{i,r_i \in V} \Delta p_{i\alpha} r_{i\beta}. \quad (4.44)$$
Note that the momentum change $\Delta p_{i\alpha}$ is only nonzero at times $t = lh$, with $l \in \mathbb{Z}_{>0}$. For further analytical treatment, we note that we can replace $r_i$ with $\Delta r_i = r_i - r_c$ in the collisional term, where $r_c$ is the center of the cell that contains particle $i$. This is due to the fact that the sum of $\Delta p_{i\alpha}$ over all particles $i$ in one cell is zero due to momentum conservation. Furthermore, for systems in which the stress does not depend on position, e.g. in equilibrium or in shear flow, we can sum over all particles, yielding

$$
\sigma_{\alpha\beta}^{i}(t) = -\frac{1}{V} \sum_{i=1}^{N} m (v_{i\alpha} - v_{\alpha}(r_i))(v_{i\beta} - v_{\beta}(r_i)) - \frac{1}{Vh} \sum_{i=1}^{N} \Delta p_{i\alpha} \Delta r_{i\beta} \quad (4.45)
$$

$$
\equiv \sigma_{\alpha\beta}^{i,k} + \sigma_{\alpha\beta}^{i,c}, \quad (4.46)
$$

where we introduced the kinetic and collisional part of the stress tensor.

### 4.4.2 Continuum stress tensor

Only for MPC+a, the continuum stress tensor has the well known form of eq. (2.9) with two viscosity parameters. For MPC-a, the more general form introduced in eqs. (2.5) and (2.8) with three viscosity parameters has to be considered

$$
\sigma = -p \mathbf{1} + \eta_1 \nabla v^T + \eta_2 (\nabla v^T)^T + \eta_3 (\nabla \cdot v) \mathbf{1}. \quad (4.47)
$$

We will establish the parameters $\eta_1, \eta_2, \eta_3$ in the next chapter. With the stress tensor (4.47), the Navier-Stokes equation (2.20) becomes

$$
\rho \left( \frac{\partial}{\partial t} v + v \cdot \nabla v \right) = -\nabla p + \eta_2 \Delta v + (\eta_1 + \eta_3) \nabla (\nabla \cdot v). \quad (4.48)
$$

We expect this continuum equation to hold for the MPC fluid on large length and long time scales, since mass and momentum are conserved in the MPC algorithm.

For low (oscillatory) Reynolds numbers, we can neglect the left-hand side (see section 2.3). If the density does not vary strongly, we can additionally assume $\nabla \cdot v = 0$ and the MPC fluid is described by Stokes equations

$$
0 = -\nabla p + \eta_2 \Delta v, \quad \nabla \cdot v = 0. \quad (4.49)
$$

### 4.5 Transport coefficients

It is of interest to know the viscosity parameters $\eta_1, \eta_2, \eta_3$ of the MPC fluid in terms of density $\rho$, time step $h$, and – in case of SRD – rotation angle $\alpha$. Additionally the diffusion coefficient $D$ of a MPC fluid particle is of interest. Using the molecular chaos approximation, analytical formulae can be derived. Here, we list all transport
coefficients for MPC±a (see also ref. [47]). In chapter 5, where the bulk viscosity of MPC will be discussed, we will also show how to determine the viscosity parameters for MPC-a analytically.

The viscosity $\eta = \eta_2$ has a kinetic contribution $\eta^k$ due to the streaming step and a collisional contribution $\eta^c$ due to the collision step. As we will show in chapter 5,

$$\eta_1 = \eta^k, \quad \eta_2 = \eta^k + \eta^c, \quad \eta_3 = -2\eta/k/3, \quad (4.50)$$

for MPC-a, whereas

$$\eta_1 = \eta = \eta^k + \eta^c, \quad \eta_2 = \eta = \eta^k + \eta^c, \quad \eta_3 = -2\eta/V + \eta^V \quad (4.51)$$

for MPC+a. Thereby $\eta^k$ and $\eta^c$ are different for MPC-a and MPC+a. However, the bulk viscosity $\eta^V = (\eta_1 + \eta_2 + 3\eta_3)/3$ is equal for MPC+a and MPC-a as we will show in chapter 5.

For MPC-a in $d = 2, 3$ dimensions, the kinetic and collisional viscosity as well as the diffusion coefficient are

$$\eta^k = \bar{N}_c k_B T h \left( \frac{\bar{N}_c/B}{N_c - 1 + e^{-N_c}} - \frac{1}{2} \right), \quad (4.52)$$

$$\eta^c = \frac{Am(\bar{N}_c - 1 + e^{-N_c})}{12a^d - 2h}, \quad (4.53)$$

$$D = \frac{k_B T h}{m} \left( \frac{\bar{N}_c/A}{\bar{N}_c - 1 + e^{-N_c}} - \frac{1}{2} \right), \quad (4.54)$$

where $A$ and $B$ are given in table 4.1 for MPC-AT and MPC-SRD.

For MPC+a in 3 dimensions, the transport coefficients are given by

$$\eta^k = \frac{\bar{N}_c k_B T h}{a^3} \left( \frac{1}{c_m} - \frac{1}{2} \right), \quad \text{where}$$

$$c_m = B(1 - e^{-N_c}(1 + \bar{N}_c)) + \left( 3A - \frac{11}{2}B \right) \frac{1 - e^{-N_c}(1 + \bar{N}_c + \bar{N}_c^2/2)}{2N_c}, \quad (4.55)$$

$$\eta^c = \frac{Am}{24a^d h} \left( \bar{N}_c - \frac{7}{5} + e^{-N_c} \left( \frac{7}{5} + \frac{2\bar{N}_c}{5} - \frac{3\bar{N}_c^2}{10} \right) \right), \quad (4.56)$$

$$D = \frac{k_B T h}{m} \left( \frac{\bar{N}_c/A}{\bar{N}_c - 2} - \frac{1}{2} \right). \quad (4.57)$$

The analytical expressions are a very good approximation for MPC-a, but rather imprecise for MPC+a (see chapter 5). Note that as we are restricted to small time steps $h$ in order to guarantee isothermality [159], the viscosity will be dominated by the collisional contribution which scales as $1/h$. As mentioned before, the free parameters are the mean number of particles per cell $\bar{N}_c$ and the collision time step $h$. To increase viscosity we can decrease the time step ($\eta \approx \eta^c \propto 1/h$) or increase the mean number of particles ($\eta \propto \bar{N}_c$). However, the computational effort scales linearly with $\bar{N}_c$ and $1/h$ as well.
### 4.5 Transport coefficients

<table>
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<th>A</th>
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</tr>
</thead>
<tbody>
<tr>
<td>SRD (d = 2)</td>
<td>(1 - \cos(\alpha))</td>
<td>(1 - \cos(2\alpha))</td>
</tr>
<tr>
<td>SRD (d = 3)</td>
<td>(\frac{2}{3}(1 - \cos(\alpha)))</td>
<td>(\frac{2}{5}(2 - \cos(\alpha) - \cos(2\alpha)))</td>
</tr>
<tr>
<td>AT</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

Table 4.1: Coefficients \(A\) and \(B\) for MPC-SRD and MPC-AT. \(d\) denotes for the number of dimensions.
Part II

Studies on MPC
Chapter 5

Bulk viscosity of multiparticle collision dynamics fluids

The results of this chapter have been published in publication [A] (see List of publications on page 203). All verbatim quotes within this chapter are quotations of [A] and are indicated as “…”[A]. Such verbatim quotes can extend over several pages and have been reformatted to fit with the main text. The label of a cited article inside a verbatim quote follows the enumeration in the bibliography (page 186).

5.1 Introduction

“The hydrodynamic properties of the MPC fluid manifest themselves in the stress tensor, which, for an isotropic system, has three viscosity parameters in general (see eq. (2.8)). For MPC, the shear viscosity has been analyzed theoretically and numerically [33, 34, 47, 119, 160, 164–167], and good agreement has been found between the theoretical expression and the numerically obtained values for a three-dimensional non-angular momentum conserving SRD fluid (MPC-SRD-a) [159]”[A]. However, no decisive results have been found for the bulk viscosity. The simulations of ref. [168] suggest a zero bulk viscosity for two-dimensional MPC fluids, while a nonzero bulk viscosity for MPC-SRD-a has been indicated in ref. [164] in connection with the stress tensor of ref. [166].

In this chapter, “we determine all relevant viscosity parameters of MPC fluids, and demonstrate that the bulk viscosity is nonzero for both angular momentum conserving and non-angular momentum conserving MPC variants. Moreover, we derive an analytical expression for the bulk viscosity and confirm it by simulations”[A].

5.2 Analytical calculation of viscosity parameters

Our aim is to find the coefficients $\eta_1, \eta_2, \eta_3$ of the hydrodynamic stress tensor (see eq. (4.47)) for the MPC fluid. Since the particle level stress tensor (4.45) is comprised of the kinetic and collisional parts $\sigma_{\alpha\beta}^{i,k}$ and $\sigma_{\alpha\beta}^{i,c}$, with a respective continuum
Chapter 5 Bulk viscosity of multiparticle collision dynamics fluids

representation, we can split the viscosities into kinetic parts $\eta_1^k$, $\eta_2^k$, and respective collisional parts $\eta_1^c$, $\eta_2^c$, and $\eta_3^c$. For an analytical treatment we will assume molecular chaos, which means that the velocities and positions of different particles are uncorrelated.

5.2.1 Relations between $\eta_1$ and $\eta_2$

Evidently, the kinetic stress tensor in eq. (4.45) is symmetric and therefore $\eta_1^k = \eta_2^k$. For the collisional stress, the symmetry requirement $\sigma_{\alpha\beta}^{i,c} = \sigma_{\beta\alpha}^{i,c}$ is equivalent to

$$0 = \sum_{\alpha\beta} \varepsilon_{\alpha\beta\gamma} \sigma_{\alpha\beta}^{i,c} = \sum_{i \in \text{cell}} (\Delta \mathbf{p}_i \times \mathbf{r}_i)_\gamma = -\sum_{i \in \text{cell}} \Delta L_{i\gamma},$$

(5.1)

where $\varepsilon_{\alpha\beta\gamma}$ is the Levi-Civita symbol. Hence, the collisional stress tensor is only symmetric, i.e., $\eta_1^c = \eta_2^c$, if the angular momentum $\mathbf{L}$ is conserved during collisions.

In order to determine $\eta_1^c$ for MPC-a, we consider a shear flow $v_\alpha(r) = \delta_{\alpha x} \dot{\gamma} r_z$ with the shear rate $\dot{\gamma}$; for which the continuum stress $\sigma_{\alpha\beta}$ can be easily computed by eq. (4.47). The only non-vanishing off-diagonal elements are

$$\sigma_{xz} = \eta_2^{\dot{\gamma}}, \quad \sigma_{zx} = \eta_1^{\dot{\gamma}}.$$

(5.2)

For the particle level stress tensor, we can perform a time average of eq. (4.45) as discussed in ref. [167], which yields

$$\sigma_{xz} = \langle \sigma_{xz}^i \rangle = -\left\langle \frac{1}{V} \sum_{i=1}^{N} m v_{ix} v_{iz} + \frac{\dot{\gamma} h}{2V} \sum_{i=1}^{N} m v_{iz}^2 + \frac{1}{Vh} \sum_{i=1}^{N} \Delta \mathbf{p}_{ix} \Delta \mathbf{r}_{iz} \right\rangle_T,$$

(5.3)

where $\langle \ldots \rangle_T$ denotes the average over time steps [167]. The velocities $\mathbf{v}_i$ are taken after the streaming step and in the primary box of the periodic system. Note that the sign of the second term changes, if velocities are taken before the streaming step [167]. "Exploiting the stress tensor (5.3), we can establish a relation between the stress, shear rate, and the MPC parameters. For the collisional stress we find

$$\langle \sigma_{i\beta}^{c,i} \rangle = -\frac{m}{Vh} \sum_i \langle \Delta v_{i\alpha} \Delta r_{i\beta} \rangle = -\frac{m}{Vh} \sum_i \langle C - 1 \rangle \langle v_{i\alpha} \Delta r_{i\beta} \rangle$$

$$= \frac{m}{Vh} \langle 1 - C \rangle \left( 1 - \frac{1}{N_c} \right) \sum_i \langle v_{i\alpha} \Delta r_{i\beta} \rangle$$

(5.4)

within the molecular chaos assumption, i.e., we set $\langle v_{j\alpha} \Delta r_{i\beta} \rangle = 0$ for $j \neq i$. Since $\langle v_{iz} \Delta r_{ix} \rangle = 0$ for MPC-a, we directly find $\sigma_{zx}^c = 0$ and hence $\eta_1^c = 0$ [48]. On the other hand $\langle v_{ix} \Delta r_{iz} \rangle = \dot{\gamma} \langle \Delta r_{ix}^2 \rangle = \dot{\gamma} a^2 / 12 > 0$, which enables us to calculate
η₂^k [167]. The result is equal to that obtained by Green Kubo relations, which are exploited in section 5.2.2[A]. In summary, we determined

\[ \eta_1^k = \eta_2^k, \]  
\[ \eta_1^c = \begin{cases} \eta_2^c, & \text{for MPC+a}, \\ 0, & \text{for MPC-a}. \end{cases} \]  

5.2.2 Green-Kubo relation for shear viscosity

"We will focus on MPC-a. Analytical results for the shear viscosity of MPC+a can be found in ref. [47]. The shear viscosity \( \eta = \eta_2 \) is obtained by the Green-Kubo relation [169]

\[ \eta = \frac{V}{k_B T} \int_0^\infty dt \langle \sigma_{xz}^i(t)\sigma_{xz}^i(0) \rangle \]  

from the stress tensor correlation function in equilibrium (\( n_i(\mathbf{r}_i) = 0 \) in eq. (4.45)). Note that the integral over a discrete time variable \( f(t) = f(jh), \; j \in \mathbb{N} \), is evaluated using the trapezoidal rule, i.e.,

\[ \int_0^\infty dt \; f(t) = \frac{h}{2} f(0) + h \sum_{j=1}^\infty f(jh). \]  

The correlation function \( \langle \sigma_{xz}(t)\sigma_{xz}(0) \rangle \) comprises autocorrelation functions of the kinetic and collisional stress tensors, respectively, as well as cross terms \( \langle \sigma_{xz}^{i,k} \sigma_{xz}^{i,c} \rangle \). By simulations, we determine the cross correlations for a wide range of MPC parameters and find \( \langle \sigma_{xz}^{i,k} \sigma_{xz}^{i,c} \rangle = 0 \) for the particle-level stress tensor of eq. (4.45)"[A]. Hence, we will assume this identity in the following\(^a\). Consequently, the viscosity is simply the sum of a kinetic and collisional contribution For the kinetic viscosity, we obtain within the molecular chaos approximation [121, 160, 165, 167, 170]

\[ \eta_2^k = \frac{V}{k_B T} \left( \frac{h}{2} \langle \sigma_{xz}^{i,k}(0)^2 \rangle + h \sum_{l=1}^\infty \langle \sigma_{xz}^{i,k}(lh)\sigma_{xz}^{i,k}(0) \rangle \right) \]  
\[ = \frac{Vh}{k_B T} \langle \sigma_{xz}^{i,k}(0)^2 \rangle \left( \frac{1}{2} + \sum_{l=1}^\infty f_l \right) \]  
\[ = \frac{Nk_B T}{V} \left( \frac{1}{1-f} - \frac{1}{2} \right) \]  

\(^a\)In ref. [35], the Navier-Stokes equation was derived from the MPC algorithm by a Chapman-Enskog expansion. It was shown that up to \( O(\varepsilon^2) \), kinetic and collisional contributions to the transport coefficients decouple.
where we used eq. (5.3) for $\dot{\gamma} = 0$ as well as the definition
\[
\langle v'_ix'_i v'_i z_i \rangle_C = f \langle v'_ix'_i v'_i z_i \rangle_C,
\]
and $\langle \cdots \rangle_C$ denotes the average over the collision operator, which we perform before the ensemble average. The factor $f$ is given by (see appendix 5.B)
\[
f = \frac{1}{\bar{N}_c} + \left(1 - \frac{1}{\bar{N}_c}\right) \frac{1}{5} \left(1 + 2 \cos(\alpha) + 2 \cos(2\alpha)\right)
\] (5.10)
for SRD-a [47, 160, 165, 167, 170] and $f = 1/\bar{N}_c$ for AT-a [47]. Similarly, the collisional viscosity of MPC-a follows as [167]
\[
\eta_c^R = \frac{V}{k_B T} \frac{h}{2} \langle \sigma^{l,c}_{xz}(0) \sigma^{l,c}_{xz}(0) \rangle \tag{5.11}
\]
\[
= \frac{1}{2k_BT k_h} \sum_{i,j} \langle \Delta p_{ix} \Delta p_{jz} \rangle \langle \Delta r_{iz} \Delta r_{jz} \rangle = \frac{N ma^2}{12Vh} g,
\]
by assuming that in MPC-a the momentum change $\Delta p_i$ is independent of the position $\Delta r_i$ \cite{A}. Furthermore we assumed molecular chaos and a uniform distribution of positions in a cell, which results in the variance $\langle \Delta r_{iz}^2 \rangle = a^2/12$. Note that we neglect all higher correlations $\langle \sigma^{l,c}_{xz}(lh) \sigma^{l,c}_{xz}(0) \rangle$ for $l = 1, 2, 3...$, which is motivated by simulation results. Furthermore, we defined $g = m \sum_a \langle \Delta v_{ia} \Delta v_{ia} \rangle/(6Nk_BT)$, which becomes (see appendix 5.C)
\[
g = \frac{2}{3} \left(1 - \frac{1}{\bar{N}_c}\right) (1 - \cos \alpha),
\] (5.12)
for SRD-a and $g = 1 - 1/\bar{N}_c$ for AT-a [121]. In ref. [170], the same result for the viscosity was derived by calculating the momentum transfer across a plane in shear flow. A calculation of the collisional viscosity by means of a Green-Kubo relation was also performed in ref. [36] with a stress tensor defined on the cell rather than the particle level. In that case, the correlations $\langle \sigma^{l,c}_{xz}(lh) \sigma^{l,c}_{xz}(0) \rangle$ and $\langle \sigma^{l,c}_{xz} \sigma^{l,c}_{xz} \rangle$ are in fact not negligible [36]\cite{A}.

### 5.2.3 Green-Kubo relation for bulk viscosity

In terms of the dynamic pressure $P$ and its fluctuations $\delta P$ defined as
\[
P = -\frac{1}{3} \sum_a \sigma_{aa},
\] (5.13)
\[
\delta P = P - \langle P \rangle - \frac{2}{3V} (E_{\text{kin}} - \langle E_{\text{kin}} \rangle),
\] (5.14)
b) In the following expression, $v_i$ and $v'_i$ denote the velocity before and after the collision step respectively. Note that the velocity does not change in the streaming step.
5.2 Analytical calculation of viscosity parameters

The bulk viscosity follows from the Green Kubo relation [169]

\[ \eta^V = \frac{V}{k_B T} \int_0^\infty dt \langle \delta P(t) \delta P(0) \rangle. \]  (5.15)

Note that by definition \( \delta P \) is independent of the kinetic stress, which therefore does not contribute to \( \eta^V \). Furthermore, the dynamic pressure \( P \) is not affected by the presence or absence of angular momentum conservation, i.e., by the term \( \mathbf{\omega} \times \mathbf{r}_{ic} \) in the collision rule of MPC+a (see eq. (4.14)). This evidently follows from the calculation

\[ \sum_{i=1}^{N_c} \mathbf{r}_i \cdot [\mathbf{\omega} \times (\mathbf{r}_i - \mathbf{r}_{cm})] = -\sum_{i=1}^{N_c} \mathbf{r}_i \cdot (\mathbf{\omega} \times \mathbf{r}_{cm}) = -N_c \mathbf{r}_{cm} \cdot (\mathbf{\omega} \times \mathbf{r}_{cm}) = 0. \]  (5.16)

Hence, the bulk viscosity is identical for MPC+a and MPC-a, which constitutes an important result [A]. Analogous to the derivation of eq. (5.11) and within the molecular chaos assumption, we find for MPC-a

\[ \eta^V = \frac{V}{k_B T} \frac{\hbar}{2} \langle \delta P(0) \delta P(0) \rangle = \frac{1}{18k_B TV h} \sum_{i=1}^{N_c} \langle \Delta p_{i\alpha} \Delta p_{i\beta} \rangle \langle \Delta r_{i\alpha} \Delta r_{i\beta} \rangle \]  (5.17)

\[ = \frac{1}{18k_B TV h} \frac{m^2 a^2}{12} \sum_{i=1}^{N_c} \langle \Delta v_{i\alpha} \Delta v_{i\alpha} \rangle = \frac{1}{3} \eta^c. \]  (5.18)

5.2.4 Resulting stress tensor for MPC fluid

**MPC-a**  
For MPC-a we established the relations \( \eta^k_1 = \eta^k_2 \equiv \eta^k \), \( \eta^c_1 = 0, 0 < \eta^c_2 \equiv \eta^c \), and \( \eta^V = \eta^c / 3 \). Using eqs. (2.10) and (4.47), this leads to the stress tensor

\[ \sigma_{\alpha\beta} = -p \delta_{\alpha\beta} + \eta^k \left( \partial_{\alpha} v_{\beta} + \partial_{\beta} v_{\alpha} - \frac{2}{3} \delta_{\alpha\beta} \sum_{\gamma} \partial_{\gamma} v_{\gamma} \right) + \eta^c \partial_{\alpha} v_{\beta}. \]  (5.19)

Analytical expressions for \( \eta^k \) and \( \eta^c \) are provided in eqs. (5.9) and (5.11). We like to emphasize that eq. (5.19) agrees with the stress tensor derived in refs. [35, 166] [A].

**MPC+a**  
For MPC+a we found \( \eta_1 = \eta_2 \equiv \eta \), and the stress tensor reads

\[ \sigma_{\alpha\beta} = -p \delta_{\alpha\beta} + \eta \left( \partial_{\alpha} v_{\beta} + \partial_{\beta} v_{\alpha} - \frac{2}{3} \delta_{\alpha\beta} \sum_{\gamma} \partial_{\gamma} v_{\gamma} \right) + \eta^V \delta_{\alpha\beta} \sum_{\gamma} \partial_{\gamma} v_{\gamma}. \]  (5.20)

Here, the bulk viscosity is \( \eta^V = \eta^c / 3 \), where \( \eta^c \) is the collisional viscosity of MPC-a. Note that in general, the viscosities \( \eta^k \) and \( \eta^c \) are different for MPC-a and MPC+a.

Table 5.1 summarizes our findings in terms of the parameters \( \eta_1, \eta_2, \) and \( \eta^V \) [A].
\[ \sigma_{\alpha\beta} = -p\delta_{\alpha\beta} + \eta_1 \partial_\alpha v_\beta + \eta_2 \partial_\beta v_\alpha + \eta_3 \delta_{\alpha\beta} \sum_\gamma \partial_\gamma v_\gamma \]

**Table 5.1:** “Viscosity relations for MPC±a variants. \( \eta = \eta_2 \) is the shear viscosity and \( \eta^V = (\eta_1 + \eta_2 + 3\eta_3)/3 \) the bulk viscosity. \( \eta_1 + \eta_2 + \eta_3 \) is the viscous contribution to the sound attenuation coefficient. Note that the viscosities \( \eta^k \) and \( \eta^c \) are different for MPC-a and MPC+a [121]. However, in any case, the bulk viscosity is given by \( \eta^V = \eta^c/3 \), where \( \eta^c \) is the collisional viscosity of MPC-a” [A]. (Table from publication [A])

<table>
<thead>
<tr>
<th></th>
<th>( h/\sqrt{ma^2/(k_B T)} )</th>
<th>0.01</th>
<th>0.03</th>
<th>0.1</th>
<th>0.3</th>
<th>1</th>
</tr>
</thead>
<tbody>
<tr>
<td>SRD+a</td>
<td>( \eta/\sqrt{mk_B T/a^4} )</td>
<td>35.2</td>
<td>11.9</td>
<td>4.26</td>
<td>2.94</td>
<td>5.75</td>
</tr>
<tr>
<td>analytical</td>
<td></td>
<td>39.3</td>
<td>13.2</td>
<td>4.44</td>
<td>2.87</td>
<td>5.59</td>
</tr>
<tr>
<td>AT+a</td>
<td>( \eta/\sqrt{mk_B T/a^4} )</td>
<td>32.0</td>
<td>10.9</td>
<td>4.09</td>
<td>3.21</td>
<td>6.92</td>
</tr>
<tr>
<td>analytical</td>
<td></td>
<td>35.9</td>
<td>12.1</td>
<td>4.23</td>
<td>3.12</td>
<td>6.79</td>
</tr>
</tbody>
</table>

**Table 5.2:** “Measured and respective theoretical (ref. [121]) shear viscosities of MPC+a variants” [A]. (Table from publication [A])

### 5.3 Measurement of viscosity parameters

#### 5.3.1 Viscosities \( \eta_1 \) and \( \eta_2 \)

“To determine \( \eta_1 \) and \( \eta_2 \), we perform shear simulations using Lees-Edwards boundary conditions [157] (see section 4.3). The viscosities follow from eq. (5.2), with the stress tensor calculated according to eq. (5.3). Our simulation results confirm eqs. (5.5) and (5.6) for both, SRD±a and AT±a. Moreover, the simulations validate the analytical formula for \( \eta^c \) for AT-a and SRD-a. As in previous simulation studies, we find that the analytical formulae for \( \eta^k \) of MPC-a become increasingly imprecise for smaller time steps due to the applied molecular chaos assumption in their derivation. However, the total shear viscosity agrees very well with the theoretical expression, even at small collision times, because the collisional viscosity dominates at small \( h \) [159].

The analytical expressions for the shear viscosity of SRD+a and AT+a given in ref. [47] are found to be less accurate than those for the non-angular momentum conserving variants, as indicated in fig. 5.1 and table 5.2” [A].

#### 5.3.2 Bulk viscosity

“We measure the bulk viscosity by means of the Green-Kubo relation (5.15) performing equilibrium simulations. The integral is evaluated using the trapezoidal
5.3 Measurement of viscosity parameters

![Graph showing viscosity parameters](image)

Figure 5.1: “Shear viscosity $\eta$ for a SRD+a fluid. The open symbols denote simulation results for the MPC parameters $\langle N_c \rangle = 10$, $\alpha = 130^\circ$, the shear rate $\dot{\gamma} = 0.01 \sqrt{k_B T/(ma^2)}$, and the size of the cubic simulation box $L = 40a$. The solid line represents the theoretical result provided in ref. [47]. The measured values are also listed in Table 5.2”[A]. (Figure from publication [A])

rule to account for the discrete time process. The correlation function $\langle \delta P(t)\delta P(0) \rangle$ decays extremely rapidly. Already after one collision step, the correlation function is essentially zero. Hence, $\eta^V$ is well described by eq. (5.18), with the essential contribution at zero time lag. The measured bulk viscosities are presented in fig. 5.2. They agree very well with the analytical prediction.

We also determined the bulk viscosity for two-dimensional MPC fluids and found $\eta^V = \eta^f/2$, in agreement with theoretical calculations similar to those of section 5.2. Hence, in general, the bulk viscosity is $\eta^V = \eta_c/d$, where $d$ denotes the spatial dimension”[A].

5.3.3 Viscous contribution to sound attenuation

The simulation results this far confirmed the theoretical predictions for all three viscosity parameters. As an additional check, we measure the longitudinal velocity autocorrelation function (LVACF) of the MPC fluid, which is connected to sound propagation. A Fourier transform of the velocity field $\mathbf{v}(\mathbf{r},t)$ yields $\mathbf{v}(\mathbf{k},t)$. The longitudinal velocity mode is defined by $\mathbf{v}_L(\mathbf{k},t) = (1 - \mathbf{k}\mathbf{k}^T/k^2)\mathbf{v}(\mathbf{k},t)$. As shown in appendix 5.A, the LVACF of an isothermal fluid in a periodic cubic box of volume
Chapter 5 Bulk viscosity of multiparticle collision dynamics fluids

\[ \eta^V \left( \sqrt{ma^2/(k_BT)} \right) \]

\[ h \left( \sqrt{ma^2/(k_BT)} \right) \]

Figure 5.2: “Bulk viscosities of MPC±a variants for various collision steps \( h \). Circles (red) and diamonds (blue) correspond to SRD-a and SRD+a fluids, respectively, while the squares (green) and triangles (magenta) correspond to AT-a and AT+a fluids. The simulation parameters are \( \langle N_c \rangle = 10 \), \( \alpha = 130^\circ \), and \( L = 10a \) for SRD, and \( \langle N_c \rangle = 5 \) for AT. The black lines represent the theoretical expectation \( \eta^V = \eta^c/3 \), where \( \eta^c \) is the collisional viscosity of MPC-a (see eq. (5.11)). The top solid and bottom dashed line correspond to SRD and AT, respectively.” [A]. (Figure from publication [A])

\[ V \text{ is given by} \]

\[ C_L(k,t) = \langle v_L(k,t) \cdot v_L(k,0) \rangle \]

\[ = \frac{k_BT}{\rho_0 V} e^{-k^2\tilde{\nu}^2/2} \left[ \cos(\Omega|t|) - \sqrt{\frac{k^2\tilde{\nu}^2}{4c^2-k^2\tilde{\nu}^2}} \sin(\Omega|t|) \right] \text{ for } \frac{4c^2}{k^2\tilde{\nu}^2} > 1. \ (5.21) \]

Here, \( \rho_0 \) is the equilibrium mass density, \( c \) is the velocity of sound, which is \( c = \sqrt{k_BT/m} \) for an ideal gas such as MPC, \( \Omega = k^2\tilde{\nu}/\sqrt{4c^2/(k^2\tilde{\nu}^2)} - 1/2 \), \( \nu = \eta/\rho_0 \), \( \tilde{\nu} = \tilde{\eta}/\rho_0 \) and the viscous contribution to the sound attenuation \( \tilde{\eta} = \eta_1 + \eta_2 + \eta_3 \). The derivation of this result and the expression for \( \frac{4c^2}{k^2\tilde{\nu}^2} < 1 \) can be found in appendix 5.A.

In order to measure the longitudinal velocity correlation function (5.21) of the MPC fluid, we perform the Fourier expansion

\[ v(k,t) = \frac{1}{V} \int d^3r \ v(r,t) e^{ik\cdot r} = \frac{1}{V} \int d^3r \ \left( \frac{1}{N/V} \sum_{i=1}^{N} \delta(r-r_i) v_i \right) e^{ik\cdot r} \ (5.22) \]

\[ = \frac{1}{N} \sum_{i=1}^{N} v_i(t) e^{ik\cdot r_i(t)} \ (5.23) \]
5.3 Measurement of viscosity parameters

Figure 5.3: “Longitudinal velocity autocorrelation function for $k = 2\pi/L$ and the length $L = 40a$ of a cubic box. The red and blue lines correspond to SRD+a with $\langle N_c \rangle = 10$, $\alpha = 130^\circ$, and $h/\sqrt{ma^2/(k_B T)} = 0.01$; the green and black lines correspond to AT+a with $\langle N_c \rangle = 10$ and $h/\sqrt{ma^2/(k_B T)} = 0.05$. Dashed lines represent simulation results. The solid lines represent the theoretical expression in eq. (5.21) with $\tilde{\nu} = (4\eta/3 + \eta^V)/\rho_0$, while for the dotted lines the viscosity $\tilde{\nu} = (4\eta/3)/\rho_0$ without bulk contribution is used”[A]. (Figure from publication [A])

of the MPC particle velocities.

“The decay of the longitudinal velocity correlation function (5.21) is governed by the viscosity $\tilde{\eta} = \eta_1 + \eta_2 + \eta_3 = 2/3(\eta_1 + \eta_2) + \eta^V$. For the non-angular momentum conserving variants of MPC, the expression reduces to $\tilde{\eta} = 4\eta^k/3 + \eta^c$, as already discussed in refs. [164, 168, 171], which includes the bulk viscosity.

More importantly, the bulk viscosity also contributes to sound attenuation in an angular momentum conserving MPC fluid. The effect of $\eta^V$ is clearly visible for both, SRD+a and AT+a in fig. 5.3. Since $\eta^c \propto h^{-1}$ and $\eta^k \propto h$, the bulk viscosity contributes significantly to the decay of the longitudinal correlation function at small collision time steps. We calculated the LVACF for several collision step sizes and find good agreement with the theoretical prediction as long as the time step $h$ is small, i.e., $h/\sqrt{ma^2/(k_B T)} \lesssim 0.1$. As discussed in refs. [159, 171], larger time steps result in a substantial heat transfer between cells in the streaming step and consequently the isothermal theory is no longer applicable”[A].

The longitudinal velocity autocorrelation function for MPC-SRD-a has been extensively studied in ref. [171]. For MPC-AT-a we find equally satisfying agreement between theory and simulations, as long as $h/\sqrt{ma^2/(k_B T)} \lesssim 0.1$ (see fig. 5.4). Hence, we have verified the stress tensor expressions of section 5.2.4 for MPC-SRD±a as well as MPC-AT±a.
Chapter 5 Bulk viscosity of multiparticle collision dynamics fluids

\[ C_l(k, t) / C_l(k, 0) \]

Figure 5.4: Longitudinal velocity autocorrelation function for \( k = 2\pi / L \) and the length \( L = 40a \) of a cubic box. The green and black lines correspond to AT-a with \( \langle N_c \rangle = 10 \) and \( h / \sqrt{ma^2/(k_BT)} = 0.05 \); the red and blue lines correspond to AT-a with \( \langle N_c \rangle = 10 \) and \( h / \sqrt{ma^2/(k_BT)} = 0.1 \). Dashed lines represent simulation results. The solid lines correspond to the theoretical expression in eq. (5.21) with \( \tilde{\nu} = (4\eta^k/3 + \eta^c)/\rho_0 \).

5.4 Summary and conclusions

"We have determined the viscous transport coefficients for SRD and AT variants of MPC fluids, by both, analytical considerations and simulations. As a main result, we find a nonzero bulk viscosity for all MPC variants, with and without angular momentum conservation.

A nonzero bulk viscosity for MPC-SRD-a has already been indicated in ref. [164] in connection with the stress tensor of ref. [166]. An alternative stress tensor has been formulated, which differs only by a term of vanishing divergence from eq. (5.19), and thus, yields the identical Navier-Stokes equations [164, 168, 171]. However, in ref. [168], it has been concluded that the bulk viscosity for this stress tensor is zero. The lack of angular momentum conservation leaves more fluid parameters undetermined than an angular momentum conserving one. Hence, by considering the shear viscosity and the sound attenuation factor only, the lack or presence of a bulk viscosity cannot be verified, which renders the various stress tensors seemingly equivalent. However, the Navier-Stokes equations have to be supplemented by boundary conditions, which can depend explicitly on the stress tensor, as is the case with (partial) slip boundary conditions (see section 2.2.4). As a consequence, all three viscosity parameters determine the velocity field and only the stress tensor eq. (5.19) is appropriate for MPC-a fluids.

The situation is different for angular momentum conserving fluids, where the sym-
metry requirement of the stress tensor reduces the number of independent viscosity parameters to two. Hence, the shear viscosity and the sound attenuation factor determine $\eta^V$ uniquely. Vice versa, the bulk viscosity is an integral part of the sound attenuation factor $\tilde{\eta}$. Specifically for MPC at small collision time steps, where the collisional viscosity $\eta^c$ dominates the shear viscosity and $\eta \approx \eta^c$, the bulk viscosity is essential for the correct sound attenuation factor. We confirmed the strong influence of the bulk viscosity on the decay of the sound correlation function for MPC+a versions by simulations. The presence of $\eta^V$ has consequences for all those correlation functions, which include the longitudinal mode. In particular, the velocity correlation functions are affected of, e.g., colloids[A].

"We found that the shear and bulk viscosity of a MPC fluid are of the same order of magnitude. For many real fluids, such as water, the bulk viscosity is hundreds to thousands times larger than the shear viscosity [172]. However, the effect of a large bulk viscosity is most pronounced at high frequency hydrodynamics, where compressibility effects matter most.

The presence of a nonzero bulk viscosity is seemingly in contradiction with the ideal gas equation of state of MPC and the zero bulk viscosity of an ideal gas. As is evident, the bulk viscosity is determined by collisions only. Hence, for a weakly interacting MPC fluid, which we may call a gas, $\eta^c$ is negligibly small and we may set it to zero. Hence, we reach the ideal gas limit for large collision time steps. This is supported by the Schmidt number, which assumes gas-like values for large collision time steps [158]. Thus, a non-vanishing bulk viscosity is natural for small collision time steps, because here the MPC fluid corresponds to a fluid rather than a gas"[A].

### Appendix 5.A Hydrodynamic correlations

In this appendix, we derive the velocity autocorrelation function of an isothermal fluid in a cubic box with periodic boundary conditions. We approximate the equation of state of our fluid up to first order in $\rho$ (which is exact for an ideal gas)

$$p = \rho c^2.$$  (5.24)

Here, $c$ is the isothermal velocity of sound. Thermal fluctuations can be modeled by adding a Gaussian and Markovian stochastic process $\sigma^R$ to the stress tensor $\sigma$ (Landau-Lifshitz Navier-Stokes approach) [62, 68, 171], with

$$\langle \sigma^R \rangle = 0,$$  (5.25)

$$\langle \sigma_{\alpha \beta}^R(r, t) \sigma_{\alpha' \beta'}^R(r', t') \rangle = 2k_B T \eta_{\alpha \alpha' \beta \beta'} \delta(r - r') \delta(t - t').$$  (5.26)

The tensor $\eta_{\alpha \beta \alpha' \beta'}$ is given in eq. (2.7). We define the random force $f^R = \nabla \cdot \sigma^R$. 

Within linearized hydrodynamics [173], it is assumed that $\rho$ is nearly constant with a small fluctuation $\delta \rho$ and the velocity itself is a small fluctuation $\delta v$. Substitution of

$$\rho = \rho_0 + \delta \rho, \quad \rho_0 = \text{const.}, \quad v = \delta v$$  \hspace{1cm} (5.27)$$

into eqs. (2.16) and (2.20) and disregarding terms quadratic in $(\delta \rho, \delta v)$ leads to the linearized equations

$$\rho_0 \frac{\partial v}{\partial t} = -\nabla p + \eta \Delta v + (\tilde{\eta} - \eta) \nabla (\nabla \cdot v) + \mathbf{f}^R,$$  \hspace{1cm} (5.28)$$

$$\frac{\partial \rho}{\partial t} = -\rho_0 \nabla \cdot \mathbf{v}.$$  \hspace{1cm} (5.29)$$

where $\tilde{\eta} = \eta_1 + \eta_2 + \eta_3$. Taking the divergence of eq. (5.28) and using the linearized continuity eq. (5.29) together with the ideal gas equation of state (5.24), we obtain the wave equation

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \rho = \nabla \cdot \{\eta \Delta v + (\tilde{\eta} - \eta) \nabla (\nabla \cdot v) + \mathbf{f}^R\}. \hspace{1cm} (5.30)$$

It is advantageous to perform a Fourier series expansion in $r$ and a Fourier transform in $\omega$ according to [171]

$$\mathbf{v}(r, t) = \frac{1}{2\pi} \sum_k \int d\omega \ \hat{\mathbf{v}}(k, \omega) e^{-ik \cdot r} e^{i\omega t}, \hspace{1cm} (5.31)$$

$$\hat{\mathbf{v}}(k, \omega) = \frac{1}{V} \int d^3r \int dt \ \mathbf{v}(r, t) e^{ik \cdot r} e^{-i\omega t}, \hspace{1cm} (5.32)$$

where $V$ is the volume of the cubic system of length $L$, and $k_\alpha = n_\alpha 2\pi/L, n_\alpha \in \mathbb{Z}, k \neq 0$. A Fourier transform of eqs. (5.28) and (5.30) yields

$$\left(-k^2 + \frac{\omega^2}{c^2}\right) \hat{\rho} = ik^T \left\{-\eta k^2 \hat{\mathbf{v}} - (\tilde{\eta} - \eta) kk^T \hat{\mathbf{v}} + \hat{\mathbf{f}}^R\right\}, \hspace{1cm} (5.33)$$

$$\rho_0 i\omega \hat{\mathbf{v}} = -ik \hat{\rho} - \eta k^2 \hat{\mathbf{v}} - (\tilde{\eta} - \eta) kk^T \hat{\mathbf{v}} + \hat{\mathbf{f}}^R. \hspace{1cm} (5.34)$$

In order to solve eqs. (5.33) and (5.34), we define the longitudinal and transverse projection operators $P_L(k) \equiv kk^T/k^2$ and $P_T(k) \equiv 1 - P_L(k)$ along with $\hat{\mathbf{v}}_L \equiv P_L \hat{\mathbf{v}}$ and $\hat{\mathbf{v}}_T \equiv P_T \hat{\mathbf{v}}$. Applying $P_T$ on eq. (5.34), we find

$$\hat{\mathbf{v}}_T = \left[\rho_0 i\omega + \eta k^2\right]^{-1} P_T \hat{\mathbf{f}}^R \equiv \hat{G}_T(k, \omega) P_T(k) \hat{\mathbf{f}}^R.$$  \hspace{1cm} (5.35)$$

Now, we multiply eq. (5.34) by $k^T$, solve for $\hat{\rho}$ and substitute this into eq. (5.33), which yields

$$\hat{\mathbf{v}}_L = \left[\tilde{\eta} k^2 + \frac{i\rho_0}{\omega} (\omega^2 - k^2 c^2)\right]^{-1} P_L \hat{\mathbf{f}}^R \equiv \hat{G}_L(k, \omega) P_L(k) \hat{\mathbf{f}}^R. \hspace{1cm} (5.36)$$
To find the velocity correlation function in Fourier space $\langle \hat{v}(k, \omega) \cdot \hat{v}(k', \omega)' \rangle$, where $z^*$ denotes the complex conjugate of $z$, we evaluate

$$\langle \hat{f}^R_{\alpha}(k, \omega) \hat{f}^R_{\beta}(k', \omega)' \rangle = \frac{4\pi k_B T}{V} \sum_{\alpha'\beta'} k_{\alpha'\beta'} \eta_{\alpha'\beta'} \delta(\omega - \omega') \delta_{k,k'}. \quad (5.37)$$

Utilizing this result, together with eq. (2.7), we find after some algebra

$$\langle \hat{v}(k, \omega) \cdot \hat{v}(k', \omega)' \rangle = \frac{4\pi k_B T}{V} k^2 \left( 2\eta |\hat{G}_T|^2 + \tilde{\eta} |\hat{G}_L|^2 \right) \delta(\omega - \omega') \delta_{k,k'}. \quad (5.38)$$

By Fourier transformation we obtain

$$C_T(k, t) = \langle v_T(k, t) \cdot v_T(k, 0) \rangle = \frac{2k_B T}{\rho_0 V} e^{-\nu k^2 |t|}, \quad (5.39)$$

$$C_L(k, t) = \langle v_L(k, t) \cdot v_L(k, 0) \rangle$$

$$\begin{cases} k_B T_{\rho_0 V} e^{-\nu k^2 |t|/2} \left[ \cos(\Omega |t|) - \sqrt{\frac{k^2 \nu^2}{4c^2 - k^2 \nu^2}} \sin(\Omega |t|) \right], & \text{if } \frac{4c^2}{k^2 \nu^2} > 1, \\
\frac{k_B T_{\rho_0 V} e^{-\nu k^2 |t|/2} \left[ \cosh(\lambda |t|) - \sqrt{\frac{k^2 \nu^2}{k^2 \nu^2 - 4c^2}} \sinh(\lambda |t|) \right], & \text{if } \frac{4c^2}{k^2 \nu^2} < 1, \end{cases} \quad (5.40)$$

where we introduced the kinematic viscosity $\nu = \eta/\rho_0$, as well as $\tilde{\nu} = \tilde{\eta}/\rho_0$, $\Omega = k^2 \tilde{\nu} \sqrt{4c^2/(k^2 \nu^2) - 1}/2$ and $\lambda = k^2 \tilde{\nu} \sqrt{1 - 4c^2/(k^2 \nu^2)}/2$.

### Appendix 5.B Derivation of decorrelation factor $f$

The decorrelation factor $f$ is defined by the relation

$$\langle v'_{ix} v'_{iy} \rangle = f \langle v_{ix} v_{iy} \rangle. \quad (5.41)$$

Here, $v'_i$ and $v_i$ are the velocities after and before the collision step, respectively. We evaluate $f$ separately for MPC-AT-a and MPC-SRD-a.

**MPC-AT-a** Using eq. (4.7), we find

$$\langle v'_{ix} v'_{iy} \rangle = \langle (v_{cm,x} + C[v_{ic}]_x)(v_{cm,y} + C[v_{ic}]_y) \rangle = \langle v_{cm,x} v_{cm,y} \rangle$$

$$\frac{1}{N_c^2} \sum_{i,j} \langle v_{ix} v_{iy} \rangle = \frac{1}{N_c^2} \sum_i \langle v_{ix} v_{iy} \rangle = \frac{1}{N_c} \langle v_{ix} v_{iy} \rangle, \quad (5.42)$$

and hence

$$f = \frac{1}{N_c}. \quad (5.43)$$

Note that in principle, we have to average our result over the Poisson distribution of $N_c$ as in ref. [47]. However, since we typically work with $N_c \geq 10$, we can put $N_c = \langle N_c \rangle = \bar{N}_c$ in our expressions without any significant error. This holds for all following calculations in this section and in section 5.C.
Using the collision rule of MPC-SRD-a and abbreviating \( R = R(\alpha) \), we find

\[
\langle v'_{ix} v'_{iy} \rangle = \left\langle \left[ v_{cm,x} + (Rv_i)_x - (Rv_{cm})_x \right] \left[ v_{cm,y} + (Rv_i)_y - (Rv_{cm})_y \right] \right\rangle. \tag{5.45}
\]

We evaluate the several resulting terms using the molecular chaos assumption [47]

\[
\langle v_{cm,x} v_{cm,y} \rangle = \frac{1}{N_c} \sum_{i,j} \langle v_{ix} v_{iy} \rangle = \frac{1}{N_c} \sum_i \langle v_{ix} v_{iy} \rangle = \frac{1}{N_c} \langle v_{ix} v_{iy} \rangle, \tag{5.46}
\]

\[
2 \langle v_{cm,x} (Rv_i)_y \rangle = \frac{2}{N_c} \sum_j \langle v_{jx} (Rv_i)_y \rangle = \frac{2}{N_c} \langle v_{ix} (Rv_i)_y \rangle, \tag{5.47}
\]

\[
-2 \langle v_{cm,x} (Rv_{cm})_y \rangle = -\frac{2}{N_c} \sum_{i,j} \langle v_{jx} (Rv_i)_y \rangle = -\frac{2}{N_c} \langle v_{ix} (Rv_i)_y \rangle, \tag{5.48}
\]

\[
\langle (Rv_i)_x (Rv_i)_y \rangle = f_0 \langle v_{ix} v_{iy} \rangle, \tag{5.49}
\]

\[
-2 \langle (Rv_i)_x (Rv_{cm})_y \rangle = -\frac{2}{N_c} \sum_j \langle (Rv_i)_x (Rv_j)_y \rangle = -\frac{2 f_0}{N_c} \langle v_{ix} v_{iy} \rangle, \tag{5.50}
\]

\[
\langle (Rv_{cm})_x (Rv_{cm})_y \rangle = f_0 \langle v_{cm,x,v_{cm}}, \rangle \tag{5.46} = \frac{f_0}{N_c} \langle v_{ix} v_{iy} \rangle. \tag{5.51}
\]

Here, we exploited that due to symmetry all terms are invariant under the exchange \( x \leftrightarrow y \) and we defined \( f_0 \) via \( \langle (Rv)_x (Rv)_y \rangle = f_0 \langle v_x v_y \rangle \). Summing up all terms yields

\[
\langle v'_{ix} v'_{iy} \rangle = \left[ \frac{1}{N_c} + \left( 1 - \frac{1}{N_c} \right) f_0 \right] \langle v_{ix} v_{iy} \rangle. \tag{5.52}
\]

The factor \( f_0 \) can be determined by averaging over velocities and rotations separately

\[
\langle (Rv)_x (Rv)_y \rangle = \sum_{\alpha,\beta} \langle R_{xa} v_{\alpha} R_{y\beta} v_{\beta} \rangle = \sum_{\alpha,\beta} \langle R_{xa} R_{y\beta} \rangle \langle v_{\alpha} v_{\beta} \rangle. \tag{5.53}
\]

We proceed in two steps. First we note that only terms with \( \alpha \neq \beta \) contribute, since \( \langle v_{\alpha}^2 \rangle = k_B T/m \) and

\[
\sum_{\alpha} R_{xa} R_{ya} = (RR^T)_{xy} = 0. \tag{5.54}
\]

Secondly, we analyze all remaining factors \( \langle R_{xa} R_{y\beta} \rangle \) separately. Here, the average is performed over the rotation axis \( n \) and hence the uniformly distributed random
5.C Derivation of decorrelation factor $g$

Numbers $\phi \in [0, 2\pi]$ and $u \in [-1, 1]$ in eq. (4.5). Different products of $n_\alpha$ arise, but the symmetries $x \leftrightarrow y \leftrightarrow z$ decrease the numerous cases to

\begin{equation}
\langle n_x^2 \rangle = \frac{1}{2\pi} \int_0^{2\pi} d\phi \frac{1}{2} \int_{-1}^{1} du \ u^2 = \frac{1}{3}, \tag{5.55}
\end{equation}

\begin{equation}
\langle n_x^2 n_z^2 \rangle = \frac{1}{4\pi} \int_0^{2\pi} d\phi \int_{-1}^{1} du (1 - u^2) \cos^2(\phi)u^2 = \frac{1}{15}, \tag{5.56}
\end{equation}

\begin{equation}
\langle n_x^3 \rangle = 0, \langle n_x n_y \rangle = 0, \langle n_x^2 n_y \rangle = 0, \langle n_x n_y^2 n_z \rangle = 0. \tag{5.57}
\end{equation}

We find that $\langle R_{\alpha\alpha} R_{\beta\beta} \rangle = 0$ unless $\alpha = x, \beta = y$ or $\alpha = y, \beta = x$. Hence, eq. (5.53) simplifies to $\langle (Rv)_x (Rv)_y \rangle = f_0 \langle v_x v_y \rangle$ with

\begin{equation}
f_0 = \langle R_{xx} R_{yy} \rangle + \langle R_{xy} R_{yx} \rangle = \begin{cases} 
\cos^2 \alpha - \sin^2 \alpha = \cos(2\alpha), & \text{in 2D,} \\
\frac{1}{5} [1 + 2 \cos(2\alpha) + \cos \alpha], & \text{in 3D.} 
\end{cases} \tag{5.58}
\end{equation}

Substituting this result into eq. (5.52) we finally obtain

\begin{equation}
f = \begin{cases} 
\frac{1}{N_c} + \left(1 - \frac{1}{N_c}\right) \cos(2\alpha), & \text{in 2D,} \\
\frac{1}{N_c} + \left(1 - \frac{1}{N_c}\right) \frac{1}{5} (1 + 2 \cos(2\alpha) + \cos \alpha), & \text{in 3D.} 
\end{cases} \tag{5.59}
\end{equation}

**Appendix 5.C Derivation of decorrelation factor $g$**

The decorrelation factor $g$ is defined as

\begin{equation}
g = \frac{m}{2d k_B T} \sum_\alpha \langle \Delta v_{i\alpha} \Delta v_{i\alpha} \rangle, \tag{5.60}
\end{equation}

where $d = 2, 3$ is the dimension and $\Delta \mathbf{v}_i$ is the change of momentum of particle $i$ in the collision step

\begin{equation}
\Delta \mathbf{v}_i = \mathbf{v}_i' - \mathbf{v}_i = \mathbf{v}_{cm} + \mathcal{C} \mathbf{v}_{ic} - \mathbf{v}_i = (\mathcal{C} - 1) \mathbf{v}_{ic}. \tag{5.61}
\end{equation}

We will determine $g$ separately for MPC-AT-a and MPC-SRD-a.
\[ \langle (C - 1) v_{ic}^2 \rangle = \left( v_{ic}^2 - \frac{1}{N_c} \sum_{j} v_{ic}^2 \right)^2 + \langle v_{ic}^2 \rangle = 2 \langle v_{ic}^2 \rangle = 2 \left( \langle v_i^2 \rangle - 2 \langle v_i \cdot 1 \rangle + \frac{1}{N_c} \sum_{j} \langle v_j^2 \rangle \right) = 2 \left( 1 - \frac{1}{N_c} \right) \langle v_i^2 \rangle \] (5.62)

\[ = 2d \frac{k_B T}{m} \left( 1 - \frac{1}{N_c} \right). \] (5.63)

Here, we used the equipartition theorem. The final result is

\[ g = 1 - \frac{1}{N_c}. \] (5.66)

**MPC-SRD-a** We define \( \mathbf{\tilde{R}} = \mathbf{R} - 1 \) and find

\[ \langle (C - 1) v_{ic}^2 \rangle = \sum_{\alpha \beta \gamma} \langle \tilde{R}_{\alpha \beta} v_{ic \beta} \tilde{R}_{\alpha \gamma} v_{ic \gamma} \rangle = \sum_{\beta \gamma} \left\langle \left( \tilde{R}^T \mathbf{\tilde{R}} \right)_{\beta \gamma} \right\rangle \langle v_{ic \beta} v_{ic \gamma} \rangle. \] (5.67)

Since \( \tilde{R}^T \mathbf{\tilde{R}} = (\mathbf{R}^T - 1)(\mathbf{R} - 1) = 1 + 1 - \mathbf{R}^T - \mathbf{R} \), the off-diagonal matrix elements of \( \langle \tilde{R}^T \mathbf{\tilde{R}} \rangle \) are zero. Hence,

\[ \langle (C - 1) v_{ic}^2 \rangle = \sum_{\beta} \left\langle \left( \tilde{R}^T \mathbf{\tilde{R}} \right)_{\beta \beta} \right\rangle \langle v_{ic \beta} v_{ic \beta} \rangle = \sum_{\beta} \left\langle \left( \mathbf{R}^T \mathbf{R} \right)_{\beta \beta} \right\rangle \left( 1 - \frac{1}{N_c} \right) \frac{k_B T}{m} \] (5.68)

\[ = \left( 1 - \frac{1}{N_c} \right) \frac{k_B T}{m} \left( 2 \text{Tr}[1] - 2 \text{Tr}[\mathbf{R}] \right) \] (5.69)

\[ = 4 \frac{k_B T}{m} \left( 1 - \frac{1}{N_c} \right) (1 - \cos \alpha). \] (5.70)

For the last equality, see eqs. (4.4) and (4.6). The relation holds equally for \( d = 2 \) and \( d = 3 \). Finally,

\[ g = \frac{2}{d} \left( 1 - \frac{1}{N_c} \right) (1 - \cos \alpha). \] (5.71)
Chapter 6

Relevance of angular momentum conservation in MPC simulations with slip boundary conditions

The results of this chapter have been published in publication [B] (see List of publications on page 203). All verbatim quotes within this chapter are quotations of [B] and are indicated as “…”[B]. Such verbatim quotes can extend over several pages and have been reformatted to fit with the main text. The label of a cited article inside a verbatim quote follows the enumeration in the bibliography (page 186).

6.1 Introduction

The hydrodynamic equations are balance equations for mass, momentum, and energy. Consequently, the conservation of these quantities is a key ingredient in the design of the original MPC-a method [32]. Angular momentum, however, is not conserved in MPC-a, which results in an asymmetric stress tensor for the MPC fluid (see section 2.2). Although this asymmetric stress tensor $\sigma$ is unphysical, its divergence $\nabla \cdot \sigma$ equals that of a symmetric stress tensor, except for a diminished sound attenuation coefficient (see chapter 5). Since the Navier-Stokes equations only depend on the divergence of the stress tensor $\nabla \cdot \sigma$, and not the stress tensor $\sigma$ itself, one could argue that MPC-a reproduces correct fluid dynamics. However, the Navier-Stokes equations have to be supplemented by boundary conditions, which in case of slip boundary conditions, depend explicitly on the stress tensor $\sigma$ (see section 2.2.4). Hence, unphysical flow fields will be obtained by MPC-a simulations of systems involving slip boundary conditions, as we will show in this chapter.

Our considerations are not limited to MPC, but apply to any simulation method that does not conserve angular momentum. As an example, we refer to the SDPD method [174], which has recently been extended to include angular momentum conservation [175].
6.2 Circular Couette flow with a slip boundary

As a particular example, we consider two concentric cylinders of radius $R_1$ and $R_2$ (see fig. 6.1). The outer cylinder has a no-slip surface and rotates at constant angular velocity $\Omega$, while we assume slip boundary conditions on the inner cylinder.

![Sketch of circular Couette flow between concentric cylinders](image)

Figure 6.1: “Sketch of circular Couette flow between concentric cylinders”[B]. (Figure adapted from publication [B])

6.2.1 Theoretical flow field

Given the symmetry of the problem, we employ cylindrical coordinates $(r, \varphi, z)$. The fluid dynamical equations in the Stokes limit read

\[ 0 = \nabla \cdot \mathbf{v}, \quad (6.1) \]
\[ 0 = -\nabla p + \eta_2 \Delta \mathbf{v}, \quad (6.2) \]

with the no-slip boundary condition on the outer wall

\[ v_\varphi(R_2) = R_2 \Omega, \quad (6.3) \]

and the slip boundary condition on the inner wall

\[ \sigma_{rr}(R_1, \varphi, z) = 0, \quad (6.4) \]
\[ \sigma_{\varphi r}(R_1, \varphi, z) = 0. \quad (6.5) \]

Due to symmetry, we choose the Ansatz $p(r) = \text{const.}$ and $\mathbf{v}(r) = v_\varphi(r) \mathbf{e}_\varphi$, which directly satisfies eqs. (6.1) and (6.4). The Stokes eq. (6.2) yields

\[ 0 = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial v}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 v}{\partial \varphi^2} = \left[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial v_\varphi(r)}{\partial r} \right) - \frac{1}{r^2} v_\varphi(r) \right] \mathbf{e}_\varphi. \quad (6.6) \]
The Ansatz $v_\phi(r) = r^n$ yields $n^2 - 1 = 0$, and hence, the general solution

$$v_\phi(r) = Ar + \frac{B}{r}. \quad (6.7)$$

The constants $A$ and $B$ can be obtained by the boundary conditions eqs. (6.3) and (6.5). For eq. (6.5), we evaluate the expression for $\sigma_\phi r = e_\phi^T \sigma e_r$, with the stress tensor (see eq. (4.47))

$$\sigma = -p1 + \eta_1 \nabla v^T + \eta_2 \left( \nabla v^T \right)^T. \quad (6.8)$$

Since $e_\phi$ and $e_r$ are orthogonal, the pressure term does not contribute to $\sigma_\phi r$. For the remaining terms we find

$$e_\phi^T \left( \nabla v^T \right) e_r = e_\phi^T \left( e_r \frac{\partial}{\partial r} v^T + e_r \frac{1}{r} \frac{\partial}{\partial \phi} v^T + e_z \frac{\partial}{\partial z} v^T \right) e_r \quad (6.9)$$

$$= \frac{1}{r} \left( \frac{\partial}{\partial \phi} v^T \right) e_r = \frac{1}{r} \frac{\partial}{\partial \phi} \left( v^T e_r \right) - \frac{1}{r} v^T \frac{\partial}{\partial \phi} e_r \quad (6.10)$$

$$= \frac{1}{r} \frac{\partial v_r}{\partial \phi} - \frac{1}{r^2} v_\phi, \quad (6.11)$$

$$e_\phi^T \left( \nabla v^T \right)^T e_r = e_r^T \left( \nabla v^T e_\phi \right) \quad (6.12)$$

$$= \frac{\partial}{\partial r} v^T \cdot e_\phi = \frac{\partial}{\partial r} \left( v^T e_\phi \right) - v^T \frac{\partial e_\phi}{\partial r} \quad (6.13)$$

$$= \frac{\partial v_\phi}{\partial r}. \quad (6.14)$$

Hence, the boundary conditions eqs. (6.3) and (6.5) yield

$$R_2 \Omega = A R_2 + \frac{B}{R_2}, \quad (6.15)$$

$$0 = -\eta_1 \left( A + \frac{B}{R_1^2} \right) + \eta_2 \left( A - \frac{B}{R_1^2} \right), \quad (6.16)$$

and can easily be solved to obtain

$$A = \frac{\Omega x}{1 + x}, \quad B = \frac{\Omega R_2^2}{1 + x}, \quad x = \frac{R_2^2 \eta_2 + \eta_1}{R_1^2 \eta_2 - \eta_1}. \quad (6.17)$$

"In the case of angular momentum conserving fluids $\eta_1 = \eta_2$ and hence $v_\phi(r) = \Omega r$. This solution is expected, since a uniformly rotating flow does not produce any stresses in a fluid with local angular momentum conservation. As eq. (6.17) for $\eta_1 \neq \eta_2$ shows, this is intrinsically different in the absence of AMC, where the fluid flow is noticeably affected" [B].
Chapter 6 Relevance of AMC in MPC simulations with slip boundary conditions

Figure 6.2: (a) Velocity in $\varphi$-direction as a function of the normalized distance to the cylinder’s center. Symbols correspond to the simulation results with AMC (green) and without AMC (red). The solid line represents the flow field for an angular momentum conserving fluid, while the dashed line corresponds to the theoretical prediction based on eqs. (6.7) and (6.17) with the viscosities from section 5.2. Parameters are given in the text. (b) Corresponding simulation for the time step $h = 0.1\sqrt{ma^2(k_B T)}$ and the angular velocity $\Omega = 10^{-4}\sqrt{k_B T/(ma^2)}$. (Fig. (a) shows the same data as fig. 4 in publication [B]).
6.2.2 Simulations

“To verify the theoretical predictions, we perform simulations with and without AMC. Thereby, we apply the MPC-SRD-a and MPC-SRD+a approach with the MBS-thermostat. The MPC fluid is confined between cylinders of radius $R_1 = 10a$ and $R_2 = 30a$, both of length $L_z = 40a$ and periodic boundary conditions are applied along the $z$-direction”[B]. The boundary conditions at $R_1$ and $R_2$ are implemented as outlined in section 4.3.2. “The angular velocity of the outer cylinder is fixed at $\Omega = 0.003\sqrt{k_B T/(ma^2)}$, the MPC rotation angle is chosen as $\alpha = 130^\circ$, and the particle density as $\rho = 10m/a^3$. For MPC-SRD-a, the collision time is $h = 0.01\sqrt{ma^2/(k_B T)}$, and for MPC-SRD+a $h = 0.0043\sqrt{ma^2/(k_B T)}$, which yields in both cases $\eta = 82\sqrt{mk_BT/a^2}$ as established by independent simulations. Given the linear velocity of the outer cylinder $R_2\Omega$, the Reynolds number is $Re \simeq 0.1$, and the flow velocity can be described within the Stokes limit.

Results for the flow velocities $v_\phi(r)$ are presented in fig. 6.2 (a)”[B]. We find a major difference between MPC-a and MPC+a, as predicted by the theoretical expressions in eqs. (6.7) and (6.17). This shows that in fact, unphysical flow fields are obtained for MPC-a in case of systems involving slip boundary conditions. In fig. 6.2 (b), simulation results for a MPC-SRD-a simulation with collision time step $h = 0.1\sqrt{ma^2/(k_B T)}$ are shown. Since the higher time step reduces viscosity, we decrease the angular velocity to $\Omega = 10^{-4}\sqrt{k_B T/(ma^2)}$ to obtain $Re \lesssim 0.1$. As for $h = 0.01\sqrt{ma^2/(k_B T)}$, we find a good agreement with eqs. (6.7) and (6.17).

6.3 Summary and conclusions

We have shown that MPC simulations without local angular momentum conservation yield unphysical flow fields for systems involving slip boundary conditions. Hence, we strongly recommend the use of MPC+a in this case. But even for systems with no-slip boundary conditions, MPC-a will yield unphysical torques. This is due to the fact, that the torque $T = \int_{\partial V} \sigma \cdot n$ on an immersed object will always depend on the stress tensor itself and not only its divergence. This issue was investigated in ref. [48].

An interesting point regarding our Couette flow simulations is that a uniformly rotating flow produces stresses in MPC-a. For an angular momentum conserving fluid, the uniformly rotating flow

$$v(r) = \Omega \times r$$

does not result in any shear forces (see section 2.1), since

$$\eta \frac{\partial v_\alpha}{\partial r_\beta} + \eta \frac{\partial v_\beta}{\partial r_\alpha} = \eta \sum_\gamma (\epsilon_{\alpha\gamma\beta} + \epsilon_{\beta\gamma\alpha})\Omega_\gamma = 0.$$
For $\eta_1 \neq \eta_2$ however, the viscosity cannot be moved in front of the summation and shear forces will arise.

In publication [B], the simulation of colloids with slip boundary conditions was discussed. As in the studies on Couette flow, an unphysical flow field is obtained if angular momentum conservation is not accounted for. Additionally, the hydrodynamic drag force on a colloid is affected, which we will discuss in chapter 7.
Part III
Colloids
Chapter 7

From local to hydrodynamic friction in Brownian motion: A MPC study

The results of this chapter have been published in publication [C] (see List of publications on page 203). All verbatim quotes within this chapter are quotations of [C] and are indicated as “…”[C]. Such verbatim quotes can extend over several pages and have been reformatted to fit with the main text. The label of a cited article inside a verbatim quote follows the enumeration in the bibliography (page 186).

7.1 Introduction

In chapter 3, we discussed the interplay between hydrodynamic drag forces and thermal random forces on a colloid. Since hydrodynamic theory is only valid for large length and time scales, we expect the resulting correlation functions determined by hydrodynamic friction kernels to only apply on long time scales. In fact, this has already been observed in MPC simulations [40, 90, 122, 126, 127, 161]. “Two different time regimes are typically identified: a short-time regime of uncorrelated fluid-particle motion and local interactions, characterized by molecular chaos, and a long-time regime determined by hydrodynamics. The first regime is typically dominated by ballistic fluid-colloid collisions and is described by the Enskog gas theory [126, 176, 177]; in a more general sense, we will denote this regime as local regime. As a consequence, the VACF decays exponentially for short times with a characteristic time given by the ratio of the local friction coefficient and the colloid mass (see refs. [40, 90, 126, 127, 161] and eq. (3.19)). On longer time scales, the VACF follows the prediction of hydrodynamic theory (Navier-Stokes) and displays an algebraic long-time tail [40, 90, 126, 127, 161, 171, 178].

The separation between local and hydrodynamic time scales affects the frictional and consequently the diffusive behavior of a colloid. Indeed, several studies suggest that the total colloid diffusivity is a sum of a hydrodynamic and a local diffusion coefficient [122, 124, 126]. However, a detailed and decisive study of the relevance of the various contributions for a colloid embedded in a MPC fluid is still missing”[C].
As discussed in chapter 6, “the standard, most often applied implementation of MPC [33, 34] does not conserve angular momentum, which can give rise to unphysical torques [48], and, in case of (partial) slip boundaries, yields flow fields, which deviate from those theoretically predicted. In publication [B], it has been found in particular that the lack of angular momentum conservation combined with (partial) slip boundary conditions leads to a reduction of the friction coefficient compared to that in a fluid with angular momentum conservation. This result is surprising, especially since it was not reported before in studies where the friction of colloids with slip boundary conditions was determined [122, 124, 126]. The reason may lie in the rather involved interpretation of simulated friction coefficients. As mentioned above, the friction measured in MPC simulations is not only determined by hydrodynamics, but also by short-time local processes. Additionally, periodic boundary conditions, typically employed in simulations, affect the frictional behavior. The importance of the various contributions has been addressed before [122, 124, 126], but the validity of Stokes’ law $F = -4\pi \eta R U$ for systems with slip boundary conditions has been presumed. In the light of the modified Stokes law (see publication [B] and eq. (7.3)), the frictional behavior needs to be reconsidered to correctly identify the hydrodynamic and local contributions”[C].

In this chapter, “we generalize the steady-state considerations for the friction coefficient of publication [B] and determine its full frequency dependence. We verify the reduction of the hydrodynamic friction of non-angular-momentum-conserving MPC methods compared to angular momentum conserving ones by measuring force as well as velocity autocorrelation functions. Moreover, the relative contributions of local and hydrodynamic friction are determined by the force autocorrelation function. We determine the diffusion and friction coefficients for colloids of different radii by integration of the autocorrelation function (Green-Kubo) and complement the analysis by sedimentation studies. This enables us to examine the additivity of various contributions to the diffusion coefficient”[C].

7.2 Brownian motion in MPC

7.2.1 Hydrodynamic and local friction

“On long time scales, the MPC solvent can be described by hydrodynamics, whereas on short time scales the molecular chaos assumption applies [158, 171]. For the latter, the relevant time range depends on the collision time $h$. We will denote the resulting friction coefficients on these time scales as hydrodynamic friction $\gamma_h$ and local friction $\gamma_l$. In our simulations, we determine the friction coefficients via velocity and force autocorrelation functions. Theoretical predictions for $C_U(t)$ and $C_K(t)$ can be found by numerical Fourier transformation of eqs. (3.7) and (3.12), with $\tilde{\gamma}(\omega)$ given in appendix 7.A.1. In an analogous manner, $C_O(t)$ and $C_N(t)$ are
determined by Fourier transformation with $\hat{\xi}(\omega)$ of appendix 7.A.2\([C]\).

**Hydrodynamic friction**  “The classical result for the frequency-dependent hydrodynamic friction $\hat{\gamma}(\omega)$ (ref. [179]) assumes local angular momentum conservation for the solvent, i.e., $\eta_1 = \eta_2$ in eq. (4.47). The respective derivation of the friction coefficients for translational and rotational motion for the case $\eta_1 \neq \eta_2$, which applies for MPC-SRD-a, is presented in appendix 7.A.1. In any case, on long time scales, where the MPC fluid is described by hydrodynamics, the correlation function $C_U(t)$ exhibits the well-known algebraic long-time tail (see refs. [40, 158, 178] and appendix 7.A.1)

$$C_U(t) \xrightarrow{t \to \infty} \frac{2 k_B T}{3 \rho_0} \left(4\pi \frac{\eta_2 |t|}{\rho_0}\right)^{-3/2}. \quad (7.1)$$

For a no-slip colloid, $\gamma_h$ is unaffected by angular momentum conservation and Stokes’ law (see publication [B] and eq. (7.44))

$$\gamma_h = 6\pi \eta R \quad (7.2)$$

applies. In contrast, for a colloid with a slip boundary condition, the classical result $\gamma_h = 4\pi \eta_2 R$ is modified, and the friction coefficient becomes

$$\gamma_h = 6\pi \eta_2 R \frac{\eta_1 + \eta_2}{\eta_1 + 2\eta_2} \quad (7.3)$$

for $\eta_1 \neq \eta_2$ (see publication [B] and eq. (7.41)). For MPC-SRD-a, eq. (7.3) reduces to $\gamma_h \approx 4\pi \eta R$ for $\eta^c \ll \eta^t$, and to $\gamma_h \approx 3\pi \eta R$ in the typical case $\eta^c \gg \eta^t$. In contrast, the stress tensor is symmetric for MPC-SRD+a, i.e., $\eta_1 = \eta_2$, and the classical result $\gamma_h = 4\pi \eta R$ is recovered”\([C]\). The zero-frequency limit of the rotational friction coefficient is (see appendix 7.A.2)

$$\xi_h = 8\pi \eta R^3. \quad (7.4)$$

for no-slip colloids. For slip colloids $\xi_h = 0$, i.e., no hydrodynamic drag torque opposes the colloid’s rotation.

**Local friction**  “At short times, the molecular chaos assumption applies for the MPC fluid [158, 171], which yields $C_K(t) = 0 \ \forall t > 0$. In the time continuum this implies that $C_K(t)$ is proportional to a delta distribution and therefore $\hat{C}_K(\omega) = \hat{C}_K(\omega = 0)$. By means of eq. (3.12), we find $\hat{\gamma}(\omega) = \hat{\gamma}(\omega = 0) \equiv \gamma_l$ and the typical Langevin equation is obtained, which yields

$$C_U(t) = C_U(0) \exp(-\gamma_l t/M). \quad (7.5)$$

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We expect to observe this behavior in MPC simulations only approximately and for very short times. Since MPC is a discrete-time random process and \( C_K(t) = 0 \) \( \forall t > 0 \), eq. (3.13) yields

\[
\gamma_l = \frac{h}{2k_BT}C_K(0).
\]  

This equation provides a simple relation to measure the local friction coefficient \( \gamma_l \) [180].

In order to find an analytical expression for \( \gamma_l \), we have to evaluate various contributions to \( C_K(0) \). The random force \( \mathbf{K} = \mathbf{K}^k + \mathbf{K}^c \) comprises the contributions \( \mathbf{K}^k \) due to collisions with MPC particles in the streaming step and \( \mathbf{K}^c \) due to the change of the ghost-particle momenta during the collision step [180]. Hence, the autocorrelation functions \( \langle \mathbf{K}^k \cdot \mathbf{K}^k \rangle \) and \( \langle \mathbf{K}^c \cdot \mathbf{K}^c \rangle \), as well as the cross-correlation function \( \langle \mathbf{K}^k \cdot \mathbf{K}^c \rangle \) contribute to \( C_K(0) \). We denote the friction by the correlation \( \langle \mathbf{K}^k \cdot \mathbf{K}^k \rangle \) as Enskog friction \( \gamma_E \). When no ghost particles are present, as is the case for slip boundary conditions, \( \mathbf{K}^c = 0 \) and \( \gamma_l = \gamma_E \). A derivation for \( \gamma_E \) is presented in appendix 7.B, which yields the result previously established in ref. [126]a)

\[
\gamma_E = \frac{8}{3} \sqrt{2\pi k_B T \mu n R^2} \frac{1 + (2 - \Gamma)\chi M/\mu}{1 + \chi M/\mu}. 
\]  

Here, \( n = \rho_0/m \) is the particle density. We could not find analytical expressions for \( \langle \mathbf{K}^c \cdot \mathbf{K}^c \rangle \) and \( \langle \mathbf{K}^c \cdot \mathbf{K}^k \rangle \). Therefore, we measure \( \gamma_l \) according to eq. (7.6) in presence of ghost particles.

For the rotational motion, the treatment is analogous. The local and Enskog friction are”[C] (see appendix 7.B)

\[
\xi_l = \frac{h}{2k_BT}C_N(0), 
\]

\[
\xi_E = (1 - \Gamma)\frac{8}{3} \sqrt{2\pi k_B T \mu n R^4} \frac{M\chi}{\mu + M\chi}. 
\]  

### 7.3 Simulations

For the simulation of a colloid embedded in the MPC fluid, we employ the method outlined in section 4.3.2, with energy conserving collisions and computation of collision times, i.e., step (iii-a) instead of (iii) in section 4.3.2. The moment of inertia tensor of a sphere is \( \mathbf{I} = (2/5)MR^2 \mathbf{1} \). “MPC simulations are performed with the rotation angle \( \alpha = 130^\circ \) and the mean number of particles per collision cell \( \langle N_c \rangle = 10 \), which corresponds to the equilibrium density \( \rho_0 = \langle N_c \rangle m/a^3 \). We focus

a) We remind the reader of the definitions \( \chi = 2/5 \) and \( \mu = mM/(M + m) \) as well as \( \Gamma = 0 \) for no-slip and \( \Gamma = 1 \) for slip boundary conditions from section 4.3.2.
7.3 Simulations

Figure 7.1: “(a) Force autocorrelation function (FACF) of a slip colloid in a MPC-SRD-a fluid (blue solid line). The inset shows the FACF on a linear scale; the FACF becomes negative at $t = 0.2 \sqrt{ma^2/(k_B T)}$. (b) FACF of a no-slip colloid in a MPC-SRD+a fluid. In both cases, the radius of the colloid is $R/a = 6$. Note that the prediction by hydrodynamic theory (red, dashed line) diverges for $t \to 0$. Therefore, both curves are normalized by the simulation value for $t = 0$, denoted as $C_K(0)$.\textsuperscript{[C]} (Figures from publication [C])

on the liquid-like regime of the MPC fluid\textsuperscript{[158]} by choosing the collision time as $h/\sqrt{ma^2/(k_B T)} = 0.05$, which corresponds to a Schmidt number of approximately 100. We employ a cubic simulation box of length $L/a = 100$ with periodic boundary conditions if not otherwise stated. Since simulation results shall be compared to hydrodynamic theory, we require values of high accuracy for the viscosities $\eta$ and $\eta_r$ for our applied methods (MPC-SRD±a) and parameters. Measuring viscosities by nonequilibrium simulations using shear flow (see refs. [159, 167] and chapter 5) yields $\eta = 7.45 \sqrt{mk_B T/\rho_0}$ and $\eta_r = 5.40 \sqrt{mk_B T/\rho_0}$ for MPC-SRD+a, as well as $\eta_k = 0.3 \sqrt{mk_B T/\rho_0}$ and $\eta_c = 16.2 \sqrt{mk_B T/\rho_0}$ for MPC-SRD-a. In the following, we assume a neutrally buoyant colloid, i.e., $M = (4\pi/3)\rho_0 R^3$.

On the order of 100 independent simulations of $10^6$ time steps each were performed for a given parameter set to extract autocorrelation functions\textsuperscript{[C]}.

7.3.1 Autocorrelation functions

“In simulations, the velocity autocorrelation function is determined by measuring the velocity of a freely diffusing colloid in the MPC solvent. For the force autocorrelation function, we fix the colloid center at the origin of the reference frame by means of a constraining force $F^C(t)$. The random force follows then as $K(t) = -F^C(t)$ [122, 180], while $F^C$ is calculated as momentum transfer per time step $h$, with contributions due to collisions of MPC particles in the streaming step and due to the change of the ghost particles’ momenta during the collision step.
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Figure 7.2: “(a) VACF of a colloid with slip boundary conditions in a MPC-SRD-a fluid, and (b) with no-slip boundary conditions in a MPC-SRD+a fluid. The solid line (blue) corresponds to the simulation results, the dashed line (red) is the prediction by hydrodynamic theory, and the dotted line (green) is calculated according to eq. (7.5). The dashed-dotted line (magenta) in (b) is the simulation result without ghost particles. The insets display the first few time steps in a semi-logarithmic representation. In both cases, the radius of the colloid is $R/a = 6$”[C]. (Figures from publication [C])

Figure 7.3: “VACFs of colloids with slip boundary conditions in a MPC-SRD-a and a MPC-SRD+a fluid. The upper curves (red and blue) correspond to MPC-SRD-a with $h/\sqrt{ma^2/(k_BT)} = 0.12$, and the lower ones (black and green) to MPC-SRD+a with $h/\sqrt{ma^2/(k_BT)} = 0.05$. The colloid radius is $R/a = 6$ and the viscosity $\eta = 7.45\sqrt{mk_BT/a^2}$. The theoretical curves (dashed) are calculated by means of eqs. (3.7) and (7.36) with the appropriate viscosities”[C]. (Figure from publication [C])
Figure 7.1 shows two typical force autocorrelation functions. In case of the slip colloid, the FACF at \( t = 0 \) matches the expected Enskog value \( 2k_B T \gamma E / h \) (see eq. (7.6) or appendix 7.B) very well. However, for the no-slip colloid, \( C_K(0) \) is only captured by Enskog theory if ghost particles are neglected, as can be expected for non-zero correlations \( \langle K^c \cdot K^c \rangle \). Taking into account the momentum transfer due to ghost particles, we obtain a significantly larger value of \( C_K(0) \), and hence, of the local friction coefficient.

Within the molecular chaos assumption, the FACF is zero for all \( t > 0 \), which implies an exponentially decaying VACF. In fact, in our MPC simulation, the FACF decreases substantially after one time step, but instead of zero, it assumes about 10\% of its initial value for the parameters of fig. 7.1 (a). This explains the approximate exponential decay of the VACF for the first few time steps, as reported in refs. [122, 124, 126, 127]. As fig. 7.1 shows, the FACF is rather noisy, and an average over many independent realizations is required to achieve a smooth curve on long time scales.

The corresponding velocity autocorrelation functions are much smoother, as revealed by fig. 7.2. At \( t = 0 \), \( C_U(t = 0) = k_B T / M \), as expected. For short times \( t > 0 \), the simulation data slightly exceed the theoretical prediction until the hydrodynamic regime is reached. For longer times, we observe the long-time tail (see eq. (7.1)). The oscillations visible in fig. 7.2 for long times originate from sound modes and are a consequence of the finite compressibility of the MPC fluid combined with the periodic boundary conditions [171]. This also leads to an exponential decay of the correlation function on long time scales [171]. We determine the VACF for colloids of radii \( R/a = 1, 2, \ldots, 8 \), and find that for no-slip boundary conditions the VACF follows the hydrodynamic prediction well for radii \( R \geq 2a \), while for slip colloids, \( R \geq 3a \) is required. A detailed analysis of the VACF at \( t = 0 \) shows a small deviation from the equipartition value \( C_U(0) = k_B T / M \), which vanishes with increasing colloid size. For a slip colloid of radius \( R = 3a \) in a MPC-SRD+a fluid, \( C_U(0) \) is just 1\% larger than the expected value, while for no-slip colloids the theoretical value is exceeded by 5\% for \( R = 2a \) and by 2\% for \( R = 3a \). Working with \( R/a \geq 3 \), these values are acceptable, and we do not see a broadened Maxwell-Boltzmann distribution due to ghost particles as reported in ref. [126]. A disregard of ghost particles, as suggested in ref [126], leads to strong deviations between VACF obtained in simulations and from hydrodynamic theory for \( t > 0 \) as shown by fig. 7.2 (b). Hence, ghost particles are essential to obtain a good representation of no-slip boundary conditions. To elucidate the relevance of angular momentum conservation on the VACF, we perform simulations of a slip colloid in MPC-SRD+a with the collision step \( h/\sqrt{ma^2/(k_B T)} = 0.05 \) and in MPC-SRD-a with \( h/\sqrt{ma^2/(k_B T)} = 0.12 \), respectively. By this choice, both fluids possess approximately the same shear viscosity \( \eta = 7.45\sqrt{mk_B T / a^2} \). As fig. 7.3 shows, the velocity autocorrelation functions deviate from each other, but both are well described by hydrodynamic theory with the appropriate stress tensor. The colloid VACF for the MPC-SRD-a fluid is consis-
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Figure 7.4: “(a) Angular velocity autocorrelation function and (b) torque autocorrelation function for a colloid with no-slip boundary conditions in a MPC-SRD+a fluid. The colloid radius is \( R/a = 6 \) and the fluid viscosity \( \eta = 7.45 \sqrt{m k_B T/a^2} \). The box sizes are \( L = 100a \) (a) and \( L = 60a \) (b), respectively. The inset in (a) shows the first few time steps on a semilogarithmic scale; that in (b) presents the torque autocorrelation function on a linear scale. The torque autocorrelation function in (b) reaches its \( t^{-5/2} \) long-time tail at \( t/\sqrt{ma^2/(k_B T)} \gtrsim 10^2 \) which is not visible in the plotted time domain. Since the theoretical hydrodynamic-torque autocorrelation function (dashed, red) diverges for \( t \to 0 \), both curves are normalized by the simulation value at \( t = 0 \), denoted as \( C_N(0) \) [C]. (Figures from publication [C])

7.3.2 Diffusion coefficient

“As is well known, the diffusion coefficient of a particle in a system with periodic boundary conditions is system-size dependent [43, 124, 181–184]. In order to find the asymptotic diffusion coefficient for an infinite system \( (L \to \infty) \), we integrate the simulation data for the VACF from \( t = 0 \) to \( t_0 > 0 \) and subsequently integrate the theoretical correlation function from \( t = t_0 \) to \( t \to \infty \) [40, 43]. Hereby, \( t_0 \) has been chosen such that hydrodynamic theory applies for \( t > t_0 \). This procedure is
tently larger than that for the MPC-SRD+a fluid, which corresponds to a reduced friction of the MPC-SRD-a fluid (see eq. (7.3)). This reconfirms the result of publication [B] that the friction of slip colloids is reduced for non-angular-momentum conserving MPC fluids.

Finally, fig. 7.4 displays simulation results for the angular-velocity-autocorrelation function as well as the torque-autocorrelation function for a no-slip colloid, which both agree well with hydrodynamic theory. Similar to the force autocorrelation, the torque autocorrelation function is very noisy” [C].
Figure 7.5: “Integrals according to eq. (7.10) of the VACF of a no-slip colloid of radius $R = 6a$ in a MPC-SRD+\(a\) fluid for the simulation box sizes $L/a = 40$ (blue), 50 (green), 60 (red), 80 (cyan) (bottom to top). The dashed line (black) indicates the integration of the combined VACF from simulation up to $t = t_0 \approx 40\sqrt{ma^2/(k_BT)}$ and the theoretical expression of hydrodynamics beyond. The horizontal line (black) marks the diffusion coefficient from hydrodynamic theory”[C]. (Figure from publication [C])

illustrated in fig. 7.5, where the expression

\[
D(t) = \int_0^t dt' C_U(t)
\]  

(7.10)

is displayed for various box sizes, and for a VACF comprised of the numerical results for $L/a = 80$ up to $t_0 = 40\sqrt{ma^2/(k_BT)}$ and the theoretical expression following from eq. (3.7) for longer times.

In this way, we determine the diffusion coefficients of slip and no-slip colloids for several radii. The results are presented in fig. 7.6. Evidently, the diffusion coefficients are in close agreement with the prediction by hydrodynamics at large colloid radii. However, for small radii, we observe certain deviations, which we attribute to the effect of local friction. In general, the simulation data are by far closer to the hydrodynamic diffusion coefficient $D_h$ than to the combination with the Enskog expression, i.e., $D_h + D_E$, where $D_h = k_B T/\gamma_h$ and $D_E = k_B T/\gamma_E$.

In the case of no-slip colloids, we have to compare $D_h$ to $D_h + D_l$, where $D_l = k_B T/\gamma_l$ accounts for local interactions. Note that $D_l$ is small compared to $D_h$, because $\gamma_l$ is very large. Our simulation results are about midway in-between the predictions $D_h$ and $D_h + D_l$. As a consequence, for both, slip and no-slip colloids, hydrodynamics dominates at large $R$ and the simulation results are well described by $D_h$”[C].
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Figure 7.6: “Diffusion coefficients as function of the radius of colloids with slip boundary conditions (a) in a MPC-SRD-a fluid, (b) in a MPC-SRD+a fluid, and (c) a colloid with no-slip boundary conditions in a MPC-SRD+a fluid. The solid lines (blue) represent $D_h$, the dotted lines (green) $D_h + D_E$, and the dashed lines (orange) $D_h + D_l$. Open circles (black) correspond to values extracted from the VACF (cf. sec. 7.3.2), while the triangles (red) are obtained by integration of the FACF"[C]. (Figures adapted from publication [C])

7.3.3 Colloid sedimentation and diffusion coefficient

“We can extract finite-system-size diffusion coefficients $D_L$ by the plateau values in fig. 7.5. Complementary, we perform simulations of sedimenting colloids by constantly accelerating a colloid with the acceleration $g = 10^{-3}k_B T/(ma)$. At the same time, the fluid experiences the acceleration $g_f = -g/(\rho_0 U^3/M - 1)$, such that the center of mass of the total system remains at rest. For the selected values, the Reynolds number is approximately 0.1. When a steady state velocity $u$ is reached, we obtain the finite-system-size friction coefficient as $\gamma_L = Mg/u$, and the diffusion coefficient as $D_L = k_B T/\gamma_L$ (Einstein-Sutherland relation).

For a hydrodynamic solvent, a relation between $D_L$ and the diffusion coefficient
Figure 7.7: “Finite-system-size diffusion coefficients $D_L$ of colloids with (a) no-slip boundary conditions in a MPC-SRD+a and (b) slip boundary conditions in a MPC-SRD-a fluid. The colloid radius is $R/a = 6$. The open circles correspond to the plateau values of fig. 7.5. The values indicated by crosses (red) are obtained by sedimentation simulations. The lines follow from eq. (7.11), where we insert $D = D_h + D_l$ for the dashed orange line, $D = D_h$ for the blue solid line, and we substitute $D$ by the value obtained from the integral extrapolation approach of section 7.3.2 for the black dotted line” [C]. (Figures from publication [C])

of an infinite system $D$ has been provided in ref. [182] to first order in $R/L$,

$$D_L = D - \frac{2.837k_B T}{6\pi \eta L}.$$  \hspace{1cm} (7.11)

Since this expression is derived for point particles, the finite-size correction should hold for both slip and no-slip colloids. We consider colloids of radius $R/a = 6$, for which hydrodynamics should dominate in our simulations, and thus, eq. (7.11) should apply.

Results for $D_L$ are presented in fig. 7.7 together with the theoretical expression eq. (7.11) for various systems sizes $L$. Our values obtained by sedimentation are only about 2% higher than those determined via the VACF, which is an excellent agreement taking the accuracy of our methods into account.

We return to the question whether diffusivity is additive, i.e., $D = D_h + D_l$. According to the sedimentation data for no-slip colloids, $D = D_h + D_l$ seems to be a decent approximation, with an error of about 2%. For slip colloids, however, the prediction $D = D_h + D_l$ exceeds the measured values by about 30%. Hence, for slip colloids the diffusivities are not additive in MPC”[C].

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7.4 Summary and conclusions

“We have determined velocity and force autocorrelation functions of rigid spherical colloids dispersed in a MPC fluid. Both, slip and no-slip boundary conditions on the colloid surface have been considered. For slip boundary conditions, MPC fluids with (MPC-SRD+a) and without (MPC-SRD-a) angular-momentum conservation have been employed. In contrast, for no-slip boundary conditions only a MPC-SRD+a fluid has been utilized, because the coupling between the colloidal rotational degrees of freedom and the fluid requires a proper angular momentum transfer. We have verified the reduction of hydrodynamic friction of colloids with slip boundary conditions in a MPC-SRD-a fluid compared to the respective Stokes law. As derived in publication [B], the friction coefficient of such a colloid is given by an expression (eq. (7.41)), which reduces to \( \gamma_h \approx \frac{3 \pi \eta R}{\eta_c} \) for \( \eta_c \gg \eta_k \), in contrast to the classical result \( \gamma_h = \frac{4 \pi \eta R}{M} \) for an angular-momentum-conserving fluid. We like to stress that differences in the transport coefficients of angular- and non-angular-momentum-conserving fluids are a general problem of fluid simulations and not particular to MPC.

By measuring correlation functions for no-slip colloids, we found that ghost fluid particles inside the colloid are essential for a proper hydrodynamic colloid-solvent coupling. We did not observe a broadening of the Maxwell-Boltzmann distribution due to ghost particles, as discussed in ref. [126], i.e., a notably deviation of \( C_U(0) \) from \( k_B T/M \). Hence, ghost particles are essential to properly describe the dynamics of colloids with no-slip boundary conditions.

In addition, we have extracted diffusion coefficients from correlation functions and indirectly via colloid sedimentation. Most importantly, our simulations clearly show that the colloid diffusion coefficient is dominated by hydrodynamics in the parameter regime where MPC is liquid-like, i.e., for \( \eta_c \gg \eta_k \). Local friction yields only a minor contribution to the overall diffusion coefficient, and disappears with increasing colloid radius. Thereby, for no-slip colloids, the sum of the local and hydrodynamic diffusion coefficients is an acceptable approximation for the total diffusivity. This sum slightly overestimates the simulation result, but the deviation disappears with increasing colloid radius, and the Stokes-Einstein relation is satisfied. We have confirmed the result for the time steps \( h/\sqrt{ma^2/(k_B T)} = 0.1, 0.02 \). Thereby, all considered collision-time steps are well within the liquid regime of MPC-SRD [158]. However, for colloids with slip boundary conditions, the total diffusion coefficient is not given by the sum of the hydrodynamic and local diffusion coefficients, regardless of angular-momentum conservation. The combination of both significantly overestimates the diffusivity. As for other colloids, the simulation result is well described by the hydrodynamic diffusivity.

In the literature, colloids with slip boundary conditions have also been modeled by finite-range (steep) central potentials between colloids and MPC fluid particles [122, 124]. For infinitely steep interaction potentials, this approach should be equivalent to
7.A Frequency-dependent hydrodynamic friction

the method of specular reflection applied in this paper. For a finite interaction range, the interpretation of simulation results is less straight-forward, since an effective hydrodynamic radius has to be introduced as a fit parameter. But also in this case, we expect the non-additivity of diffusion coefficients to prevail"[C].

In this study, we chose step (iii-a) instead of step (iii) for the implementation of the colloid, i.e., we computed the collision times between fluid particles and the colloid instead of assuming collisions to occur at \( t + h/2 \) (see section 4.3.2). For the small time-steps applied here, we verified in test simulations that equally accurate correlation functions are obtained with step (iii). Furthermore, one can safely apply the momentum transfer specified in eqs. (4.23) and (4.24), instead of the energy-conserving momentum transfer of eq. (4.35), since the numerical difference between the expressions is negligible for buoyant colloids with \( R \geq 3a \) considered here, and since a thermostat is applied.

“Our studies clearly underline the dominance of hydrodynamic interactions in colloid diffusion. Even for moderately large colloids, hydrodynamics dominates already. The necessity of a local friction contribution in previous studies for colloids with slip boundary conditions may partially originate from an overestimation of the hydrodynamic friction coefficient by assuming the applicability of the Stokes law. The actual coefficient is up to a factor of 3/4 smaller, and hence the diffusion coefficient correspondingly larger"[C].

Appendix 7.A Frequency-dependent hydrodynamic friction

7.A.1 Translational friction

“In the following, we derive expressions for the friction coefficient \( \dot{\gamma}(\omega) \) of a spherical colloid with slip or no-slip boundary conditions moving with velocity \( \mathbf{U}(t) = U(t)(0,0,1)^T \) in a fluid, with a stress tensor specified by eq. (4.47). Thereby, we follow the derivation of ref. [179]. The Navier-Stokes equation

\[
\rho \left( \frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) = \nabla \cdot \mathbf{\sigma}, \tag{7.12}
\]

together with the continuity equation, can be linearized in the velocity \( \mathbf{v}(r,t) \), the pressure, and density fluctuations (see ref. [173] and appendix 5.A), which yields in the frequency domain

\[
-i\omega \rho_0 \tilde{\mathbf{v}} = -\nabla \tilde{p} + \eta_2 \Delta \tilde{\mathbf{v}} + (\eta_1 + \eta_3) \nabla (\nabla \cdot \tilde{\mathbf{v}}), \tag{7.13}
\]

\[
i\omega \tilde{\mathbf{p}} = \rho_0 (\nabla \cdot \tilde{\mathbf{v}}), \tag{7.14}
\]

\[
\tilde{\mathbf{p}} = \rho c^2. \tag{7.15}
\]
Equation (7.15) is the linearized relation between pressure and density, i.e., \( c^2 = \frac{\partial p}{\partial \rho} \), where \( c \) is either the adiabatic or isothermal sound velocity. MPC obeys the ideal-gas equation of state [167], and in simulations we apply the MBS-thermostat, hence \( c = \sqrt{k_B T/m} \). The pressure can be eliminated from eq. (7.13) by means of eqs. (7.14) and (7.15), which yields

\[
-i\omega \rho_0 \hat{v} = -\eta_2 \nabla \times (\nabla \times \hat{v}) + \frac{i\omega \rho_0}{\beta^2} \nabla (\nabla \cdot \hat{v}),
\]

(7.16)

where we define

\[
\beta^2 = \omega^2 \left( c^2 - \frac{i\omega(\eta_1 + \eta_2 + \eta_3)}{\rho_0} \right)^{-1}, \quad \text{Im}\{\beta\} > 0.
\]

(7.17)

With the Helmholtz decomposition

\[ \hat{v} = \nabla \phi + \nabla \times A, \]

(7.18)

eq. (7.16) becomes

\[
\Delta \phi + \beta^2 \phi = 0,
\]

(7.19)

\[
\nabla \times (\nabla \times A) = \alpha^2 A,
\]

(7.20)

with \( \alpha^2 = i\omega \rho_0/\eta_2 \), \( \text{Im}\{\alpha\} > 0 \)\[\textbf{C}\]. In terms of spherical coordinates \((r, \theta, \varphi)\), the Ansatzb) \( \phi = \hat{u} \cos(\theta) h(r) \) and \( A = \hat{U} \times \nabla f(r) \), yields the ordinary differential equations

\[
0 = \frac{\partial^2 h}{\partial r^2} + 2 \frac{\partial h}{r \partial r} + h(r) \left( \beta^2 - \frac{2}{r^2} \right),
\]

(7.21)

\[
0 = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial f}{\partial r} \right) + \alpha^2 f(r),
\]

(7.22)

which are solved by

\[
h(r) = (r^{-2} - i\beta r^{-1})e^{i\beta r},
\]

(7.23)

\[
f(r) = r^{-1}e^{i\alpha r}.
\]

(7.24)

We construct a solution as the linear combination

\[ \hat{v} = c_2 \nabla \phi - c_1 \nabla \times A, \]

(7.25)

\[ \text{b) Note that } \phi \text{ is a scalar that should depend linearly on } \hat{U}. \text{ Hence, we assume } \phi \text{ to be proportional to the scalar product of } \hat{U} \text{ and } r, \text{ i.e., } \phi = \hat{u} \cos(\theta) h(r). \text{ The vector } A \text{ should also be linear in } \hat{U}, \text{ and since } \hat{v} \text{ is a polar vector, } A \text{ has to be an axial vector. This motivates the cross product } A = \hat{U} \times \nabla f(r) \text{ [62]\[\textbf{C}].} \]
7.A Frequency-dependent hydrodynamic friction

which yields after some manipulations

\[ v_r = u \cos(\theta) \left\{ 2c_1 e^{i\alpha r} \left( -\frac{1}{r^3} + \frac{i \alpha}{r^2} \right) + c_2 e^{i\beta r} \left( -\frac{2}{r^3} + \frac{2i \beta}{r^2} + \frac{\beta^2}{r^2} \right) \right\}, \tag{7.26} \]

\[ v_\theta = u \sin(\theta) \left\{ c_1 e^{i\alpha r} \left( -\frac{1}{r^3} + \frac{i \alpha}{r^2} + \frac{\alpha^2}{r} \right) + c_2 e^{i\beta r} \left( -\frac{1}{r^3} + \frac{i \beta}{r^2} \right) \right\}, \tag{7.27} \]

and \( v_\phi = 0 \). The coefficients \( c_1 \) and \( c_2 \) are determined by slip boundary conditions,

\[ \hat{v}_r (r = R) = \hat{u} \cos(\theta), \tag{7.28} \]

\[ \hat{\sigma}_{\theta r} (r = R) = 0, \tag{7.29} \]

or no-slip boundary conditions

\[ \hat{v}(r = R) = \hat{U}. \tag{7.30} \]

Note that for eqs. (7.28)–(7.30), we chose a reference frame, in which the origin always lies in the colloid’s center \([185] \)\([C]\). The hydrodynamic force \( \mathbf{F}(\omega) \) acting on the colloid can be evaluated by the surface integral (see eq. (2.2))

\[ \mathbf{F} = \int_{r=R} dA \, \hat{\sigma} \, \mathbf{e}_r. \tag{7.31} \]

To evaluate the integrand, we apply the partition of unity, i.e., \( 1 = \sum_\alpha e_\alpha e_\alpha^T \), yielding

\[ \sigma = \sum_{\alpha, \beta \in \{r, \theta, \phi\}} e_\alpha e_\alpha^T \sigma e_\beta e_\beta^T = \sum_{\alpha, \beta \in \{r, \theta, \phi\}} \sigma_{\alpha \beta} e_\alpha e_\beta^T. \tag{7.32} \]

Using the symmetry with respect to \( \phi \), we find after some algebra

\[ \mathbf{F} = \int_{r=R} dA \left( -\sin(\theta) \hat{\sigma}_{\theta r} + \cos(\theta) \hat{\sigma}_{rr} \right) (0, 0, 1)^T, \tag{7.33} \]

where \( \hat{\sigma}_{\theta r} \) and \( \hat{\sigma}_{rr} \) follow by eqs. (4.47), (7.14), and (7.15) as (see chapter 6 for a similar calculation in cylindrical coordinates)

\[ \hat{\sigma}_{\theta r} = \eta_1 \left( \frac{1}{r} \frac{\partial \hat{v}_r}{\partial \theta} - \frac{\hat{v}_\theta}{r} \right) + \eta_2 \frac{\partial \hat{v}_\theta}{\partial r}, \tag{7.34} \]

\[ \hat{\sigma}_{rr} = \left( \eta_3 - \frac{\rho_0 c^2}{i \omega} \right) \nabla \cdot \hat{\mathbf{v}} + \left( \eta_1 + \eta_2 \right) \frac{\partial \hat{v}_r}{\partial r}. \tag{7.35} \]

Here, we expressed the pressure as \( p = \rho_0 c^2 \nabla \cdot \mathbf{v} / (i \omega) \), which follows from eqs. (7.14) and (7.15). With the definition of \( \hat{\gamma}(\omega) \) by \( \mathbf{F} = -\hat{\gamma}(\omega) \hat{U} \), eq. (7.31) yields

\[ \hat{\gamma}(\omega) = -\frac{4\pi}{3} \eta_2 Rx^2 \left[ Q(1 - y) + 2P(x - 1) \right], \tag{7.36} \]

with the abbreviations

\[ x = i \alpha R, \quad y = i \beta R, \quad P = \frac{c_1}{R^3} e^{i\alpha R}, \quad Q = \frac{c_2}{R^3} e^{i\beta R}. \tag{7.37} \]
**Chapter 7 From local to hydrodynamic friction in Brownian motion: A MPC study**

**Slip boundaries:** Determining the coefficients $c_1$ and $c_2$ by the slip boundary conditions (7.28) and (7.29), we find

\[
P = (\eta_1 + \eta_2)(y^2 - 3y + 3)/\Delta,
\]
\[
Q = \left[\eta_1(-3 + 3x - x^2) + \eta_2(-3 + 3x - 2x^2 + x^3)\right]/\Delta,
\]
\[
\Delta = (\eta_2 x^3 - (\eta_1 + 2\eta_2) x^2) (-2 + 2y - y^2) + (\eta_1 + \eta_2)(1 - x)y^2.
\]

From eqs. (3.12) and (7.36), and the analog of Watson’s lemma for Fourier transformations [186], we find the asymptotic long-time behavior of the random-force autocorrelation function

\[
C_K(t) \xrightarrow{t \to \infty} -3\pi \eta_2 R^2 \frac{\eta_1 + \eta_2}{(\eta_1 + 2\eta_2)^2} \sqrt{\frac{\rho_0}{\pi \eta_2}} |t|^{-3/2}.
\]

Similarly, the long-time tail of the velocity autocorrelation function follows as [185]

\[
C_U(t) \xrightarrow{t \to \infty} 2\frac{\eta_2}{3\rho_0} \left(4\pi \frac{\eta_2}{\rho_0} |t|\right)^{-3/2}.
\]

In the Stokes limit $\beta \to 0$, $\alpha \to 0$, we obtain the friction coefficient $\hat{\gamma}(\omega = 0) = 6\pi \eta_2 R \frac{\eta_1 + \eta_2}{\eta_1 + 2\eta_2}$, consistent with the result of publication [B].

**No-slip boundaries:** Determining the coefficients $c_1$ and $c_2$ by the no-slip boundary conditions (7.30), we find

\[
P = (y^2 - 3y + 3)/\Delta,
\]
\[
Q = -(x^2 - 3x + 3)/\Delta,
\]
\[
\Delta = x^2 y^2 - y^2 x - 2x^2 y + y^2 + 2x^2,
\]

and for the long-time tail of the random-force autocorrelation function

\[
C_K(t) \xrightarrow{t \to \infty} -3\pi \eta_2 R^2 \sqrt{\frac{\rho_0}{\pi \eta_2}} |t|^{-3/2},
\]

while the long-time tail of the velocity autocorrelation function is the same as for slip boundary conditions $\hat{\gamma}(\omega = 0) = 6\pi \eta_2 R$. In the Stokes limit $\beta \to 0$, $\alpha \to 0$, we obtain the friction coefficient

\[
\hat{\gamma}(\omega = 0) = 6\pi \eta_2 R.
\]
7.A Frequency-dependent hydrodynamic friction

7.A.2 Rotational friction

For a rotating sphere, the no-slip boundary condition is \( \hat{v}(r = R) = \hat{\Omega} \times r \). Without loss of generality, we assume \( \hat{\Omega} = \hat{\Omega} \hat{e}_z \). “Since the rotational motion of the no-slip colloid should not excite longitudinal sound modes, we can restrict ourselves to an incompressible description of the fluid [187]”[C]. The linearized incompressible Navier-Stokes equations read

\[
-i \omega \rho_0 \hat{v} = \eta \nabla \Delta \hat{v} - \nabla p, \tag{7.45}
\]

\[
\nabla \cdot \hat{v} = 0. \tag{7.46}
\]

We can apply the curl operator to eq. (7.45) to eliminate the pressure

\[
\nabla \times \left[ \Delta \hat{v} + \alpha^2 \hat{v} \right] = 0. \tag{7.47}
\]

Here, \( \alpha^2 = i \omega \rho_0 / \eta^2 \), \( \text{Im}\{\alpha\} > 0 \), as before. We employ the Ansatz\(^c\)

\[
\hat{v} = \nabla \times A, \tag{7.48}
\]

with \( A = f(r) \hat{\Omega} \), and obtain

\[
0 = \nabla \times \left[ \left( \Delta + \alpha^2 \right) \nabla f \times \hat{\Omega} \right]. \tag{7.49}
\]

The above equation is satisfied for \( \left( \Delta + \alpha^2 \right) f = 0 \), which again can be solved by \( f(r) = c_1 r^{-1} \exp \left( i \alpha r \right) \) [62]. For the velocity field, we obtain

\[
\hat{v}(r) = c_1 e^{i \alpha r} \left( \frac{i \alpha}{r^2} - \frac{1}{r^3} \right) r \times \hat{\Omega}. \tag{7.50}
\]

The no-slip boundary condition \( \hat{v}(r = R) = \hat{\Omega} \times r \) determines \( c_1 \). Hence,

\[
\hat{v}(r) = e^{i \alpha (r - R)} \frac{1 - i \alpha r}{1 - i \alpha R} \frac{R^3}{r^3} \hat{\Omega} \times r. \tag{7.51}
\]

The torque can be evaluated as \( \hat{T} = \int dA \ r \times \hat{\sigma} \ e_r \). Due to symmetry, we expect no torque in \( x \)- and \( y \)-direction. For the \( z \)-component, we obtain

\[
e_z \cdot \hat{T} = \int dA \ r e_z \cdot \left[ e_r \times \hat{\sigma} \ e_r \right] = \int dA \ r \left[ e_z \times e_r \right] \cdot \hat{\sigma} \ e_r \tag{7.52}
\]

\[
= \int dA \ r \sin(\theta) e_z^T \hat{\sigma} \ e_r = \int_0^\pi d\theta \int_0^{2\pi} d\varphi \ R^3 \sin^2(\theta) \hat{\sigma}_{\varphi r} \tag{7.53}
\]

\[
= 2\pi R^3 \int_0^\pi d\theta \ \sin^2(\theta) \hat{\sigma}_{\varphi r}. \tag{7.54}
\]

\(^c\) Since \( \hat{v} \) is a polar vector, \( A \) has to be an axial vector. But as \( \hat{\Omega} \) already is an axial vector, and \( \hat{v} \) should be linear in \( \hat{\Omega} \), the Ansatz \( A = f(r) \hat{\Omega} \) is well motivated [62]”[C].
We evaluate $\hat{\sigma}_{rr} = \eta (\partial \hat{v}_r / \partial r - \hat{v}_r / r)$ and utilize $\hat{\Omega} \times r = \hat{\Omega} \sin(\theta) \hat{e}_\varphi$. Finally, we obtain $\hat{T}_z = -\hat{\xi}(\omega) \hat{\Omega}$, with the rotational friction coefficient

$$\hat{\xi}(\omega) = \frac{8 \pi \eta_2 R^3 3 - 3x + x^2}{1 - x},$$

(7.55)

where $x = i\alpha R$. In the Stokes limit $\omega \to 0$, we obtain the friction constant $\xi_h = 8 \pi \eta R^3$.

Applying analogous calculations as for the translational case, we find the angular-velocity autocorrelation and the random-torque autocorrelation function

$$C_{\Omega}(t) \xrightarrow{t \to \infty} \frac{\pi}{\rho_0} \left( \frac{4 \pi \eta_2 |\Omega|}{\rho_0} \right)^{-5/2},$$

(7.56)

$$C_N(t) \xrightarrow{t \to \infty} -2 \sqrt{\frac{\pi \rho_0^3 R^6}{\eta_2}} |t|^{-5/2}.$$  

(7.57)

**Appendix 7.B Enskog friction**

“For short times, the force autocorrelation function of a colloid is determined by uncorrelated collisions with fluid particles (molecular chaos). Hence, during the first streaming and collision step, no hydrodynamic correlations build up. (We will neglect ghost particles for the following considerations.) Since force is change of momentum per time, the force $\mathbf{K}$ on the colloid during a streaming step is

$$\mathbf{K} = \frac{1}{h} \sum_{k=1}^{N} \mathbf{J}_k.$$  

(7.58)

Hence, the force-autocorrelation function at time $t = 0$ is

$$\langle \mathbf{K}(0)^2 \rangle = \frac{1}{h^2} \sum_{k=1}^{N} \sum_{l=1}^{N} \langle \mathbf{J}_k \cdot \mathbf{J}_l \rangle = \frac{1}{h^2} \left( \sum_{k=1}^{N} \mathbf{J}_k^2 \right),$$

(7.59)

within the molecular-chaos assumption. Instead of summing over particles, we can integrate the respective distribution over the colloid surface. Consider an infinitesimal surface element of the spherical colloid of area $dA = R^2 \sin(\theta) d\theta d\varphi$. During a time step, solvent particles with a relative velocity

$$\mathbf{v} = \mathbf{v} - \mathbf{U} - \hat{\Omega} \times \mathbf{R} n,$$  

(7.60)

which is negative in the normal direction, i.e., $\mathbf{v} \cdot n = \mathbf{v} n < 0$, collide with the colloid if they are located in the volume element $dV = -\mathbf{v} \cdot n dA$ (see fig. 7.8). With the
average particle density \( n = N/V \), we find for the number \( dN \) of colliding particles per surface element \( dA \)

\[
dN = -n \bar{v}_n h dA, \quad (7.61)
\]

and consequently for the force autocorrelation function

\[
\langle K(0)^2 \rangle = \frac{1}{h^2} \int dA \left\langle \frac{dN}{dA} J^2 \right\rangle. \quad (7.62)
\]

The dependence of \( J \) on \( \bar{v} \) is specified in eq. (4.35), and the average is defined as

\[
\langle \ldots \rangle = \int_{-\infty}^{0} d\bar{v}_n \int_{-\infty}^{\infty} d\bar{v}_\theta \int_{-\infty}^{\infty} d\bar{v}_\varphi P_n(\bar{v}_n) P_t(\bar{v}_\theta) P_t(\bar{v}_\varphi) \ldots, \quad (7.63)
\]

where \( \bar{v}_\theta \) and \( \bar{v}_\varphi \) are the components of the tangential velocity \( \bar{v}_t \) in \( \theta \) and \( \varphi \) direction"[C]. The probability distribution functions \( P_n \) and \( P_t \) are Gaussian, since the Cartesian components of \( v, U \), and \( \Omega \) are Gaussian distributed. “The variance of \( \bar{v}_n \) is \( \sigma_n^2 = k_B T/m + k_B T/M \), while the variance of \( \bar{v}_\theta \) as well as \( \bar{v}_\varphi \) is

\[
\sigma_t^2 = k_B T R^2/I + k_B T/m + k_B T/M, \quad \text{where} \quad I = \chi M R^2.
\]

Evaluation of the average yields

\[
\langle K(0)^2 \rangle = \frac{16}{h} \sqrt{2\pi (k_B T)^3} \mu n R^2 \frac{1 + (2 - \Gamma) \chi M/\mu}{1 + \chi M/\mu}. \quad (7.64)
\]

The friction coefficient, which we will denote as Enskog friction, follows by integration of the force autocorrelation function according to eq. (3.13). Since MPC is a discrete-time-random process, the integral is

\[
\int_0^\infty dt \langle K(t) \cdot K(0) \rangle = \frac{h}{2} \langle K(0)^2 \rangle + h \sum_{l=1}^\infty \langle K(lh) \cdot K(0) \rangle. \quad (7.65)
\]
Within the molecular chaos approximation, $\langle \mathbf{K}(l) \cdot \mathbf{K}(0) \rangle = 0$ for $l \neq 0$, and we find the Enskog friction coefficient

$$\gamma_E = \frac{8}{3} \sqrt{2\pi k_B T \mu n R^2 \frac{1 + (2 - \Gamma)\chi M/\mu}{1 + \chi M/\mu}}. \quad (7.66)$$

In the time continuum limit, the force autocorrelation function is a delta distribution

$$\langle \mathbf{K}(t) \cdot \mathbf{K}(0) \rangle = 6k_B T \gamma_E \delta(t), \quad (7.67)$$

which leads to the exponentially decaying velocity-autocorrelation function

$$C_U(t) = \frac{k_B T}{M} e^{-\gamma_E t/M}. \quad (7.68)$$

The rotational motion is treated in a similar manner$^*$[C]. For the torque autocorrelation function at time $t = 0$ we obtain

$$\langle \mathbf{N}(0)^2 \rangle = \int d\Omega \left\langle \frac{dN}{d\Omega} (R\mathbf{n} \times \mathbf{J})^2 \right\rangle / h^2 \quad (7.69)$$

$$= \frac{16nR^4}{h} \sqrt{2\pi (k_B T)^3 \mu(1 - \Gamma)} \frac{M\chi}{\mu + M\chi}, \quad (7.70)$$

and hence, for the rotational friction coefficient

$$\xi_E = \frac{8}{3} \sqrt{2\pi k_B T \mu n R^4 (1 - \Gamma)} \frac{M\chi}{\mu + M\chi}. \quad (7.71)$$
Chapter 8

Position fluctuations of a colloid in a harmonic trap near a plane wall

8.1 Introduction

Recent experimental progress allows for the measurement of a colloid’s Brownian motion with a time resolution of microseconds and a positional accuracy on the order of nanometers [49]. With this precision, it has been demonstrated that noise in Brownian motion is actually not white, but rather colored [39] (see section 3.2). This is a consequence of hydrodynamic memory, a feature also giving rise to the well known long-time tails, first observed in computer simulations in the 1970’s [178] (see chapter 7 and eq. (7.1)).

It has been argued that beyond the experiment of ref. [39], the observation of a colloidal particle’s Brownian motion could be exploited to deduce properties of its environment [49, 50]. In other words, a colloid could function as a sensor, with possible applications in microbiology [50]. Studying the motion of a colloid close to a wall or membrane, footprints of surface elasticity [51] or adsorption layers [49, 52] could be observed.

A recent experimental study of Jannasch et al. [53] takes the same line and suggests that the distance between a colloid in an optical trap and a plane wall can be inferred from the colloid’s spectrum of position fluctuations, i.e., its power spectral density (PSD). The measurement of the colloid-wall distance hereby relies on the fit of a theoretical expression for the PSD to the experimental data. Although the theoretical expression used by Jannasch et al. fits their experimental results very well, Felderhof argues in ref. [54] that its derivation is not convincing and proposes an alternative expression.

In such a case, a computer simulation is a promising approach to check the two proposed theoretical expressions for correctness. Since we showed in chapter 7 that the MPC method yields the correct Brownian motion of a colloid including hydrodynamic correlations, it is a reliable tool for this task.
Chapter 8 Position fluctuations of a colloid in a harmonic trap near a plane wall

8.2 Setup and theoretical predictions

We consider a spherical colloid in an incompressible fluid, which is confined by a planar wall to \( z > 0 \). The colloid is placed in an isotropic harmonic trap of stiffness \( k \) at position \( C_0 = (0, 0, d)^T \). Both, on the surface of the colloid as well as on the planar wall, no-slip boundary conditions apply. We measure the position parallel to the wall \( C_x(t) \equiv x(t) \), and perform a Fourier transformation to obtain \( \hat{x}(\omega) \). The fluctuation dissipation theorem demands

\[
\langle \hat{x}(\omega) \hat{x}^*(\omega') \rangle = \frac{1}{2\pi} \delta(\omega - \omega') \frac{2k_B T}{\omega} \text{Im} \{ \chi_{xx}(\omega) \},
\]

where \( \chi_{xx}(\omega) \) is the susceptibility, which determines the response \( \hat{x}(\omega) = \chi_{xx}(\omega)\hat{E}_x(\omega) \) to an external force \( \hat{E}_x(\omega) \) in \( x \)-direction. In eq. (8.1), the power spectral density \( S(\omega) \) can be identified as

\[
S(\omega) = \frac{2k_B T}{\omega} \text{Im} \{ \chi_{xx}(\omega) \}.
\]

References [53] and [54] propose different expressions for the susceptibility \( \chi_{xx}(\omega) \) and hence the PSD \( S(\omega) \).

- In ref. [54], Felderhof found the susceptibility

\[
\chi_{xx,F}(\omega) = \frac{1 + \gamma_0(1 + \alpha R + \frac{1}{3} \alpha^2 R^2) F_{xx}(\omega)}{k - (M - M_f)\omega^2 - \gamma_0(1 + \alpha R + \frac{1}{3} \alpha^2 R^2)(i\omega - kF_{xx}(\omega))},
\]

where \( \alpha^2 = -i\omega\rho_0/\eta, \text{Re}\{\alpha\} > 0, M_f = \rho_0(4\pi/3)R^3, \gamma_0 = 6\pi \eta R, \) and \( F_{xx}(\omega) \) is the reaction field tensor, given in eq. (3.9) of ref. [188]. Felderhof’s derivation is based on Faxén’s theorem (see appendix 8.A).

- In ref. [53], the susceptibility is effectively cast into the form

\[
\chi_{xx,J}(\omega) = \frac{1}{k - M\omega^2 - i\omega\zeta(\omega)},
\]

with a frequency dependent friction \( \zeta(\omega) \). Note that Jannasch et al. use the frequency \( f \) instead of \( \omega = 2\pi f \), denote the power spectral density by \( P(f) \) and the friction by \( \gamma(f, R/l) \) [53]. For comparison, \( S(\omega) = P(\omega/(2\pi)) \), \( \zeta(\omega) = \gamma(\omega/(2\pi), R/l) \), where \( \gamma(f, R/l) \) is given in eq. (2) of ref. [53].

8.3 Implementation of colloid and wall

The colloid and the planar wall are implemented according to section 4.3.2. Since we cannot represent a semi-infinite system in MPC, we implement two planar walls.
8.4 Simulations

We implement a neutrally buoyant colloid of radius \( R = 3a \) in MPC-SRD+a with \( h = 0.05 \sqrt{ma^2/(k_B T)} \), \( \bar{N}_c = 10 \) and rotation angle \( \alpha = 130^\circ \), yielding a viscosity of \( \eta = 7.45 \sqrt{mk_B T/a^4} \). MPC is a compressible fluid, but simulation results are to be compared to incompressible theory. Therefore, we choose \( \omega_k = \sqrt{k/M} \) rather small, so that the important features of the PSD occur at small frequencies, for which MPC is approximately incompressible. On the other hand, \( \omega_k \) is not chosen too small either, since strong traps were employed in the experiments and the colloid’s distance to the wall should be nearly constant. We set the trap stiffness to \( k = 60k_B T/a^2 \), yielding \( \omega_k \approx 0.23 \sqrt{k_B T/(ma^2)} \) and \( \omega_k h \approx 10^{-2} \ll 1 \). We determine \( \sqrt{\langle (C(t) - C_0)^2 \rangle / R} \) from simulations and find a value of 0.04, indicating a strong confinement.

Figure 8.1: PSD of the colloid in a trap without a wall. The solid black line represents the expression of ref. [54], while the dashed red line represents the expression of ref. [53]. The blue bullets represent simulation results.

and choose their distance so large, that the colloid effectively only interacts with one wall. To include the harmonic trap of stiffness \( k \) at position \( C_0 \), we employ the analytical solution for a particle in a harmonic trap in the streaming step

\[
C(t + h) = C_0 + (C(t) - C_0) \cos(\omega_k h) + \frac{U(t)}{\omega_k} \sin(\omega_k h),
\]

(8.5)

\[
U(t + h) = U(t) \cos(\omega_k h) - (C(t) - C_0)\omega_k \sin(\omega_k h),
\]

(8.6)

with the frequency \( \omega_k = \sqrt{k/M} \). We implement step (iii-a) instead of (iii) of section 4.3.2. Thereby, we expand eqs. (8.5) and (8.6) for small \( h \), i.e, \( \cos(\omega_k h) \approx 1 \) and \( \sin(\omega_k h) \approx \omega_k h \), to calculate collision times.
Figure 8.2: PSD of the colloid in a trap with a distance (a) $d = 3R$ and (b) $d = 2R$ to the wall. The solid black line represents the expression of ref. [54], while the dashed red line represents the expression of ref. [53]. The blue bullets represent simulation results.

For the data analysis, we block-average the measured discrete PSD, and normalize by its value at the smallest available frequency. The theoretical expressions are rescaled by a factor $s$, where $s$ is chosen for each expression such that the error between theory and simulation data is minimized for the first 10 frequency values.

In fig. 8.1, simulation results without wall are compared to the PSD of refs. [53] and [54] for the wall distance $d \to \infty$. In this limit, the different theoretical expressions coincide and match the data nicely. The resonance peak in the PSD reflects the fact that the noise experienced by the colloid is colored, which is a consequence of hydrodynamic memory [49]. For large frequencies, we observe a deviation of the data from theory, which has two reasons. First, compressibility effects becomes stronger, and second, for large frequencies, i.e., short times, hydrodynamic correlations have not yet developed in the MPC fluid.
In fig. 8.2, the influence of the wall on the PSD is shown. We can observe that the wall dampens the resonance peak. The simulation results are nicely described by Felderhof’s theoretical expression, while the expression utilized by Jannasch et al. shows strong deviations, especially for low frequencies. The agreement of Felderhof’s expression with the simulation data even for \(d = 2R\) indicates that the approximations in its derivation (see appendix 8.A) only weakly affect the result.

8.5 Conclusions

Our simulation study strongly supports Felderhof’s theoretical expression for the PSD. Felderhof found that the deviation of the PSD from its zero frequency value is first positive and then negative [54]. For the theoretical PSD used in the experimental study of Jannasch et al. [53], the deviation is first negative, then positive and finally negative again for typical cases. This behavior was also observed in the experiments. However, as Felderhof’s theory and our simulations suggest, this experimental observation will not be shown by a no-slip colloid above a no-slip wall. In ref. [189], Felderhof suggested that a dynamic partial-slip boundary condition at the wall could explain the experimental results. Here, further experimental work is desirable.

Appendix 8.A Felderhof’s derivation of the susceptibility

Here, we summarize Felderhof’s derivation for the susceptibility. Note that Franosch extended Felderhof’s framework to include the harmonic trap [49]. The equation of motion in frequency space of a spherical colloid subject to the external force \(E\) and the harmonic force \(-kC\) in an infinite fluid described by the linearized incompressible Navier-Stokes equation with a background flow \(v^*\), is given by Faxén’s law [49]

\[
-i\omega(M - M_f)\hat{U}(\omega) = -\tilde{\zeta}(\omega)(\hat{U}(\omega) - v^*) + \hat{E} - k\hat{C}(\omega). \tag{8.7}
\]

Here, \(M_f = (4\pi/3)\rho R^3\) is the mass of the displaced fluid and \(\tilde{\zeta}(\omega) = \gamma_0(1 + \alpha R + \alpha^2 R^2/3)\) with \(\alpha^2 = -i\omega\rho_0/\eta, \Re\{\alpha\} > 0\). We can use this expression to find the equation of motion for a colloid in a harmonic trap above a no-slip wall. We denote the flow field created by the colloid above a no-slip wall by \(v\) and the corresponding flow in an infinite fluid by \(v^\infty\). Considering the external flow \(v^* = v - v^\infty\), we can regard the problem of the colloid above the wall with solution \(v\) as equivalent to the problem of a colloid in an infinite fluid with background flow \(v^*\), since \(v = v^\infty + v^*\).a)

---

\(a^)\) The same argument was used in section 2.5.3 to find the induced (angular) velocity of a swimmer above a no-slip wall.
Hence, substituting \( \mathbf{v}^* = \mathbf{v} - \mathbf{v}^\infty \) into eq. (8.7), yields the equation of motion for the colloid above a wall. Finally, we approximate the velocity field created by the colloid \( \mathbf{v} \) as the flow field of a point force \( \mathbf{E} - k \mathbf{C} \) at position \( \mathbf{C}_0 \) acting on the fluid. Thus, \( \mathbf{v}(\mathbf{r}, \omega) = G(\mathbf{r}, \mathbf{C}_0, \omega)(\mathbf{E} - k \mathbf{C}) \) and \( \mathbf{v}^\infty(\mathbf{r}, \omega) = G^\infty(\mathbf{r} - \mathbf{C}_0, \omega)(\mathbf{E} - k \mathbf{C}) \), where \( G \) and \( G^\infty \) are the Green’s functions of the linearized incompressible Navier-Stokes equations in a system bounded by an infinite planar no-slip surface and in an infinite unbounded system, respectively. The velocity fields have to be evaluated at the position of the particle, which we approximate by \( \mathbf{C}_0 \). Then, we obtain

\[
\mathbf{v}^*(\mathbf{r} = \mathbf{C}_0) = \lim_{\mathbf{r} \to \mathbf{C}_0} [G(\mathbf{r}, \mathbf{C}_0, \omega) - G^\infty(\mathbf{r} - \mathbf{C}_0, \omega)] (\mathbf{E} - k \mathbf{C})
\]

(8.8)

\[
= F(\mathbf{C}_0, \omega)(\mathbf{E} - k \mathbf{C}),
\]

(8.9)

where we defined the reaction-field tensor \( F(\mathbf{C}_0, \omega) \). For a derivation of the explicit expression for \( F(\mathbf{C}_0, \omega) \), see ref. [188]. Using \( \mathbf{U}(\omega) = -i\omega \mathbf{C}(\omega) \), we can solve eq. (8.7) for \( \mathbf{C}(\omega) \), yielding

\[
\left[(k - (M - M_f)\omega^2 - \tilde{\zeta}(\omega)(i\omega - k F))\right] \mathbf{C}(\omega) = (\tilde{\zeta}(\omega)F + 1) \mathbf{E}(\omega).
\]

(8.10)

Since \( \chi_{xx} \) is defined by \( C_x(\omega) = \chi_{xx}(\omega)E_x(\omega) \) and \( F(\mathbf{C}_0, \omega) \) is diagonal [188], we obtain

\[
\chi_{xx}(\omega) = \frac{1 + \gamma_0(1 + \alpha R + \frac{1}{3}(\alpha^2 R^2)F_{xx}(\omega))}{k - (M - M_f)\omega^2 - \gamma_0(1 + \alpha R + \frac{1}{3}(\alpha^2 R^2)(i\omega - k F_{xx}(\omega)))},
\]

(8.11)

which is Felderhof’s result eq. (8.3).
Part IV

Microswimmers
Chapter 9

Modeling a spheroidal microswimmer

Most results of this chapter have been published in publication [D] (see List of publications on page 203). All verbatim quotes within this chapter are quotations of [D] and are indicated as “...”[D]. Such verbatim quotes can extend over several pages and have been reformatted to fit with the main text. The label of a cited article inside a verbatim quote follows the enumeration in the bibliography (page 186).

9.1 Introduction

“Generic models, which capture the essential swimming aspects, are crucial in theoretical studies of microswimmers. On the one hand, they help to unravel the relevant interaction mechanisms and, on the other hand, allow for the study of sufficiently large systems. A prominent example is the squirmer model introduced by Lighthill [15] and revised by Blake [16] (see section 2.5.4)”[D]. In modern studies, the spherical squirmer is typically characterized by two modes accounting for its swimming velocity and its force-dipole [17] (modes $B_1$ and $B_2$ in eq. (2.96)). The latter distinguishes between pushers, pullers, and neutral squirmers. “The assumption of a spherical shape is adequate for swimmers like Volvox, however, the shape of bacteria such as E. coli or the time-averaged shape of cells such as Chlamydomonas is nonspherical. Hence, an extension of the squirmer concept to spheroidal objects is desirable. In 1977, Keller and Wu proposed a generalization of the squirmer model to a prolate-spheroidal shape, which resembles real biological microswimmers such as Tetrahymenapryiformis, Spirostomum ambiguum, and Paramecium multiclonucleatum [98]. However, that squirmer model accounts for the swimming mode only and does not include a force-dipole mode. This is unfortunate, since the force-dipole mode determines swimmer-swimmer and swimmer-wall interactions [81, 90, 92, 190]. A route to incorporate the force-dipole mode into the spheroidal squirmer model was proposed in ref. [86]. However, to the best of our knowledge, the resulting hydrodynamic model is not solvable analytically. In section 9.2, we propose an alternative model for a spheroidal squirmer, taking into account both, a swimming and a force-dipole mode. The major advantage of our approach is that

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the flow field can be determined analytically.

Various mesoscale simulation techniques have been applied to study the dynamics of squirmers embedded in a fluid, comprising Stokesian dynamics [92, 93, 96], the boundary-element method [7, 86, 91, 190, 191], the multiparticle collision dynamics (MPC) approach [8, 30, 90], lattice Boltzmann simulations [89, 94, 192], the smoothed profile method [95], and the force-coupling approach [193]" [D]. Here, we will present an implementation of a spheroidal squirmer within the MPC approach. MPC "has been successfully applied in various studies of active systems underlining the importance of hydrodynamic interactions for microswimmers [3, 30, 33, 73, 75, 90, 145–147, 149–151, 194–196]" [D]. We study the resulting flow field and thermal properties and compare them with theoretical predictions.

**9.2 Hydrodynamic model of a spheroidal squirmer**

**9.2.1 Spheroid geometry**

"We describe a nonspherical squirmer as a prolate spheroidal rigid body with a prescribed surface velocity $\mathbf{u}_{sq}$. In Cartesian coordinates $(x, y, z)$, the surface equation of a spheroid, or ellipsoid of revolution, is

$$\frac{x^2}{b_x^2} + \frac{y^2}{b_y^2} + \frac{z^2}{b_z^2} = 1,$$

(9.1)

with $b_z$ and $b_x$ the semi-major and semi-minor axis, respectively, and $b_z \geq b_x$ (see fig. 9.1). We denote half of the focal length by $c = \sqrt{b_z^2 - b_x^2}$, which yields the
9.2 Hydrodynamic model of a spheroidal squirmer

eccentricity \( e = c/b_z \). Furthermore, we define a swimmer diameter as \( \sigma = 2b_z \). In terms of prolate \((b_z > b_x)\) spheroidal coordinates \((\zeta, \tau, \varphi)\), the Cartesian coordinates are given by

\[
\begin{align*}
    x &= c\sqrt{\tau^2 - 1} \sqrt{1 - \zeta^2} \cos \varphi, \\
    y &= c\sqrt{\tau^2 - 1} \sqrt{1 - \zeta^2} \sin \varphi, \\
    z &= c\tau\zeta,
\end{align*}
\]

(9.2) where \(-1 \leq \zeta \leq 1, 1 \leq \tau < \infty, \) and \(0 \leq \varphi \leq 2\pi\) [D]. Note, that \((\zeta, \tau, \varphi)\) form a right-handed system. 

The intersection of the spheroid and a meridian plane, where \( \varphi \) is constant, is an ellipse. The normal \( \mathbf{n} \) and tangent \( \mathbf{s} \) to this ellipse are given by the unit vectors \( \mathbf{e}_\tau \) and \( -\mathbf{e}_\zeta \), respectively, which follow by partial derivative of eqs. (9.2) to (9.4) with respect to the coordinates \( \zeta \) and \( \tau \). For \( b_x = b_z \), the spheroid becomes a sphere. The spherical coordinates

\[
(x, y, z)^T = r(\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)^T
\]

(9.5) are obtained from eqs. (9.2) to (9.4) for \( \tau \to \infty, c\tau = r, \) and \( \zeta = \cos \theta \). In this limit, the unit vectors turn into \( \mathbf{e}_r \to \mathbf{e}_r \) and \( \mathbf{e}_\zeta \to -\mathbf{e}_\theta \) (see fig. 9.1). The Lamé metric coefficients for prolate spheroidal coordinates are \( h_\zeta = c(\tau^2 - \zeta^2)^{1/2}(1 - \zeta^2)^{-1/2} \), \( h_\tau = c(\tau^2 - \zeta^2)^{1/2}(\tau^2 - 1)^{-1/2} \) and \( h_\varphi = c(\tau^2 - 1)^{1/2}(1 - \zeta^2)^{1/2} \) [D].

9.2.2 Flow field

The squirmer is immersed in an incompressible fluid at zero Reynolds number, which is described by the incompressible Stokes equations (see section 2.3). Since the squirmer shall be axisymmetric, we can express the velocity field in terms of the stream function \( \Psi \) as (see section 2.3.4)

\[
\mathbf{v}(\zeta, \tau) = -\text{curl} \left( \frac{1}{h_\varphi} \Psi(\tau, \zeta) \mathbf{e}_\varphi \right). 
\]

(9.6)

\[a\)There is a typo in publication [D]. The minus sign on the right hand side of eq. (9.6) is missing.

\[b\)The stream function itself satisfies the equation [70]

\[
E^4 \Psi = 0,
\]

(9.7) with the operator [197]

\[
E^2 = \frac{1}{c^2(\tau^2 - \zeta^2)} \left( (\tau^2 - 1) \frac{\partial^2}{\partial \tau^2} + (1 - \zeta^2) \frac{\partial^2}{\partial \zeta^2} \right).
\]

(9.8)
Each function $\Psi$ in the kernel of $E^2$ can be represented as [197]

$$
\Psi(\tau, \zeta) = \sum_{n=0}^{\infty} \sum_{i=1}^{4} c_n^i \Theta_n^i(\tau, \zeta),
$$

(9.9)

with constants $c_n^i$ and the functions

$$
\Theta_n^1(\tau, \zeta) = G_n(\tau)G_n(\zeta), \quad \Theta_n^2(\tau, \zeta) = G_n(\tau)H_n(\zeta),
\Theta_n^3(\tau, \zeta) = H_n(\tau)G_n(\zeta), \quad \Theta_n^4(\tau, \zeta) = H_n(\tau)H_n(\zeta).
$$

Here, $G_n(x)$ and $H_n(x)$ are Gegenbauer functions of the first and second kind, respectively (see Appendix 9.A). The velocity components follow from the stream function via [70]

$$
v_\tau = \frac{1}{h_\zeta h_\varphi} \frac{\partial \Psi}{\partial \zeta} = c^{-2}(\tau^2 - 1)^{-\frac{1}{2}}(\tau^2 - \zeta^2)^{-\frac{1}{2}} \frac{\partial \Psi}{\partial \zeta},
$$

(9.10)

$$
v_\zeta = -\frac{1}{h_\tau h_\varphi} \frac{\partial \Psi}{\partial \tau} = -c^{-2}(1 - \zeta^2)^{-\frac{1}{2}}(\tau^2 - \zeta^2)^{-\frac{1}{2}} \frac{\partial \Psi}{\partial \tau}.
$$

(9.11)

An important feature of a squirmer is the hydrodynamic boundary condition at its surface, which demands $v(\tau) = u_{sq}$. For the squirming velocity $u_{sq}$ we propose

$$
u_{sq} = -B_1(s \cdot e_z)s - B_2\zeta(s \cdot e_z)s
$$

(9.12)

$$
= -B_1(1 + \beta \zeta)(s \cdot e_z)s
$$

(9.13)

$$
= -B_1\tau_0(1 - \zeta^2)^{-\frac{1}{2}}(\tau_0^2 - \zeta^2)^{-\frac{1}{2}}(1 + \beta \zeta)e_\zeta.
$$

(9.14)

Here, $s$ is the tangent vector, $e_z = (0, 0, 1)^T$ is the unit vector in $z$-direction, $B_1$ and $B_2$ are the two surface velocity modes, and $\beta = B_2/B_1$ (see fig. 9.1). $B_1$ determines the swimming velocity, while the $B_2$ term introduces a force-dipole, or pusher ($B_2 < 0$) and puller ($B_2 > 0$) mode. Note that the spherical squirmer introduced by Lighthill and Blake with modes $B_1$ and $B_2$ [15, 16] is recovered for the spherical limit of a spheroid, where $\zeta \rightarrow \cos(\theta) = n \cdot e_z$.

For $B_2 = 0$, this model of a spheroidal squirmer was already introduced and analysed in refs. [98] and [99]. An additional force-dipole mode has been introduced in refs. [86] and [191] as $u_{sq}(\zeta) = -B_1 s \cdot e_z (1 + \beta n \cdot e_z) s$. However, we prefer the squirming velocity introduced in eq. (9.13), since it yields an analytically solvable boundary value problem for the Stokes equation. The two approaches provide a somewhat different flow field in the vicinity of the squirmer, but both yield the model of Lighthill and Blake in the limit of zero eccentricity.

In the swimmer’s rest frame, and with eq. (9.14), the boundary value problem
becomes
\[ \Psi(\tau, \zeta) \to \frac{1}{2} U_0 c^2 (\tau^2 - 1)(1 - \zeta^2) \text{ for } \tau \to \infty, \quad (9.15) \]
\[ \Psi(\tau_0, \zeta) = 0 \text{ for all } \zeta, \quad (9.16) \]
\[ \left. \frac{\partial \Psi}{\partial \tau} \right|_{\tau=\tau_0} = (B_1 + B_2 \zeta) c^2 \tau_0 (1 - \zeta^2) \text{ for all } \zeta. \quad (9.17) \]

Equation (9.15) implies a constant background flow \( \mathbf{v} = -U_0 \mathbf{e}_z \) infinitely far from the squirmer” [D], which corresponds to the stream function \( \Psi = \frac{1}{2} U_0 \bar{r}^2 = \frac{1}{2} U_0 c^2 (\tau^2 - 1)(1 - \zeta^2) \) (see eq. (2.56)). The no-slip boundary condition in direction of \( \mathbf{e}_\tau \) reads as \( v_\tau(\tau = \tau_0) = \mathbf{u}_{sq}(\zeta) \cdot \mathbf{e}_\tau = 0 \), which guarantees zero fluid influx into the body and is fulfilled for \( \partial \Psi/\partial \zeta = 0 \big|_{\tau = \tau_0} \) (see eq. (9.10)), which in turn is satisfied for \( \Psi(\tau = \tau_0) = 0 \), i.e., eq. (9.16). Finally, the no-slip boundary condition in direction of \( \mathbf{e}_\zeta \) is \( v_\zeta = \mathbf{u}_{sq}(\zeta) \cdot \mathbf{e}_\zeta \), which – together with eqs. (9.11) and (9.14) – yields eq. (9.17). “Due to linearity of the Stokes stream function equation (9.7), we can solve this boundary value problem for \( B_2 = 0 \) first, which yields the stream function \( \Psi_1 \). Subsequently we solve the problem

\[ \Psi(\tau, \zeta) \text{ converges for } \tau \to \infty, \quad (9.18) \]
\[ \Psi(\tau_0, \zeta) = 0 \text{ for all } \zeta, \quad (9.19) \]
\[ \left. \frac{\partial \Psi}{\partial \tau} \right|_{\tau=\tau_0} = B_2 c^2 \tau_0 (1 - \zeta^2) \zeta \text{ for all } \zeta. \quad (9.20) \]

Equation (9.18) imposes a vanishing velocity field infinitely far from the squirmer, eq. (9.19) again guarantees \( v_\tau = 0 \) at the spheroid surface, and eq. (9.20) demands \( v_\zeta = \mathbf{u}_{sq}(\zeta, B_1 = 0) \cdot \mathbf{e}_\zeta \). We denote the solution of the problem eqs. (9.18) to (9.20) by \( \Psi_2 \). Finally, \( \Psi = \Psi_1 + \Psi_2 \) solves the initial problem (9.15)-(9.17) for arbitrary \( B_1 \) and \( B_2 \).

The boundary value problem eqs. (9.15) to (9.17) for \( B_2 = 0 \) can be solved by the ansatz

\[ \Psi_1(\tau, \zeta) = \alpha_1 G_2(\tau) G_2(\zeta) + \alpha_2 H_2(\tau) G_2(\zeta) + \alpha_3 \tau (1 - \zeta^2). \quad (9.21) \]

Here, the third term is found by the separation ansatz \( \Psi(\tau, \zeta) = g(\tau)(1 - \zeta^2) \) for eq. (9.7). Equation (9.15) directly yields \( \alpha_1 = -2U_0 c^2 \). The remaining coefficients \( \alpha_2 \) and \( \alpha_3 \) are determined by eqs. (9.16) and (9.17), keeping in mind that \( B_2 = 0 \). This yields

\[ \alpha_2 = 2c^2 \frac{U_0 (\tau_0^2 + 1) - 2B_1 \tau_0^2}{(\tau_0^2 + 1) \coth^{-1} \tau_0 - \tau_0}, \quad (9.22) \]
\[ \alpha_3 = c^2 B_1 \tau_0 (\tau_0 - (\tau_0^2 - 1) \coth^{-1} \tau_0) - U_0 \frac{1}{(\tau_0^2 + 1) \coth^{-1} \tau_0 - \tau_0}. \quad (9.23) \]
Chapter 9 Modeling a spheroidal microswimmer

The boundary value problem eqs. (9.18) to (9.20) can be solved by the ansatz
\[ \Psi^2(\tau, \zeta) = \alpha_4 G_3(\tau) G_3(\zeta) + \alpha_5 H_3(\tau) G_3(\zeta) + \alpha_6 \zeta (1 - \zeta^2). \] (9.24)

As before, the third term follows by a separation ansatz \( \Psi(\tau, \zeta) = g(\tau) \zeta (1 - \zeta^2) \) for eq. (9.7). Equation (9.18) yields \( \alpha_4 = 0 \). The coefficients \( \alpha_5 \) and \( \alpha_6 \) are determined by eqs. (9.19) and (9.20) such that
\[ \alpha_5 = \frac{4 B_2 \tau_0}{3 \tau_0 + (1 - 3 \tau_0^2) \coth^{-1} \tau_0}, \] (9.25)
\[ \alpha_6 = \frac{c^2 B_2 \tau_0}{3 \tau_0 + (1 - 3 \tau_0^2) \coth^{-1} \tau_0} \frac{2/3 - \tau_0^2 + \tau_0 (\tau_0^2 - 1) \coth^{-1} \tau_0}{3 \tau_0 + (1 - 3 \tau_0^2) \coth^{-1} \tau_0}. \] (9.26)

The total stream function \( \Psi = \Psi_1 + \Psi_2 \) can be transformed to the laboratory frame (see fig. 9.3) by adding the background flow \( v = U_0 e_z \), which yields
\[ \Psi_{lab} = \Psi - \frac{1}{2} U_0 c^2 (\tau^2 - 1)(1 - \zeta^2) \]
\[ = \alpha_2 H_2(\tau) G_2(\zeta) + \alpha_3 \tau (1 - \zeta^2) + \alpha_5 H_3(\tau) G_3(\zeta) + \alpha_6 \zeta (1 - \zeta^2). \] (9.27)

The force on the spheroid by the fluid follows from a multipole expansion [70, 71], with a Stokeslet as the dominating contribution far away from a swimmer. Hence, for \( r \to \infty \) the stream function \( \Psi_{lab} \) has to be equal to the stream function of a Stokeslet [70], namely
\[ \Psi^F = \frac{F_z}{8 \pi \eta} \frac{\tilde{r}^2}{r}, \] (9.28)
and, thus, the force on the spheroidal squirmer is given by [70]
\[ F_z = 8 \pi \eta \lim_{r \to \infty} \frac{r \Psi_{lab}}{r^2} = 8 \pi \eta \frac{\alpha_3}{c}, \] (9.29)

where \( r = \sqrt{x^2 + y^2 + z^2} \) and \( \tilde{r} = \sqrt{x^2 + y^2} \). As expected, \( \Psi_2 \) does not contribute to the force, since it assumes a constant value at infinity. Since a swimmer must be force free, \( F_z = 0 \), which implies \( \alpha_3 = 0 \). Then, eq. (9.23) yields the swimming velocity of the squirmer \( (\tau_0 = 1/e) \)
\[ U_0 = B_1 \tau_0 (\tau_0 - (\tau_0^2 - 1) \coth^{-1} \tau_0), \] (9.30)
which was already found by Keller and Wu for the case \( B_2 = 0 \) [98]. As a consequence, \( \alpha_2 \) in eq. (9.22) simply becomes \( [D]^{b)} \)
\[ \alpha_2 = -2 B_1 c^2 \tau_0 (\tau_0^2 - 1). \]
In summary,\[ \Psi_{lab} = -2 B_1 c^2 \tau_0 (\tau_0^2 - 1) H_2(\tau) G_2(\zeta) + \alpha_5 H_3(\tau) G_3(\zeta) + \alpha_6 \zeta (1 - \zeta^2), \] (9.31)
with the coefficients \( \alpha_5 \) and \( \alpha_6 \) of eqs. (9.25) and (9.26). Different fluid velocity fields of a spheroidal squirmer are presented in figs. 9.2 and 9.3.

\[ ^b) \text{Note that in publication [D], the following simplified expression for } \alpha_2 \text{ misses a minus sign.} \]
Figure 9.2: “Fluid velocity fields of a spheroidal squirmer in the laboratory frame for (a) $B_1 = 1, B_2 = 0$, and (b) $B_1 = 0, B_2 = 1$. The corresponding stream function is given by eq. (9.27). The magnitude of the velocity field is color coded logarithmically. Note that the pusher velocity field with $B_1 = 0, B_2 = -1$ is not shown, since it follows from that of the puller with $B_1 = 0, B_2 = 1$ by inverting the arrows”[D]. (Figure from publication [D])

**Far field:** “The far field of a cylindrically symmetric microswimmer in terms of a multipole expansion was discussed in section 2.5.2. To obtain the far field expansion of our spheroidal squirmer, we expand the stream function, eq. (9.27), in powers of $1/\tau$. Similarly, we determine the stream functions of the first few singularity solutions appearing in the multipole expansion (force dipole, force quadrupole, source dipole, rotlet dipole, etc.) and Taylor expand them in $1/\tau$. Note that we can omit the non-axisymmetric singularity solutions of the multipole expansion like the rotlet dipole ($\mathbf{v}_{RD}$), which is cylindrically symmetric but not axisymmetric ($\mathbf{v}_{RD} \cdot \mathbf{e}_\phi \neq 0$). Equating the coefficients of $(1/\tau)^n$ for $n = 0, 1, 2$, we find that the squirmer is well described in the far field by the flow fields of a force dipole, a source dipole, and a source quadrupole

$$\mathbf{v}(r) = \kappa^{FD} \mathbf{v}^{FD}(r) + \kappa^{SD} \mathbf{v}^{SD}(r) + \kappa^{SQ} \mathbf{v}^{SQ}(r) + \mathcal{O}(r^{-5}),$$

(9.32)
Figure 9.3: Flow field of (a),(d) a spheroidal pusher with $\beta = -3$, (b),(d) a neutral squirmer, and (c),(f) a puller with $\beta = 3$. In (a)-(c) flow fields are shown in the laboratory frame, while (d)-(f) depicts flow fields in the body-fixed frame. The magnitude of the velocity field is color coded logarithmically.

where

\[ \mathbf{v}^{FD}(r) = \frac{r}{r^3} \left( \frac{3z^2}{r^2} - 1 \right), \]
\[ \mathbf{v}^{SD}(r) = \frac{1}{r^3} \left( -e_z + \frac{3zr}{r^2} \right), \]
\[ \mathbf{v}^{SQ}(r) = \frac{3}{r^4} \left( \frac{5z^2r}{r^3} - \frac{2ze_z + r}{r} \right), \]

which decay like $r^{-2}$, $r^{-3}$, and $r^{-4}$ for large $r$, respectively [7][D]. The corresponding stream functions are

\[ \Psi^{FD} = -\frac{r^2z}{r^3}, \quad \Psi^{SD} = -\frac{r^2}{r^3}, \quad \Psi^{SQ} = -\frac{3r^2z}{r^5}, \]

where
and are found by integration of eq. (2.56)
\[ \Psi = \int dz \bar{r}v_x = -\int d\bar{r} \bar{r}v_z. \] (9.37)

Since \( \Psi \) is a physical quantity (see section 2.3.4), we obtain \( \Psi^{FD}, \Psi^{SD} \) and \( \Psi^{SQ} \) in spheroidal coordinates by substituting \( \bar{r} = \bar{r}(\zeta, \tau), \ z = z(\zeta, \tau) \) into eq. (9.36). The Taylor expansions in \( 1/\tau \) are
\[ \Psi^{FD} = -\zeta(1 - \zeta^2)\tau^0 - (\zeta/2)(1 - \zeta^2)(1 - 3\zeta^2)\tau^{-2} + \mathcal{O}(\tau^{-4}), \] (9.38)
\[ \Psi^{SD} = -c^{-1}(1 - \zeta^2)\tau^{-1} + \mathcal{O}(\tau^{-3}), \] (9.39)
\[ \Psi^{SQ} = -3c^{-2}\zeta(1 - \zeta^2)\tau^{-2} + \mathcal{O}(\tau^{-4}). \] (9.40)

For the squirmer’s stream function, an expansions in \( 1/\tau \) up to \( \mathcal{O}(\tau^{-2}) \) yields
\[ \Psi_{lab}^{FD} = \alpha_6\zeta(1 - \zeta^2)\tau^0 + \alpha_2 \frac{1}{6}(1 - \zeta^2)\tau^{-1} + \alpha_5 \frac{1}{30}\zeta(1 - \zeta^2)\tau^{-2} + \mathcal{O}(\tau^{-3}). \] (9.41)

Equating the coefficients of \( 1/\tau \), we find the multipole coefficients
\[ \kappa^{FD} = -\alpha_6, \] (9.42)
\[ \kappa^{SD} = -c\alpha_2 \frac{1}{6} = \frac{B_1}{3}c^3\tau_0(\tau_0^2 - 1), \] (9.43)
\[ \kappa^{SQ} = -c^2\alpha_5 \frac{1}{90}, \] (9.44)

with the coefficients \( \alpha_5 \) and \( \alpha_6 \) of eqs. (9.25) and (9.26). “The values of the multipole coefficients in the spherical limit \( (b_z \to b_x = R, \text{ where } R \text{ is the radius}) \) follow from the above coefficients for \( \tau_0 \to \infty, \ c \to 0, \) and \( c\tau_0 = R \) as”[D] \( \kappa^{FD} = -B_2R^2/2, \ \kappa^{SD} = \frac{B_1R^3}{3}, \) and \( \kappa^{SQ} = B_2R^4/6 \) as expected for a spherical squirmer (see section 2.5.4)\(^c\).

### 9.3 Implementation of a spheroidal squirmer in MPC

A spheroidal squirmer is a homogeneous rigid body characterized by its mass \( M \), center-of-mass position \( C \), orientation quaternion \( q \), translational velocity \( U \), and angular momentum \( l \) or equivalently angular velocity \( \Omega \). “The orientation vector of a spheroid is denoted by \( e = D^T e^b = D^T (0, 0, 1)^T \). The moment of inertia tensor in the body-fixed frame \( I^b \) is a constant diagonal matrix with diagonal elements \( I_x = (M/5)(b_x^2 + b_y^2) = I_y \) and \( I_z = (2M/5)b_z^2 \). For all simulations we choose a neutrally bouyant spheroid, i.e., \( M = \rho(4\pi/3)b_z b_x^2 \), where \( \rho \) is the fluid mass density. The coupling to the MPC method is implemented as outlined in section 4.3.2”[D]. To

\(^c\)Note that the coefficient \( \kappa^{SD} \) has a minus sign in publication [D], which is a typo.
find the tangent vector when computing the squirming velocity $u_{sq}$, the expression

$$s = \frac{1}{\sqrt{b_z^2 - e^2z^2}}(-\sqrt{b_z^2 - z^2}e_z + z\sqrt{1 - e^2}e_r)$$

(9.45)

in cylindrical coordinates $(\bar{r}, \varphi, z)$ is very useful. As outlined in section 4.3.2, fluid particles have to be projected onto the body’s surface after they were streamed back half a time step. Similarly, for the evaluation of the squirming velocity of ghost particles, their positions are temporarily projected onto the body’s surface as well. We implemented this projection of particles onto the body surface in two different ways. One possibility is to construct a virtual spheroid with semi-axes $\tilde{b}_z$, $\tilde{b}_x$, $\tilde{b}_z/\tilde{b}_x = b_z/b_x$ and the particle position on its surface. “The particle is then translated along the normal vector of the virtual spheroid until it is on the real spheroid’s surface. Alternatively, the difference vector $\bar{r} - C$, where $\bar{r}$ is the particle position, can be scaled such that the particle position lies on the spheroid’s surface. We tried both approaches and found no significant difference”[D]. All results presented in the following, were found using the scaling method.

During the streaming step, the rigid body equations of motion for the spheroidal squirmers are integrated numerically. For this purpose, the widely applied leap-frog method [58] is not useful, since velocity, angular momentum, position, and orientation are required at the same point in time for the coupling to the MPC method. Hence, we employ the Verlet algorithm of ref. [60], which is presented in section 1.3. The forces $F$ and torques $T$ acting on the squirmer result from steric interactions with walls and other squirmers. They are derived from interaction potentials as presented in Appendix 9.B.

### 9.4 Simulations – thermal properties and flow field

All following simulations employ MPC-SRD+a with the mean number of particles per collision cell $\langle N_c \rangle = 10$, the rotation angle $\alpha = 130^\circ$, and the time step $h = 0.02\sqrt{ma^2/(k_B T)}$, which yields a fluid viscosity of $\eta = 17.8\sqrt{mk_B T/a^4}$.

#### 9.4.1 Passive colloid

“For the passive spheroidal colloid ($B_1 = B_2 = 0$), we perform equilibrium simulations and determine $\langle U_{\alpha}^2 \rangle$ as well as $\langle (\Omega^\alpha_{\alpha})^2 \rangle$ for $\alpha \in \{x, y, z\}$. Due to the equipartition of energy, we expect

$$\langle U_{\alpha}^2 \rangle = \frac{k_B T}{M},$$

(9.46)

$$\langle (\Omega^\alpha_{\alpha})^2 \rangle = \frac{k_B T}{I_\alpha}.$$  

(9.47)
Figure 9.4: “Orientation correlation functions $\langle \mathbf{e}(t) \cdot \mathbf{e}(0) \rangle$ for passive spheroids with $b_z = 6a, b_x = 3a$ (bottom blue line) and $b_z = 9a, b_x = 3a$ (top black line). The plot shows the simulation data (blue and black solid lines), an exponential fit to that data (red dashed), and the theoretical prediction according to eq. (9.48) (green dotted)”[D]. (Figure from publication [D])

We fix the aspect ratio $b_z/b_x = 2$ and vary $b_x$ in the range $b_x \in [2a, 4a]$. The simulation results agree very well with the theoretical values (9.46) and (9.47). As expected, the deviations from theory decrease with increasing spheroid size, due to a better resolution in terms of collision cells. In general, the relative error $\sigma_r = (\langle x_{\text{theo}}^2 \rangle - \langle x_{\text{sim}}^2 \rangle)/\langle x_{\text{theo}}^2 \rangle$ is larger for $\Omega^b_\alpha$ than for $U^b_\alpha$. We find the largest relative error for $\langle (\Omega^b_\alpha)^2 \rangle$, namely $\sigma_r = 9.5\%, 5.3\%$, and $3.1\%$ for $b_x = 2a, 3a$, and $4a$. Hence, we choose the minor axis $b_x \geq 3a$ in the following.

In addition, we determine the orientation correlation function $\langle \mathbf{e}(t) \cdot \mathbf{e}(0) \rangle$. The theory of rotational Brownian motion [112] predicts

$$\langle \mathbf{e}(t) \cdot \mathbf{e}(0) \rangle = \exp (-2D_R^-t),$$

(9.48)

where $D_R = (2D_R^+ + D_R^\parallel)/3$, $D_R^\parallel = k_BT/\xi^\parallel$, $D_R^\perp = k_BT/\xi^\perp$, and $\xi^\parallel$ and $\xi^\perp$ are the parallel and perpendicular rotational friction coefficients of a prolate spheroid with respect to the major semi-axis”[D] (see eqs. (2.64) and (2.65)). “Simulation results for the orientational auto-correlation function are shown in fig. 9.4 for two spheroids of different eccentricity. The correlation functions decay exponentially. However, for the spheroid with the smaller eccentricity, we find a somewhat faster decay than predicted by theory, whereas good agreement is found for the larger spheroid. We attribute the difference to finite-size effects related to the discreteness of the collision lattice. For larger objects, discretization effects become smaller”[D].
Figure 9.5: “Mean swimming velocity as function of the eccentricity e for a spheroidal squirmer with \( B_1 = 0.05\sqrt{k_B T/m} \) and \( B_2 = 0 \). The solid line shows the theoretical prediction of eq. (9.30). Black dots are simulation results. The eccentricity was varied by changing \( b_z \) and keeping \( b_x = 3a \) constant. For the red triangle, we simulated a larger spheroid with \( b_x = 6a \), which shows a better agreement with theory”[D]. (Figure from publication [D])

9.4.2 Squirmer

“We determine the steady state swimming velocity of a squirmer via \( \langle e \cdot U \rangle \), which should be equal to \( U_0 \) (see eq. (9.30)). Results for various eccentricities are displayed in fig. 9.5. The velocity \( U_0 \) increases with increasing eccentricity \( e \) in close agreement with the theoretical prediction of eq. (9.30). We confirm that the force-dipole parameter \( \beta \) does not affect the velocity of the squirmer, as long as the Reynolds number Re is low, i.e., \( Re = \rho U_0 b_z / \eta \lesssim 0.1 \). We also determine the orientational correlation function and find that a squirmer exhibits the same orientational decorrelation as the corresponding passive particle (see fig. 9.4)”[D].

The simulation data for the mean-square displacement \( \langle (C(t) - C(0))^2 \rangle \), shown in fig. 9.6, nicely agrees with the theoretical expectation for an active Brownian particle (see section 3.4) [148]

\[
\langle (C(t) - C(0))^2 \rangle = 2U_0^2 \tau_R t - 2U_0^2 \tau_R^2 (1 - \exp(-t/\tau_R)).
\] (9.49)

Here, \( \tau_R = 1/(2D^-) \), and we use the value of \( \tau_R \) gained from a fit to the orientation correlation function (see fig. 9.4 and eq. (9.48)). This agreement with active Brownian particle theory, which neglects inertia, results from the fact that the inertia time scales in our simulations \( M/\gamma_z \approx 2\sqrt{k_B T/(ma^2)} \) and \( I_z/\xi_z \approx 2\sqrt{k_B T/(ma^2)} \) are much smaller than the swimming time scale \( \sigma/U_0 \approx 300\sqrt{k_B T/(ma^2)} \) (see
Figure 9.6: Mean-square displacement of a spheroidal squirmer with \( b_z = 6a, b_x = 3a, B_1 = 0.05\sqrt{k_B T/m} \), and \( \beta = 0 \). The blue solid corresponds to simulation data, while the red dashed line represents the theoretical prediction according to eq. (9.49).

Moreover, we calculate the flow field from the simulation data and compare it with the theoretical prediction. As shown in fig. 9.7, the two fields are in close agreement. The two-dimensional flow field of the MPC fluid, averaged over the rotation angle \( \varphi \), is determined at the vertices of a fine resolution mesh. The velocities at these vertices include averages over time of an individual realization as well as ensemble averages over various realizations. By the latter, we determine an estimate for the error of the mean velocity. The median (over vertices) of this error is approximately 5% for the parameters of fig. 9.7 (b) and 10% for that of fig. 9.7 (e). Note that we choose a smaller swimming mode \( B_1 \) for the puller (fig. 9.7 (b)) than for the neutral squirmer (fig. 9.7 (e)). The reason is that the agreement with theory was not satisfactory for the puller with \( B_1 = 0.05\sqrt{k_B T/m} \), which we attribute to a non-vanishing Reynolds number (\( Re \approx 0.1 \)) in the simulation. In figs. 9.7 (c) and (f), we observe lines of high relative errors (yellow in the color code). They appear because theory predicts \( v_r = 0 \) or \( v_z = 0 \) for these lines, which is difficult to achieve in simulations. Hence, the overall agreement between simulations and theory is very satisfactory, and the implementation is very valuable for the simulation of squirmer-squirmer and squirmer-wall interactions, where the details of the flow field matter. For a benchmark of the code on a current GPU see Appendix 9.C”[D].
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Figure 9.7: “Fluid flow fields of a spheroidal squirmer in the laboratory frame with $b_x = 3a$, $b_z = 6a$, $B_1 = 0.01\sqrt{k_BT/m}$, and $\beta = 3 ((a),(b),(c))$, and with $B_1 = 0.05\sqrt{k_BT/m}, \beta = 0 ((d),(e),(f))$. The magnitude of the velocity field (in units of $\sqrt{k_BT/m}$) is color coded logarithmically. The plots (a), (d) show theoretical results, (b), (e) simulation results, and (c), (f) relative errors. The relative error of the flow field is defined as $\Delta v_\alpha = (v^{\text{theo}}_\alpha - v^{\text{sim}}_\alpha)/[(|v^{\text{theo}}_\alpha| + |v^{\text{sim}}_\alpha|)/2]$. Note, due to the discrete representation of the velocity field, some streamlines end abruptly” [D]. (Figure from publication [D])
9.5 Surface accumulation

We study the dynamics of a squirmer between two parallel no-slip walls. For the implementation of planar no-slip walls in MPC see section 4.3.2. Steric interactions between a squirmer and a wall are taken into account by the procedure described in appendix 9.B. The distance between the walls is $L_y$. We are interested in surface accumulation and the squirmer orientation near a wall (see section 2.5.3). For spheroidal squirmers with $b_z = 6a$, $b_x = 3a$, $B_1 = 0.01\sqrt{k_BT/m}$ and $\beta \in \{-5, 0, 5\}$ in a channel of height $L_y = 5\sigma$, results are shown in fig. 9.8. We observe accumulation at the surface in fig. 9.8 (a), which is strongest for pullers (red) and weakest for neutrals (black). Regarding the orientation toward the bottom wall, shown in fig. 9.8 (b), we find that pushers are mainly oriented away from the wall with $e_y \approx 0.25$, corresponding to $\theta \approx 15^\circ$ and that pullers are mainly oriented toward the wall with $e_y \approx -0.25$, corresponding to $\theta \approx -15^\circ$. Finally, neutral squirmers show a broad spectrum of orientations, without an obvious interpretation.

For spherical squirmers we choose $b_z = b_x = 3a$, $B_1 = 0.02\sqrt{k_BT/m}$ and $\beta \in \{-11.4, 0, 11.4\}$, which results in the same force dipole coefficient (see eq. (9.42)) and Pécel number

$$Pe = \frac{U_0}{D_R\sigma} = 25$$

(9.50)

as for the spheroidal squirmers. Here, the Pécel number compares the time scale for rotational diffusion $1/D_R$ with the time it takes to swim one diameter $\sigma/U_0$. The results for spherical swimmers, shown in fig. 9.9, are similar to those of spheroidal swimmers. We find that for spherical squirmers, puller accumulate at the surface as
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Figure 9.9: (a) Probability distribution of the center-of-mass position $C_y$ between the walls for spherical pullers, neutrals and pushers. (b) Probability density of the orientation $e_y$ in the region $0 < C_y < L_y/2$. Positive $e_y$ corresponds to orientation away from the wall.

strongly as pushers. Neutrals still accumulate weakest. The probability distribution of orientations (see fig. 9.9 (b)) reveals that pullers point directly at the wall ($e_y \approx -1$). This can be understood visualizing the puller flow field (see section 2.5.3). It is also plausible that we do not observe a similar state for spheroidal pullers due to their shape. For spherical pushers, we observe a peak at $e_y \approx 0.4$ corresponding to $\theta \approx 24^\circ$. Hence they swim close to the surface but pointing slightly away from it like the spheroidal pushers. Our results for spherical squirmers are in close agreement with the studies in ref. [85]. Note that our repulsive potential slightly differs from that of ref. [85]. For spheroidal squirmers, the orientation with respect to the wall is in agreement with findings of ref. [86]. In ref. [86], no steric interactions with the wall are modeled and fixed-points are determined exclusively by hydrodynamic theory. For the fixed-points of close-by swimming to the wall, ref. [86] finds significantly larger distances to the wall compared to our studies. However, only hydrodynamic interactions with the walls were accounted for in ref. [86]. We conjecture that the inclusion of steric interactions yields fixed points closer to the wall.

9.6 Spheroidal squirmer with a rotlet dipole

9.6.1 Squirming velocity

The rotlet dipole $v^{RD}(r)$ (see section 2.5.2)

$$v^{RD}(r) = \frac{3Fz}{r^5}e_\varphi$$ (9.51)

leads to interesting behavior such as the swimming of *E. coli* in circles above a surface. We can easily conceive a way to model a spheroidal squirmer with a rotlet.
9.6 Spheroidal squirmer with a rotlet dipole

Figure 9.10: Flow field in the $x$-$y$-plane at $z = b_z/2$ of a spheroidal squirmer with $b_x = 3a$, $b_z = 6a$, $B_1 = B_2 = 0$, and $\kappa^{RD} = 10\sqrt{k_BT/ma}$. Plot (a) shows simulation results, while (b) corresponds to the relative error flow field $\Delta v_\alpha = (|v_{\alpha}^{theo} - v_{\alpha}^{sim}|)/(|v_{\alpha}^{theo}| + |v_{\alpha}^{sim}|)/2$. The magnitude of the velocity field (in units of $\sqrt{k_BT/m}$) is color coded logarithmically.

dipole mode. When we evaluate $v^{RD}(r)$ on the spheroid’s surface, we find that it has no components in direction of $e_\tau$. Other singularities, such as the force dipole, do not share this property. Reminding ourselves that a squirmer is defined as a swimmer with a tangential surface velocity (see section 2.5.1), we can prescribe the squirming velocity

$$u^{RD}_{sq}(r) = \kappa^{RD} v^{RD}(r), \quad \text{for } \tau = \tau_0. \quad (9.52)$$

Since $v = \kappa^{RD} v^{RD}(r)$ is a solution of Stokes equations and satisfies the boundary condition $v = u^{RD}_{sq}$ on the spheroid’s surface, the velocity field of such a squirmer in an infinite fluid will be given by $v = \kappa^{RD} v^{RD}(r)$.

Due to the linearity of Stokes equations, we can add $u^{RD}_{sq}$ to our squirming velocity in eq. (9.13). The resulting spheroidal squirmer is controlled by the parameters $B_1$, $\beta = B_2/B_1$, and $\kappa^{RD}$. Note that a different approach to model a squirmer with a rotlet dipole is discussed in ref. [86].

9.6.2 Simulations

The MPC algorithm remains unchanged, except for the function that computes the squirming velocity. As a check, we determine the velocity field of the squirmer at $z = b_z/2$ in the body-fixed frame. Figure 9.10 (a) shows the flow field in the $xy$-plane at $z = b_z/2$, which corresponds to a counter-clockwise rotation as expected. The deviation from theory, shown in Figure 9.10 (b), is about 10%.

Next, we study the motion of a squirmer with a rotlet dipole mode near a no-slip wall with surface normal vector $e_y$. We choose $\beta = -8$, such that the swimmer
remains at the surface, and the rotlet dipole coefficient $\kappa_{RD}^{rd} = 4\sqrt{k_B T/ma^3} \approx 0.1B_1\sigma^3$. Figure 9.11 (a) shows the trajectory of the squirmer at the wall in the $zx$-plane, i.e., viewed from above the surface. We observe clockwise circles, as expected by the far field prediction (see eq. (2.94)) and observed in experiments with $E. coli$ [6]. Due to noise, the swimmer does not trace perfect circles. Figure 9.11 (b) shows the angle between the squirmer’s orientation and the surface, which fluctuates between $10^\circ$ and $20^\circ$. The approximate theoretical prediction for the radius of the circular trajectory (see section 2.5.3) assumes a parallel alignment with the surface, i.e., $\theta = 0$. Nevertheless, we compare the theoretical expression with simulations. To obtain an average radius $R_{rot}$ from simulation data, we determine the trajectory’s curvature $\kappa$, where $\kappa = 1/R_{rot}$ for a circle. Given a trajectory $(z(t), x(t))$, we can find the curvature as [198]

$$\kappa(t) = \frac{\ddot{z} \dddot{x} - \dddot{z} \dddot{x}}{\dot{x}^2 + \dot{z}^2}.$$ \hfill (9.53)

From our discrete data, we determine the derivative according to $\dot{z}(t) = (z(t + \Delta t) - z(t - \Delta t))/(2\Delta t)$. The average radius $R_{rot}$ is determined as the inverse of the mean curvature $R_{rot} = 1/\bar{\kappa}$, where $\bar{\kappa}$ is an average of $\kappa$ over time steps and realizations. Figure 9.12 (a) shows how the radius $R_{rot}$ depends on $\kappa_{FD}$ for a spheroidal swimmer with $b_x = 3a$, $b_z = 6a$, $B_1 = 0.05\sqrt{k_B T/m}$, and $\beta = -8$. To evaluate the theoretical
9.7 Summary and conclusions

Figure 9.12: (a) Radius of the circle which a swimmer with a rotlet dipole traces, as a function on the rotlet dipole coefficient $\kappa^{RD}$. Red bullets represent simulation results, while blue triangles correspond to the theoretical expression eq. (9.54). (b) Dependence of the height above the wall on $\kappa^{RD}$.

expression (see section 2.5.3)

$$R_{rot}/a = \frac{32h^4U_0}{3\kappa^{RD}(1 - \Upsilon)}.$$  \hfill (9.54)

we determine the height $h = C_y$ from simulations and substitute the measured velocity $\langle e \cdot U \rangle$ for $U_0$. The agreement between theory and simulations is surprisingly good. Equally satisfying results were found for other aspect-ratios $b_z/b_x$. For $\kappa^{RD} \geq 5\sqrt{k_B T/ma^3}$ however, the theoretical prediction becomes less and less accurate. We also found that the height of the squirmer above the wall increases with $\kappa^{RD}$. For $\kappa^{RD} = 4\sqrt{k_B T/ma^3}$, the relative increase compared to a squirmer without rotlet dipole is around 4% (see fig. 9.12 (b)). Similarly, the velocity $\langle e \cdot U \rangle$ increases by around 20% from $\kappa^{RD} = 0$ to $\kappa^{RD} = 4\sqrt{k_B T/ma^3}$. An increase of the height or swimming velocity has not been predicted by the theoretical analysis of ref. [7]. We conjecture that it could result from a non-vanishing Reynolds number in the simulations and hence restrict ourselves to $\kappa^{RD} \leq 4\sqrt{k_B T/ma^3}$.

9.7 Summary and conclusions

"We have introduced a spheroidal squirmer model, which comprises the swimming and force-dipole modes. It is a variation of previously proposed squirmer models. On the one hand, it includes the force-dipole mode as an extension to the model of ref. [98]. On the other hand, it is an alternative approach compared to refs. [86] and [191], with the major advantage that our model allows for the analytical calculation of the flow field. In the present calculations we employed the Stokes stream function equation. Very recently a full set of solutions to Stokes’ equations in spheroidal
coordinates were given in ref. [199], which opens an alternative approach to derive the flow field for our choice of boundary conditions.

Furthermore, we have presented an implementation of our spheroidal squirmer in a MPC fluid. In contrast to other frequently employed simulation approaches, MPC includes thermal fluctuations. The comparison between the fluid flow profile of a squirmer extracted from the simulation data with the theoretical prediction yields very good agreement. As a consequence of the MPC approach with its discrete collision cells, the minor axis of the spheroid has to be larger than a few collision cells to avoid discretization effects. The analysis of the squirmer orientation correlation function shows that very good agreement between theory and simulations is already obtained for $b_z = 9a$ (major axis) and $b_x = 3a$ (minor axis)”[D].

Moreover, we studied the surface accumulation of spherical and spheroidal squirmers and found results in agreement with literature. Additionally, we presented how to implement a spheroidal squirmer with a rotlet dipole and found that such a squirmer traces circles above a no-slip wall, as predicted by far field hydrodynamics [7] and observed in experiments with *E. coli* [6].

“Our studies confirm that spheroidal squirmers can accurately be simulated by the MPC method. The proposed implementation opens an avenue to study collective and non-equilibrium effects in systems of anisotropic microswimmers. Even large-scale systems (10$^3$ to 10$^4$ swimmers) can be addressed by the implementation of MPC and the squirmer dynamics on current GPUs”[D].

## Appendix 9.A Gegenbauer functions

“For $n \geq 2$ and $x \in \mathbb{R}$ the Gegenbauer functions of the first and second kind $G_n$ and $H_n$, are defined in terms of the Legendre functions of the first and second kind $P_n$ and $Q_n$ as [197, 200]

$$G_n(x) = \frac{P_{n-2}(x) - P_n(x)}{2n - 1}, \quad H_n(x) = \frac{Q_{n-2}(x) - Q_n(x)}{2n - 1}. \quad (9.55)$$

For $n = 0, 1$, they are defined as

$$G_0(x) = -H_1(x) = 1, \quad G_1(x) = H_0(x) = -x. \quad (9.56)$$

For the reader’s convenience, we give the formula for the Gegenbauer functions of the first kind for $n = 2, 3$, and $x \in \mathbb{R}$

$$G_2(x) = \frac{1}{2}(1 - x^2), \quad (9.57)$$

$$G_3(x) = \frac{1}{2}(1 - x^2)x. \quad (9.58)$$
9.B Steric interactions

Furthermore, the Gegenbauer functions of the second kind for \( n = 2, 3 \), and \( x > 1 \) are given by

\[
H_2(x) = \frac{1}{2} (1 - x^2) \coth^{-1}(x) + \frac{x}{2},
\]

\[
H_3(x) = \frac{1}{2} (1 - x^2) x \coth^{-1}(x) + \frac{1}{6} (3x^2 - 2).
\]

(9.59)

(9.60)

Here, we used \( \coth^{-1}(x) = \ln([x + 1]/[x - 1])/2 \). [D]

**Appendix 9.B Steric interactions**

Here, we illustrate our implementation of the excluded-volume interactions between spheroids and walls following the approach provided in ref. [59].

The spheroid’s surface in the laboratory frame is given by the quadratic form

\[
1 = A(x) \equiv (x - C)^T A(x - C),
\]

(9.61)

where the orientation matrix \( A \) can be expressed as

\[
A = (1 - ee^T)/b_x^2 + ee^T/b_z^2.
\]

(9.62)

For the steric interactions, we introduce a virtual safety distance \( d_v \), which is small compared to \( b_x \) and \( b_z \). When computing steric interactions, we replace \( b_x \) and \( b_z \) by \( b_x + d_v \) and \( b_z + d_v \), respectively"[D]. Within this thesis, we use \( d_v = 0.05a \) for all simulations.

**9.B.1 Interaction between spheroids**

We introduce a repulsive interaction potential between spheroids to prevent their overlap. The potential is given by

\[
U = 4\epsilon_0 \left[ \left( \frac{\sigma_0}{d_R + \sigma_0} \right)^{12} - \left( \frac{\sigma_0}{d_R + \sigma_0} \right)^6 \right].
\]

(9.63)

Here, \( \sigma_0 \) and \( \epsilon_0 \) correspond to a length and energy scale, respectively. We choose \( \epsilon_0 = k_B T \) and \( \sigma_0 = 2d_v \). The directional contact distance \( d_R \) between two spheroids, with orientation matrices \( A_1, A_2 \) and center positions \( C_1, C_2 \), is an approximation to their true distance of closest approach and is defined by

\[
d_R = R (1 - F(A_1, A_2)^{-1/2})
\]

(9.64)
Here, $R = C_2 - C_1$, $R = |R|$, and $F(A_1, A_2)$ is the elliptic contact function, defined as [59]

$$F(A_1, A_2) = \max_{\lambda} \min_x S(x, \lambda)$$

$$= \max_{\lambda} \min_x (\lambda A_1(x) + (1 - \lambda) A_2(x)).$$

Minimization with respect to $x$ demands $\nabla S(x, \lambda) = 0$, and hence,

$$x(\lambda) = \{\lambda A_1 + (1 - \lambda) A_2\}^{-1} \{\lambda A_1 C_1 + (1 - \lambda) A_2 C_2\}. \quad (9.67)$$

The critical value $\lambda = \lambda_c$ that maximizes $S(x(\lambda), \lambda)$ can be found by the root finding problem

$$A_1(x(\lambda)) - A_2(x(\lambda)) = 0. \quad (9.68)$$

We implement Brent’s root finding approach [111]. The forces and torques arising from the potential (9.63) can be calculated analytically and are given by [59] \(^d\)

$$F_1 = \frac{24\varepsilon_0}{\sigma_0} \left[ 2 \left( \frac{\sigma_0}{d_R + \sigma_0} \right)^{13} - \left( \frac{\sigma_0}{d_R + \sigma_0} \right)^7 \right] \left( \frac{R}{R} (R^{-1/2} - 1) - \frac{R}{2} R^{-3/2} X_c \right),$$

and

$$T_1 = -\frac{12R\varepsilon_0}{\sigma_0} \left[ 2 \left( \frac{\sigma_0}{d_R + \sigma_0} \right)^{13} - \left( \frac{\sigma_0}{d_R + \sigma_0} \right)^7 \right] F^{-3/2}(x_c - C) \times X_c \quad (9.70)$$

for the first spheroid, where $X_c = 2\lambda_c A_1(x_c - C_1)$. The force and torque on the second spheroid follow by Newton’s action-reaction law, namely

$$F_2 = -F_1, \quad (9.71)$$

$$T_2 = -T_1 + R \times F_1. \quad (9.72)$$

We restrict ourselves to short-range repulsive interactions by setting the potential $U$ to a constant value for $d_R > (\sqrt{2} - 1)\sigma_0$, which implies that $F_1$ and $T_1$ are zero for this range of $d_R$ values. Note that an upper bound to $d_R$ is $R - 2b_z$, which means that two spheroids will not interact if $R > 2b_z + (\sqrt{2} - 1)\sigma_0$. This inequality is checked before a numerical calculation of $d_R$ is employed”[D].

\(^d\)Note that eq. (54) of ref. [59] contains a typographical error. The factor 24 needs to be replaced by 12.
9.B Steric interactions

9.B.2 Interaction between a spheroid and a wall

"We assume that two parallel walls are positioned at \( y = 0, L_y \), which—taking into account the safety distance \( d_v \)—results in the effective wall positions \( y = d_v \) and \( L_y - d_v \). We propose an interaction between a spheroid and a wall in the style of the spheroid-spheroid interaction presented in ref. [59]. First, we find the point \( x \) on the spheroid’s surface that is closest to a wall. For the wall at \( y = d_v \), this is achieved by minimizing the height \( h(x) = e_y \cdot x - d_v \) under the constraint \( A(x) = 1 \). Using the method of Lagrange multipliers, we have to minimize \( \Lambda(x, \lambda) = h(x) + \lambda(A(x) - 1) \).

The necessary condition for a minimum \( \frac{\partial \Lambda}{\partial x} = 0 \) yields

\[
e_y + \lambda \nabla A(x) = e_y + 2\lambda A(x - C) = 0, \tag{9.73}
\]

and hence,

\[
x = C - A^{-1}e_y/(2\lambda). \tag{9.74}
\]

Substitution of eq. (9.74) into \( A(x) = 1 \) yields

\[
\lambda = \pm \sqrt{(A^{-1})_{yy}/2}. \tag{9.75}
\]

Finally, we obtain the point closest to the wall as

\[
x = C \mp (A^{-1}e_y)/\sqrt{(A^{-1})_{yy}.} \tag{9.76}
\]

Here, the minus sign has to be chosen, which can be visualized by the example of a sphere of radius \( R \), for which \( A = R^{-2}1 \). This finally yields the height

\[
h = C_y - d_v - \sqrt{(A^{-1})_{yy}}. \tag{9.77}
\]

We employ the Lennard-Jones potential

\[
U_w = 4\epsilon_0 \left[ \left( \frac{\sigma_0}{h + \sigma_0} \right)^{12} - \left( \frac{\sigma_0}{h + \sigma_0} \right)^{6} \right], \tag{9.78}
\]

for a repulsive wall, and \( U_w \) assumes a constant value for all \( h \geq (\sqrt{2} - 1)\sigma_0 \). We can derive the force \( F_\alpha = -\partial U_w/\partial C_\alpha \) and torque \( T_\alpha = -\partial U_w/\partial \psi_\alpha \) acting on the spheroid analytically (see section 1.2.2). For the force, we find \( h < (\sqrt{2} - 1)\sigma_0 \)

\[
F = -\frac{\partial U_w}{\partial h} \frac{\partial h}{\partial C_y} e_y \tag{9.79}
\]

\[
= -24\epsilon_0 \frac{\sigma_0}{\sigma_0} \left[ 2 \left( \frac{\sigma_0}{h + \sigma_0} \right)^{13} - \left( \frac{\sigma_0}{h + \sigma_0} \right)^{7} \right] e_y, \tag{9.80}
\]

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and for the torque
\[ T_\alpha = -\frac{\partial U_\alpha}{\partial h} \frac{\partial h}{\partial \psi_\alpha}, \]  
(9.81)

with”[D]
\[ \frac{\partial h}{\partial \psi_\alpha} = -\left( \frac{1}{2\sqrt{(A^{-1})_{yy}}} \right) \frac{\partial (A^{-1})_{yy}}{\partial \psi_\alpha} = \frac{1}{2\sqrt{(A^{-1})_{yy}}} \mathbf{e}_y^T A^{-1} \frac{\partial A}{\partial \psi_\alpha} e_y = \frac{1}{\sqrt{(A^{-1})_{yy}}} (\delta_{\alpha x}(A^{-1})_{yz} - \delta_{\alpha z}(A^{-1})_{yz}), \]  
(9.82)
\[ \frac{\partial h}{\partial \psi_\alpha} = \frac{1}{\sqrt{(A^{-1})_{yy}}} \mathbf{e}_y^T A^{-1} \frac{\partial A}{\partial \psi_\alpha} e_y = \frac{1}{\sqrt{(A^{-1})_{yy}}} \mathbf{e}_y^T A^{-1} e_\alpha \times A A^{-1} e_y \]  
(9.83)
\[ \frac{\partial h}{\partial \psi_\alpha} = \frac{1}{\sqrt{(A^{-1})_{yy}}} \mathbf{e}_y^T A^{-1} \frac{\partial A}{\partial \psi_\alpha} e_y = \frac{1}{\sqrt{(A^{-1})_{yy}}} \mathbf{e}_y^T A^{-1} e_\alpha \times A A^{-1} e_y \]  
(9.84)

"where we used \( d(B^{-1})/dt = -B^{-1}(dB/dt)B^{-1} \) which holds for an invertible matrix \( B \) depending on a scalar parameter \( t \), and eq. (C9) of ref. [59].

For the wall at \( y = L_y - d_v \), we have to minimize \( h(x) = L_y - d_v - \mathbf{e}_y \cdot x \), with \( x \) on the spheroid’s surface. This yields
\[ x = C + A^{-1} e_y \left[ (A^{-1})_{yy} \right]^{-1/2}. \]  
(9.85)

The formulas for torque and force do not change, except that we have to insert \( h = L_y - d_v - C_y - \sqrt{(A^{-1})_{yy}} \) and need to change the sign of the force”[D].

Appendix 9.C Benchmark simulation

Computation time “We perform benchmark simulations on an NVIDIA K80 GPU.

First we determine the computation time for one MPC step in a periodic system, normalized by the number of fluid particles. We obtain approximately 4.5 nanoseconds per time step and particle, which is in accordance with the value of about 3 nanoseconds reported in ref. [37], since the extension to angular momentum conserving MPC roughly doubles the computation time, while the NVIDIA K80 is approximately twice as fast as the NVIDIA GTX580 employed in ref. [37].

Next, we perform a benchmark MPC simulation of many spheroidal squirmers with \( b_x = 3a, b_z = 6a \) in a narrow slit. The slit height and length are chosen as \( L_y = 7a \) and \( L_x = L_z = 600a \), respectively. We vary the number \( N_{sq} \) of squirmers, i.e., the two-dimensional packing fraction \( N_{sq} \pi b_x b_z / (L_x L_z) \), and determine the computation time per MPC time step, normalized by the number of fluid and ghost particles. A simulation without squirmers takes approximately 4.9 nanoseconds per time step and particle. Hence, the addition of no-slip walls leads only to a minor increase in simulation time by approximately 10%. When spheroidal squirmers are introduced,
the computation time increases linearly with the number of squirmers up to a value of 13.2 nanoseconds for the two-dimensional packing fraction 0.6, which corresponds to 3825 squirmers in our set-up. Similar values are found for smaller \((L_x = L_y = 300a)\) and larger systems \((L_x = L_y = 900a)\). The increase in computation time is mostly related to the squirmers’ ghost particles. Unlike the fluid particles and the ghost particles for the no-slip walls, they are not spatially ordered in memory, which leads to a significantly lower processing speed (see fig. 3 of ref. [37]).

The steric interactions are computed sequentially on the CPU exploiting a cell-linked list [58]. The computation time spent for the steric interactions increases linearly with \(N_{sq}\). However, it only contributes by about 4\% to the total simulation time for the considered set-up”[D].

### Memory limitations

“The size of the systems that can be studied with our code is limited by the available memory on the used GPU. Explicitly, the required memory to store fluid particle, collision cell, and spheroid properties are listed in Table 9.1.

<table>
<thead>
<tr>
<th>Memory usage per cell:</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>particles per cell</td>
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</tr>
<tr>
<td>random seed</td>
<td>8 bytes</td>
</tr>
<tr>
<td>cell energy/thermostat</td>
<td>4 bytes</td>
</tr>
<tr>
<td>center of mass velocity</td>
<td>24 bytes</td>
</tr>
<tr>
<td>rotation matrix</td>
<td>24 bytes</td>
</tr>
<tr>
<td>center of mass</td>
<td>24 bytes</td>
</tr>
<tr>
<td>inertia tensor</td>
<td>48 bytes</td>
</tr>
<tr>
<td>angular momentum</td>
<td>24 bytes</td>
</tr>
<tr>
<td>total per cell</td>
<td>160 bytes</td>
</tr>
</tbody>
</table>

<table>
<thead>
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<th>Memory usage per fluid particle:</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
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</tr>
<tr>
<td>particle velocity (double precision)</td>
<td>24 bytes</td>
</tr>
<tr>
<td>cell index</td>
<td>4 bytes</td>
</tr>
<tr>
<td>temporary memory for performance optimization</td>
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</tr>
<tr>
<td>total per particle</td>
<td>52 bytes</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Memory usage per spheroid:</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>parameters describing current state</td>
<td>296 bytes</td>
</tr>
<tr>
<td>parameters describing state after half time step</td>
<td>296 bytes</td>
</tr>
<tr>
<td>total per spheroid</td>
<td>592 bytes</td>
</tr>
</tbody>
</table>

Table 9.1: “Memory requirement to store fluid particle, collision cell, and spheroid properties”[D]. (Table from publication [D])
Compared to the number of fluid particles, the number of spheroidal squirmers is several orders of magnitude smaller and their impact on GPU memory usage is negligible. With a typical density of $\langle N_c \rangle = 10$ particles per cell, the required GPU memory is 680 bytes per collision cell. Therefore, approximately $1.5 \times 10^6$ collision cells can be studied per 1 GB of GPU memory. Hence, the memory capacity of 4-12 GB of recent GPUs corresponds approximately to $6 \times 10^6 - 2 \times 10^7$ collision cells. The volume of a spheroidal squirmer with $b_x = 3a$, $b_z = 6a$ is equivalent to 226 cells. Assuming a 30% volume fraction of squirmers, memory limits the total number of squirmers to $8 \times 10^3 - 2.4 \times 10^4$. Note that for systems of many squirmers, the time scales of interest can be very long. Hence, in applications, the considered system size has often to be smaller ($\approx 10^3$ squirmers) for acceptable total simulation times”[D].
Chapter 10

Cooperative swimming in a narrow slit

The results of this chapter have been published in publication [D] (see List of publications on page 203). All verbatim quotes within this chapter are quotations of [D] and are indicated as “…”[D]. Such verbatim quotes can extend over several pages and have been reformatted to fit with the main text. The label of a cited article inside a verbatim quote follows the enumeration in the bibliography (page 186).

10.1 Simulations

“We simulate the cooperative swimming behavior of two squirmers in a slit geometry. The slit is formed by two parallel no-slip walls located at $y = 0$ and $y = L_y$”[D].

“Steric interactions between two squirmers and between a squirmer and a wall are taken into account by the procedure described in appendix 9.B. The initial positions and orientations of the two squirmers $(i = 1, 2)$ are

$$C_{1/2} = \left( \frac{L_x}{2} \pm \frac{d_{cm}}{2}, \frac{L_y}{2}, \frac{L_z}{2} \right)^T,$$

$$e_{1/2} = (\pm \cos(\alpha_0), 0, \sin(\alpha_0))^T. \quad (10.1)$$

Here, $d_{cm}$ is the initial center-of-mass distance and $\alpha_0 = (\pi - \theta_0)/2$, where $\theta_0$ is the initial angle between $e_1$ and $e_2$. The swimming mode is chosen as $B_1 = 0.05 \sqrt{k_B T/m}$ and the force dipole mode $\beta \in \{-4, 0, 4\}$. We choose $d_{cm}$ such that the squirmers are well separated and vary $\theta_0$. The squirmers major and minor axes are $b_x = 3a$ and $b_z = 6a$, respectively, and the simulation box size is $L_x = L_z = 15b_z$, and $L_y = 7a$. Note that $L_y \gtrsim b_x$ which keeps the swimming orientation essentially in the $xz$-plane.

Results for the mean surface-to-surface distance between squirmers $\langle d_s \rangle$ and the mean alignment $\langle e_1 \cdot e_2 \rangle = \langle \cos \theta \rangle$ are shown in fig. 10.1 for pushers, pullers, and neutral swimmers with an initial angle $\theta_0 = 3\pi/8$. Due to the setup, the squirmers initially approach each other and collide at $tU_0/\sigma \approx 0.5^\circ[D]$. The Péclet number
Chapter 10  Cooperative swimming in a narrow slit

Figure 10.1: “Average surface-to-surface distance $d_s$ and orientation of squirmers, where $\cos(\theta) = e_1 \cdot e_2$, as function of time. The solid blue, dashed black, and dotted red lines correspond to pullers $\beta = 4$, neutrals $\beta = 0$ and pushers $\beta = -4$. The standard deviation of the blue line ($\beta = 4$) is indicated by the cyan shaded region”[D]. (Figure adapted from publication [D])

$Pe = U_0/(D_R \sigma) \approx 120$ is sufficiently high, such that the squirmer orientation has hardly changed before collision. “When the neutral swimmers collide, they initially align parallel ($\cos \theta \approx 1$ at $tU_0/\sigma \approx 1$ in fig. 10.1), but their trajectories start to diverge immediately thereafter. Pushers remain parallel for an extended time window, which is expected as pushers are known to attract each other [90], but at $tU_0/\sigma \approx 3$ (see fig. 10.1) their trajectories diverge as well. This is probably due to noise, since we observe several realizations where pushers remain parallel. Interestingly, pullers, which are known to repel each other when swimming in parallel[90], swim cooperatively and reach a stable orientation with $\langle \cos(\theta) \rangle \approx 0.77$ shortly after they collided (at $tU_0/\sigma \approx 1$). Thereby, their cooperative swimming velocity is about $0.8U_0$. The flow field of this stable state, determined by MPC simulations, is shown in fig. 10.2. Note that the velocity field in the swimming plane is left-right symmetric, and that there is a stagnation point in the center behind the swimmers. Figure 10.2 reveals that this point actually corresponds to a line normal to the walls. In ref. [201], it was shown that the flow field of a force dipole in a narrow slit exhibits a recirculating pattern with loops in a plane parallel to the walls [77] and a parabolic flow profile perpendicular to them. Both features can be observed in fig. 10.2. Figure 10.3 shows that the fixed point of cooperatively swimming pullers is reached for nearly all simulated initial conditions $\theta_0 \in (0, \pi/2)$. Only pullers that are nearly parallel initially ($\theta_0 = \pi/8$, $\cos \theta_0 \approx 0.92$ in fig. 10.3), repel each other such that they will not reach the fixed point. For Péclet numbers $Pe < 60$, the fixed point remains at $\langle \cos(\theta) \rangle \approx 0.77$. However, it becomes more likely for the swimmers to escape (or never reach) the fixed point.
Figure 10.2: “Flow field of two cooperatively swimming pullers in the laboratory frame. The magnitude of the velocity field (in units of $\sqrt{k_B T/m}$) is color coded logarithmically. We denote the direction normal to the walls by $y$, the cooperative swimming direction by $z$, the remaining Cartesian axis by $x$, and choose the swimmers’ center of mass as origin. Panel (a) shows the flow field at $x = 0$ in the $zy$-plane, while panel (b) shows the flow field at $y = 0$ in the $xz$-plane. The black elliptical shapes (“transparent”, solid) indicate the projection of the swimmers onto the considered plane”[D]. (Figure from publication [D])
Figure 10.3: “Time dependence of the average alignment $\langle e_1 \cdot e_2 \rangle = \langle \cos \theta \rangle$ of two pullers with $\beta = 4$, $b_z/b_x = 2$ in a slit of height $L_y = 7a$, and various initial angles $\theta_0 \in (0, \pi/2)$” [D]. (Figure from publication [D])

A detailed study reveals that the fixed point vanishes, when the walls are replaced by periodic boundary conditions (we use a cubic simulation box of length $10b_z$). This is even true when we apply three-dimensional periodic boundaries, but keep the wall potential implemented, i.e., the squirmers are still confined in a narrow slit of height $7a$ as before (the fluid simulation box is cubic with length $10b_z$). These observations are quantified in fig. 10.4. Note that the slow increase of the average squirmer surface-to-surface distance in the case of a periodic system with wall potential is due to the fact that the pullers still swim together in several realizations. However, in these realizations they no longer swim in a plane as for the slit geometry, but rather show three-dimensional configurations with tilted major axes. Evidently, hydrodynamic interactions with the walls [78] are essential for the swimmers’ dynamics.

In addition, we studied the swim behavior of spherical squirmers. Here, we observe diverging trajectories for all squirmer types, i.e., pushers, neutral squirmers, and pullers. Such diverging trajectories have already been reported in ref. [90] for spherical squirmers in bulk. In contrast, in ref. [202] a cooperative swimming mode for spherical squirmers has been observed (see Fig. 22(c) of ref. [202]). However, this cooperative swimming—termed pair-swimming by the authors—is unstable to perturbations that displace one swimmer out of the swimming plane [202]. Since our simulations and those of ref. [90] include thermal fluctuations, we consequently do not observe the cooperative swimming mode of ref. [202].

Hence, the stable close-by cooperative swimming of pullers is governed by the squirmer anisotropy, by the hydrodynamic interactions between them and, impor-
Figure 10.4: “Average surface-to-surface distance $d_s$ between two pullers with $\beta = 4$, $b_z = 6a$, and $b_x = 3a$ as a function of time for various geometries. The distance $d_s$ increases rapidly after collision for squirmers in a cubic simulation box of side length $10b_z$ with periodic boundary conditions (black dotted line). Trajectories still diverge, when a confining potential for the squirmers is present (red dashed line); for details see text. Squirmers confined in a slit of width $L_y = 10a$ with no-slip walls swim together (blue solid line). However, their trajectories diverge for wide slits ($L_y = 11a$ for the green dash-dotted line and $L_y = 12a$ for the cyan dash-dot-dotted line)”[D]. (Figure from publication [D])

This conclusion is in contrast to results presented in ref. [191], where a monolayer of spheroidal squirmers is considered, with their centers and orientation vectors fixed in the same plane, however, without confining walls. The study reports a stable cooperative motion for pullers with angles $\theta \in (0, \pi/2)$ by nearest-neighbor two-body interactions, where all angles between 0 and $\pi/2$ are stable. The difference to our study is that in ref. [191] cooperative features were extracted from a simulation of many swimmers, whereas we explicitly studied two swimmers. Furthermore our study explicitly models no-slip walls and includes thermal noise.

To shed light on the stability of the cooperative puller motion, we varied the puller strength $\beta$, the aspect ratio $b_z/b_x$, and the width of the slit $L_y$. Thereby, we started from our basic parameter set $b_x = 3a$, $b_z = 6a$, $L_y = 7a$, and $\beta = 4$. With decreasing $\beta$, the stable alignment disappears, i.e., the pullers’ distance increases after collision. For increasing $\beta$ the fixed point remains, but the value of $\cos \theta$ decreases, i.e., the squirmers form a larger angle. With increasing wall separation, the fixed-point value of $\cos \theta$ decreases, i.e, the angle between the swimmers increases. For $L_y/b_z \gtrsim 11/6$, the mean distance between swimmers increases rapidly after collision (see fig. 10.4).
An increase of the aspect ratio $b_z/b_x$ from 2 to 3 and 4 increases the fixed-point value of $\langle \cos \theta \rangle$ from 0.77 to 0.84 and 0.88. The more elongated shape leads to a more parallel alignment of the squirmers. For $b_z/b_x \geq 2$, the minimal value of $\beta$ required to achieve cooperative motion depends weakly on the aspect ratio. In particular, for $b_z/b_x = 2, 3, \text{and} 4$, we find the minimal value $\beta \approx 3.5$ [D].

10.2 Summary and conclusion

“To shed light on the cooperative swimming motion and on near-field hydrodynamic interactions, we investigated the collision of two spheroidal squirmers in a slit geometry. We found a stable stationary state of close-by swimming for spheroidal pullers, which is determined by hydrodynamic interactions between the anisotropic squirmers, and, even more important, by squirmers and surfaces. This stationary state disappears for low puller strengths and low eccentricities. We expect the stable close-by swimming of pullers to strongly enhance clustering in puller suspensions in narrow slits” [D]. This presumption will be verified in the following chapter.
Chapter 11

Collective dynamics of microswimmers in a narrow slit

11.1 Introduction

Bacteria in quasi-two-dimensional confinement exhibit remarkable phenomena, such as the formation of clusters observed for *Myxococcus xanthus* [19], swarming, swirling and raft formation observed, e.g., for *E. coli* [18, 203–206] and the emergence of meso-scale turbulence reported for suspensions of *Bacillus subtilis* [20]. Cluster formation, swarming, and meso-scale turbulence have been modeled by self-propelled rods [19, 20, 207–209], which are rod-shaped active Brownian particles (see section 3.4) and hence capture self-propulsion, anisotropic drag, steric interactions, thermal fluctuations – although some studies set $k_B T = 0$ [20, 209] –, but exclude hydrodynamic interactions. The intuitive explanation for the emergence of clusters is that steric interactions align bacteria or self-propelled rods, which then swim collectively. If the packing fraction and Péclet number are high enough, a small cluster can maintain its size until it interacts with other swimmers or clusters, which results in fusion and cluster growth.

Experiments on self-phoretic artificial spherical microswimmers, such as Janus particles, self-propelled liquid droplets, and photo-activated colloids [21, 210–213], as well as simulations of circular or spherical active Brownian particles [22, 23] have shown that motility-induced cluster formation can occur without any shape anisotropy. In contrast to high aspect-ratio self-propelled rods, cluster formation of isotropic swimmers is not due to alignment, but rather due to jamming\footnote{Note that this is different in three dimensions, where active spherical particles where shown to move cooperatively [25, 214]. Here, we restrict ourselves to 2D and quasi-2D systems.}. In ref. [29], the mechanism is described as a positive feedback loop between the facts that swimmers accumulate where they move more slowly, and move more slowly in a crowded environment. Aspects on clustering of microswimmers and active particles are discussed in the review articles [2, 3, 11, 14, 24].

An interesting point regarding motility-induced phase separation of microswimmers is the role of hydrodynamics. In ref. [30], the dynamics of many spherical
Chapter 11 Collective dynamics of microswimmers in a narrow slit

squirmers in a narrow slit was studied. This setup captures the geometry of experiments, where glass plates are used for confinement. Additionally, the flow field of self-phoretic spherical microswimmers is very close to that of a neutral squirmer, which has been argued theoretically [14, 215] and been observed in experiments [211]. By means of a comparison between Brownian dynamics simulations and MPC simulations, the authors of ref. [30] found that hydrodynamics enhances cluster formation. However, in ref. [29], a two dimensional system of squirmers was studied, and contrary to ref. [30], hydrodynamics was found to suppress clustering.

In this chapter, we reconsider parts of the study of ref. [30] and additionally consider spheroidal squirmers. We compare MPC and Langevin dynamics (LD) simulations to elucidate the role of hydrodynamics. The extension to spheroidal swimmers is of interest, since most biological swimmers are non-spherical, and since the mechanisms for cluster formation are different for anisotropic and isotropic swimmers. We conjecture that the role of hydrodynamics may depend on the swimmer’s shape.

For a closer match to bacteria like *Bacillus subtilis* and *E. coli*, we add a rotlet dipole mode to the velocity field of our squirmer (see section 9.6), which captures the flow induced by the rotation and counter-rotation of flagella and cell body [6, 151]. It has been shown that the rotlet dipole significantly influences the hydrodynamic interactions between a microswimmer and a surface, leading to the circular trajectories observed for *E. coli*. The influence of the rotlet dipole on hydrodynamic interactions between swimmers, and consequently on the collective dynamics of spheroidal squirmers, is investigated in section 11.6.

11.2 Simulation setup and parameters

The simulation setup consists of $N_{sw}$ spheroidal squirmers with minor axis $b_z = 3a$ and aspect ratio $b_z/b_x \in \{1, 2, 3, 4\}$ in a narrow slit, i.e., a simulation box of dimension $L_x = L_z \equiv L$, $L_y = 7a$ with no-slip walls in $y$-direction and periodic boundary conditions in $x$- and $z$-direction. We employ MPC-SRD+a simulations with the time step $h = 0.02 \sqrt{ma^2/(k_BT)}$, rotation angle $\alpha = 130^\circ$, and the mean number of particles in a cell $\bar{N}_c = 10$, which yields a fluid viscosity of $\eta = 17.8 \sqrt{mk_BT/a^4}$. Complementary to the MPC simulations, we perform Langevin dynamics simulations of active spheroidal particles (see section 3.5). To investigate the collective behavior for different packing fractions $\phi = N_{sw}(4\pi/3)b_x^2 b_z/(L^2 L_y)$, the length $L$ of the square-shaped slit is varied, while the number of swimmers is fixed at $N_{sw} = 196$ for $b_z/b_x = 1$ and $N_{sw} = 200$ for all other aspect ratios. Since the system is quasi-two dimensional, we will often refer to the area packing fraction $\phi^{2D} = N_{sw} \pi b_x b_z / L^2$, instead of the packing fraction $\phi$. The ratio $\phi^{2D}/\phi = 3L_y/(4b_z) = 7/4$ is constant for our choice of parameters, as we keep $L_y = 7a$ and $b_x = 3a$ constant. We choose the swimming modes $B_1/\sqrt{k_BT/m} = 0.01, 0.005, 0.003$, and 0.002 for $b_z/b_x = 1, 2, 3,$
and 4, respectively, yielding an identical Pécelt number of $Pe = 12$ for all aspect ratios. The squirming mode $\beta$ is chosen as $\beta \in \{-1, 0, 1\}$. We restrict ourselves to small squirming velocities $B_1$ and $B_2 = B_1 \beta$, since we observed very low MPC-fluid densities between clustering swimmers otherwise.

**Problem of density inhomogeneities for high squirming velocities** Figure 11.1 (a) and (b) show the density of fluid and ghost particles in the slice $\{(x, y, z) | L_y/2 - a/2 < y < L_y/2 + a/2\}$ for a system of spherical neutral squirmers ($b_z = b_x = 3a$) with $B_1 = 0.04 \sqrt{k_B T/m}$ in a periodic slit of length $L = 96a$ and height $L_y = 7a$. In fig. 11.1 (a), the circles of zero MPC fluid density (blue in the color code) correspond to the squirmers. We can observe that the fluid particle density outside of the cluster is around $\rho_{fl} = 15m/a^3$ (green in the color code), which is higher than the equilibrium density of $\rho_{fl} = 10m/a^3$. Hence, the fluid particle density inside the cluster must be diminished. To visualize this, we can additionally plot the ghost particles (see fig. 11.1 (b)). Since the squirmers are chosen to be neutrally buoyant, the ghost particle density $\rho_{gh}$ is by construction equal to the global fluid density $\rho_{fl} = 10m/a^3$. If the fluid density between squirmers was equal to the global density $\rho_{fl} = 10m/a^3$, the cluster region would appear uniformly colored in fig. 11.1 (b). The fact that it does not, shows that the fluid density in between squirmers is in fact reduced. This is particularly striking for the dark blue circular region in the top right corner of fig. 11.1 (b), which corresponds to a defect in the cluster, i.e., a region where one spherical swimmer is “missing” in the cluster. Here, there are cells with 0 MPC particles. Averaged over all cells in that region, the fluid particle density is around $\rho_{fl} = 1.5m/a^3$.

There are two explanations for this finding. On the one hand, the squirmers could pump fluid particles out of the gaps in-between them. In MPC, which is compressible, the only mechanism acting against this is diffusion of particles back into the gaps. If the squirming mode $B_1$ is too strong, diffusion cannot balance the pumping, and we obtain regions of low fluid density between squirmers\(^b\). If this reasoning is correct, the reduced density is an artifact of the simulation method and hence unphysical. Furthermore, the reduced density will result in a reduced local pressure, and hence, artificially enhance clustering. On the other hand, we may regard pressure gradients due to squirming as physical. In ref. [216], it was argued that clustered squirmers in quasi-2D build up a hydrodynamic pressure gradient due to their source dipole field. Since MPC satisfies the ideal gas equation of state, this pressure gradient must translate into a density gradient.

Independent of the reason for a reduced MPC fluid density inside a cluster – be it artificial or not –, the variation of density in a MPC simulation should always be small compared to the total density, since the viscosity of the MPC fluid is density

\(^b\)Adam Wysocki discovered the problem of a nonuniform MPC fluid particle density in squirmer systems and suggested this argument (unpublished, private communication).
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Figure 11.1: (a),(c),(e): MPC fluid particle density $\rho_{fl}/m$ in the slice $\{(x, y, z) \mid L_y/2 - a/2 < y < L_y/2 + a/2\}$ for a system of neutral ($\beta = 0$) spherical squirmers in a slit. (b),(d),(f): Sum of fluid particle and ghost particle density $(\rho_{fl} + \rho_{gh})/m$. For (a) and (b) the squirmer’s swimming mode is $B_1 = 0.04 \sqrt{k_B T/m}$, while the global fluid particle density is $\rho_{fl} = 10m/a^3$. Plots (c) and (d) correspond to (a) and (b), but with $B_1 = 0.02 \sqrt{k_B T/m}$, while (e) and (f) correspond to (a) and (b) but with $\rho_{fl} = 20m/a^3$. 

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11.2 Simulation setup and parameters

dependent (see section 4.5). Figure 11.1 (b) shows that for \( B_1 = 0.04 \sqrt{k_B T/m} \) the fluid density in between squirmers is less than half of the global density. Hence, for the parameters of fig. 11.1 (a) and (b), MPC does not yield reliable results, i.e., results that would agree with a direct numerical solution of Stokes equations, for which viscosity is constant.

Figure 11.1 (c) and (d) correspond to the simulations of fig. 11.1 (a) and (b), but with a decreased swimming mode of \( B_1 = 0.02 \sqrt{k_B T/m} \). In this case the fluid density is homogeneous and the cluster has vanished. A rule of thumb to estimate when pumping might appear, can be constructed by comparing the time a fluid particle needs to diffuse over a swimmer’s diameter with the time it needs to cover a swimmer’s diameter when moving with velocity \( U_0 \). We denote the resulting dimensionless number as pumping number

\[
Pu = \frac{t_{\text{diff}}}{t_{\text{pump}}} = \frac{\sigma^2/(6D_f)}{\sigma/U_0} = \frac{U_0\sigma}{6D_f} \propto \frac{B_1\sigma}{h}.
\]

Here, \( h \) is the MPC time step and \( D_f \) the fluid particle diffusivity (see section 4.5). For a small pumping number \( Pu \), fluid particles diffuse fast into the gaps, guaranteeing a nearly uniform density. A small pumping number can only be obtained by decreasing \( B_1 \), since we already choose the minimum value of \( \sigma \) that still yields a good spatial resolution of the swimmer in terms of collision cells. A considerable increase of \( h \) is problematic, since the implementation of immersed objects (see section 4.3.2) is based on the assumption that the distance a fluid particle travels in one time step is small compared to the object’s size. Furthermore we are restricted to small \( h \) to guarantee isothermality [159]. For fig. 11.1 (a) and (b) \( Pu = 2 \), while \( Pu = 1 \) for (c) and (d). The pumping number is only a guideline, since the problem will also depend on the size of the cluster and the type of swimmer, e.g., pusher, puller or neutral. We found that also \(|\beta|\) needs to be moderate in order to avoid density inhomogeneities.

One might also argue that increasing the global fluid density \( \rho \) could resolve the problem of reduced viscosity, since the relative variation of density \( \Delta \rho/\rho \) might decrease, which would result in a more uniform viscosity. However, this is not the case, as fig. 11.1 (e) shows. Here, the global density is twice as high as in fig. 11.1 (a), i.e. \( \rho = 20m/a^3 \) instead of \( \rho = 10m/a^3 \), but the value of \( \Delta \rho/\rho \) is still around \( 0.5 = (15 - 10)/10 = (30 - 20)/20 \) for the region outside of the cluster.

In the following, we restrict ourselves to to parameter regimes of small \( B_1, \beta, \) and moderate packing fraction, for which density and hence viscosity are approximately constant. We verified for all of the following simulations that the density is in fact homogeneous. Note that the simulations of refs. [30, 216] were performed with \( B_1 = 0.1 \sqrt{k_B T/m} \). Hence, we suspect that the fluid density is highly inhomogeneous in these simulations.
11.3 Characterizing cluster structures by Voronoi decomposition and polar pair correlations

Figure 11.2 shows the different kinds of structures that can be observed for neutral squirmers with parameters specified in section 11.2 at the fixed area packing fraction \( \phi^{2D} = 0.5 \), namely a gas phase for the aspect ratio \( b_z/b_x = 1 \) (see fig. 11.2 (a)), a jammed cluster for \( b_z/b_x = 2 \) (see fig. 11.2 (b)), and collectively swimming clusters for \( b_z/b_x = 3 \) and 4 (see fig. 11.2 (c) and (d)). For the spherical swimmers in fig. 11.2 (a), we can observe a small cluster of hexagonal order in the top left of the simulation box. However, such clusters only have a short life time. They form and break dynamically. Note that there is not a clear separation between jammed and collectively swimming clusters in terms of the aspect ratio \( b_z/b_x \). Other realizations at \( b_z/b_x = 2 \) also show an aligned cluster.

We perform a Voronoi decomposition to quantify clustering and determine polar pair correlation functions to quantify polar alignment.

Local packing fraction via Voronoi decomposition — To quantify clustering, we employ a Voronoi decomposition [217] of the slit, which returns a partition of the slit volume into Voronoi cells. The Voronoi cell of a generator point consists of all points in space whose distance to the generator point is less than or equal to its distance to any other generator point. For spherical swimmers, we choose their centers as generator points. The Voronoi decomposition is applied for the full three dimensional slit. For a clear visualization, fig. 11.3 (a) shows a two-dimensional Voronoi decomposition of the system of spherical swimmers of fig. 11.2 (a), where the coordinates perpendicular to the wall are discarded. For spheroidal swimmers we cannot use their centers as generator points, since this would disregard the swimmers’ anisotropy. Hence, we mesh their surfaces using the algorithm of ref. [218], and choose the resulting vertices as generators (see fig. 11.3 (b)). The Voronoi cell volume of a spheroidal swimmer is then defined as the sum of Voronoi cell volumes of all its vertices.

The local packing fraction \( \phi_{loc} \) of a swimmer is defined as the fraction \( V_s/V_v \), where \( V_s = (4\pi/3)b_x^2b_z^2 \) is the swimmer’s volume and \( V_v \) is the volume of its Voronoi cell. If the histogram of local packing fractions \( P(\phi_{loc}) \) shows the majority of local packing fractions to be larger than the global packing fraction, the system is clustering.

Check of simulation time and system size — The probability density of local packing fraction \( P(\phi_{loc}) \) for \( N_{sw} = 200 \) swimmers with \( b_z = 6a \), \( b_x = 3a \), \( B_1 = 0.005\sqrt{k_B T/m} \), and \( \beta = 0 \) in a slit of length \( L = 150a \) is shown in fig. 11.4 (a) for several points in time. This setup corresponds to a global packing fraction of \( \phi = 0.29 \) or equivalently an area packing fraction of \( \phi^{2D} = 0.5 \). For each point in time \( t \) we include snapshots from an interval \( (t - \Delta t, t + \Delta t) \) in the histogram,
11.3 Characterizing cluster structures by Voronoi decomposition and polar pair correlations

Figure 11.2: Snapshots of microswimmers in a narrow slit. We represent the quasi-two dimensional system of spheroid-shaped swimmers by a two-dimensional image of ellipse-shape swimmers. Any apparent overlap between swimmers is due to this representation. The velocity of each swimmer is colored coded and its orientation is indicated by a white line. Short white lines as in (a) indicate a swimming orientation perpendicular to the walls.
where $\Delta t = 7\sigma/\mathcal{U}_0$. Figure 11.4 (a) reveals that most of the swimmers have a local packing fraction of around 0.6 which is much larger than the global packing fraction of 0.29. Hence, the system is clustering. Furthermore, fig. 11.4 (a) verifies that the simulation time is long enough to capture the system’s structural properties, since $P(\phi_{loc})$ is essentially constant in time for $t\mathcal{U}_0/\sigma \gtrsim 70$, apart from fluctuations.

Next, we check how the probability density of local packing fraction depends on the system size. Complementary to the simulations of 200 swimmers in a slit of length $150a$, we perform simulations with 800 swimmers in a slit of length $300a$. A snapshot of this large system is shown in fig. 11.5. Figure 11.4 (b) shows that the probability densities of local packing fraction $P(\phi_{loc})$ are very similar for the small and large system. Hence, our restriction to systems of 200 swimmers is justified.

**Polar pair correlation function** We introduce the polar pair correlation function [219]

$$C_1(r) = \frac{\left\langle \sum_{i,j>i} \mathbf{e}_i \cdot \mathbf{e}_j \delta(r - d_s(i,j)) \right\rangle}{\left\langle \sum_{i,j>i} \delta(r - d_s(i,j)) \right\rangle}, \quad (11.2)$$

where $d_s(i,j)$ is the surface-to-surface distance between swimmer $i$ and swimmer $j$. The polar pair correlation function $C_1(r)$ measures how aligned a swimmer is with its neighbors at a surface-to-surface distance $r$. For discrete simulation data, $C_1(r)$
11.3 Characterizing cluster structures by Voronoi decomposition and polar pair correlations

Figure 11.4: (a) Probability distribution $P$ of the local packing fraction $\phi_{loc}$ for 200 swimmers with $b_z = 6a$, $b_x = 3a$, $B_1 = 0.005\sqrt{k_BT/m}$, and $\beta = 0$ in a slit of length $L = 150a$ for several subsequent times. The global packing fraction is 0.29. (b) Probability distribution $P$ of the local packing fraction $\phi_{loc}$ for $N_{sw}$ swimmers with $b_z = 6a$, $b_x = 3a$, $B_1 = 0.005\sqrt{k_BT/m}$, and $\beta = 0$ in a slit of length $L$. The blue line corresponds to $N_{sw} = 800$ and $L = 300a$, while the green line corresponds to $N_{sw} = 200$ and $L = 150a$.

Figure 11.5: Snapshot of a simulation of $N_{sw} = 800$ spheroidal neutral squirmers with area packing fraction $\phi^{2D} = 0.5$ and aspect ratio $b_z/b_x = 2$. 
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Figure 11.6: Polar pair correlation $C_1(r)$ for $N_{sw}$ swimmers with $b_z = 6a$, $b_x = 3a$ and $B_1 = 0.005\sqrt{k_B T/m}$, $\beta = 0$ in a slit of length $L$. The blue line corresponds to $N_{sw} = 800$ and $L = 300a$, while the green line corresponds to $N_{sw} = 200$ and $L = 150a$.

is not evaluated using the above definition including a delta-function but rather by means of binning. We divide the distance $r$ into intervals $I_m = [m\Delta r, (m + 1)\Delta r]$ with $m \in \mathbb{Z}_{\geq 0}$ and $\Delta r > 0$ and define $C_1^m$ as the value of $C_1(r)$ at the midpoint of $I_m$. We initialize all $C_1^m$ with zero. In a loop over $i$ and $j > i$ we determine the surface to surface distance $d_s(i,j)$ for each squirming pair and in which interval $I_m$ it lies. Then, we add $e_i \cdot e_j$ to $C_1^m$. Finally, we divide $C_1^m$ by the number of squirming pairs which contributed to its value.

Check of system size — In fig. 11.6 the polar pair correlation $C_1(r)$ is shown for systems of 200 and 800 spheroidal squirmers, both with equal area packing fraction $\phi^{2D} = 0.5$. We can observe that the curves overlap for $r \lesssim 2\sigma$. For larger distances, a system-size effect is apparent.

11.4 Simulation results – spherical swimmers

For the spherical swimmers, the Péclet number $Pe = 12$ is too low for phase separation to occur. Hence, we modify the parameters specified in section 11.2. We can increase the Péclet number by increasing the swimming velocity or by decreasing the rotational diffusion. We do both and increase the swimming mode to $B_1 = 0.02\sqrt{k_B T/m}$, and the global MPC fluid density to $\bar{N}_c = 40$ or 80 particles per cell, which increases the viscosity and hence decreases rotational diffusion$^c)$. Simulations at $\bar{N}_c = 40$, and hence $Pe = 116$, and area packing fraction $\phi^{2D} = 0.6$ show cluster formation for both, the MPC and Langevin dynamics (LD) simula-

$^c)$We verified that only minor fluid density inhomogeneities occur.

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Figure 11.7: (a), (c) Probability distribution of local packing fraction and (b), (d) probability distribution of the hexagonal order parameter $|q_6|^2$ for a system of spherical swimmers with $B_1 = 0.02 \sqrt{k_B T / m}$ and $\phi^{2D} = 0.6$ in a narrow slit. The fluid viscosity is $\eta = 86.4 \sqrt{mk_B T / a^4}$, resulting in the Péclet number $Pe = 116$ for (a) and (b), and $\eta = 178.1 \sqrt{mk_B T / a^4}$, $Pe = 230$, for (c) and (d).

tions. Thereby temporal clusters of hexagonal order form and dissolve quickly. For LD simulations, a cluster extending over nearly the whole system can be observed. A first look at videos of the simulations suggests that clusters are smaller and less long-lived in the MPC simulations. Figure 11.7 (a) shows the histogram of local packing fractions for swimmers in MPC and LD simulations at $Pe = 116$. We can observe that a high local packing fraction is much more likely in the Langevin dynamics simulations. To quantify hexagonal order, we introduce the hexagonal order parameter $|q_6|^2$ [23, 30, 220]. For squirmer $k$, it is defined as

$$ q_6 \equiv \frac{1}{6} \sum_{j \in N^{(6)}} e^{i \alpha_{kj}}, $$

where the sum runs over the six nearest neighbors of squirmer $k$ and $\alpha_{kj}$ is the angle between the position difference $C_k - C_j$ and the $x$-axis. For a perfect hexagonal
lattice $|q_6|^2 = 1$. Figure 11.7 (b) shows that the largest hexagonal order $|q_6|^2 \approx 1$ is about twice as likely for the active Brownian particles compared to the neutral squirmers. The force dipole mode $\beta$ does not significantly change the results in the studied parameter regime. For $N_c = 80$, and hence $Pe = 230$, clusters still form and break, but they are longer-lived in the LD simulations. In contrast to the active Brownian particles, the squirmers do not form a large cluster spanning the whole system, but rather show local clusters of hexagonal order which quickly dissolve (see fig. 11.8). Histograms of local packing fraction and the hexagonal order parameter for $Pe = 230$ are shown in fig. 11.7 (c) and (d). The results are similar but somewhat more pronounced compared to those at $Pe = 116$. Hence, our simulations suggest that hydrodynamics suppresses clustering of spherical swimmers in quasi-2D, as already found in ref. [29] for circular swimmers in 2D. What might be the reason for suppressed clustering of squirmers compared to active Brownian particles? In ref. [29], it was argued that squirmers strongly reorient due to hydrodynamic torques during squirmer-squirmer collisions. Hence, the average orientation autocorrelation function

$$C_e(t) = \frac{1}{N_{sw}} \sum_{i=1}^{N_{sw}} \langle \mathbf{e}_i(t) \cdot \mathbf{e}_i(0) \rangle,$$ (11.4)

decays like $\exp(-t/\tau_{sq})$ instead of $\exp(-2D_R t)$. Here, $\tau_{sq}$ characterizes the time-scale of squirmer-squirmer collisions and $\tau_{sq} \ll 1/(2D_R)$. This can can be interpreted as a source to yield a decreased effective Péclet number $Pe' = \tau_{sq} U_0 / \sigma$, and hence suppressed phase separation. We verified that $\tau_{sq} \ll 1/(2D_R)$ in fig. 11.9 (a). In fig. 11.9 (b), it is shown that neutral squirmers are less likely oriented toward a
11.5 Simulation results – spheroidal swimmers

Figure 11.9: (a) Orientation correlation function $C_e(t)$ averaged over all swimmers for a system of spherical squirmers and active Brownian particles at $\phi^{2D} = 0.6$ and $Pe = 116$. (b) Probability distribution of the $y$-component of the orientation vector, $e_y = e \cdot e_y$, for the same system. For the active Brownian particles, all orientations are equally likely. Neutral squirmers are mainly orientated parallel to the wall.

wall compared to active Brownian particles. This might be another reason for their suppressed phase separation: orienting toward a wall slows down a swimmer, which leads to an effectively decreased Péclet number.

The results of ref. [30] are opposite to our findings. We conjecture that this is due to an artifact related to the high value of $B_1$ in the simulations of ref. [30], which is discussed in section 11.2.

11.5 Simulation results – spheroidal swimmers

We employ the parameters for spheroidal squirmers specified in section 11.2. Although $Pe = 12$ is quite small, we can observe cluster formation (see fig. 11.10). Figure 11.11 (g) reveals that for the area packing fraction $\phi^{2D} = 0.5$, the squirmers with aspect ratio $b_z/b_x = 2$ form clusters, which translates into a peak of $P(\phi_{loc})$ at a local packing fraction $\phi_{loc}$ nearly twice as large as the global packing fraction. In the corresponding LD simulations, however, the local packing fraction is close to the global packing fraction, i.e., there is no cluster formation. The active Brownian particles do not phase separate until $b_z/b_x = 3$ and $\phi^{2D} = 0.5$ (see fig. 11.12).

The tendency of hydrodynamics to enhance phase separation of elongated squirmers ($b_z/b_x > 1$) is found for all studied parameters, which is shown in fig. 11.11 and fig. 11.12. Thereby, a high area packing fraction and a high aspect ratio $b_z/b_x$ are beneficial for cluster formation. Hence, for moderate $\beta$ ($|\beta| \leq 1$), the results for spherical and elongated swimmers are opposite. Hydrodynamics enhances clustering for elongated swimmers and suppresses it for spherical swimmers. We can observe that pullers cluster strongest as we already conjectured from our studies in chap-
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Figure 11.10: Snapshots of spheroidal swimmers with $Pe = 12$, $b_z/b_x = 2$, at area packing fraction $\phi^{2D} = 0.5$. (a) shows active Brownian particles, while (b) shows neutral squirmers.

Pullers exhibit phase separation already at $\phi^{2D} = 0.2$, which is indicated by a bimodal $P(\phi_{loc})$ (see fig. 11.11 (a) and (b)), while pushers tend to cluster less than neutrals and pullers. We run an additional parameter set with $\beta = -5$ and aspect ratio $b_z/b_x = 2$ at the area packing fraction $\phi^{2D} = 0.5$. Note that we cannot run simulations with high positive $\beta$ because of the resulting MPC fluid inhomogeneities (see section 11.2). However, for $\beta = -5$ we did not observe these inhomogeneities.

The probability distribution of local packing fractions $P(\phi_{loc})$ for $\beta = -5$ is represented by a dark orange line in fig. 11.11 (g). It shows that very strong pushers cluster less or equally weak as active Brownian particles.

The results are summarized in a state diagram fixed at $Pe = 12$ and dependent on $\phi^{2D}$ and $b_z/b_x$ in fig. 11.13. Based on our data in fig. 11.12, we dissected the $(b_z/b_x, \phi^{2D})$ parameter-space into regions where cluster formation is observed, and regions where it is not. The line separating these regions is drawn by hand to give a qualitative impression. The finding that pullers cluster strongest, while pullers cluster weakest, is represented in fig. 11.13 by an upward shift of said line for increasing $\beta$ and a corresponding downward shift for decreasing $\beta$. For $|\beta| \leq 1$, the line for active Brownian particles (magenta in fig. 11.13) is above all squirmer-lines. Our finding of section 11.4, that hydrodynamics suppresses motility-induced phase separation of spherical swimmers, will be reflected by a crossing of the lines for squirmers and active Brownian particles when $b_z/b_x$ decreases to 1. However, for a low Péclet number as in fig. 11.13 we might not find cluster formation of spherical swimmers at all, which means that the lines might reach $\phi^{2D} = 1$ at $b_z/b_x > 1$. 
11.5 Simulation results – spheroidal swimmers

Figure 11.11: Probability density of the local packing fraction $\phi_{\text{loc}}$ for different global area packing fractions $\phi^{2D}$ and aspect ratios $b_z/b_x$. 
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Figure 11.12: Each image in this table shows the probability density $P(\phi_{loc})/P_{max}$ of local packing fraction $\phi_{loc}$ color coded for different global packing fractions $\phi$. Thereby, $P_{max}$ is the maximum value for $P(\phi_{loc})$ in that image. The white dashed lines correspond to $\phi_{loc} = \phi$, i.e., the line where the local packing fraction equals the global packing fraction. All packing fractions $\phi$ are also given in terms of area packing fractions $\phi^{2D}$.
11.5 Simulation results – spheroidal swimmers

Figure 11.13: State diagram of spheroidal microswimmers at $Pe = 12$ dependent on global area packing fraction $\phi^{2D}$ and aspect ratio $b_z/b_x$. Thereby, (a) corresponds to simulation results of active Brownian particles, while (b), (c), and (d) correspond to squirmers with $\beta = 0$, $-1$, and $1$ respectively. A bullet at a point $(\phi^{2D}, b_z/b_x)$ indicates that clusters were observed for these parameters, while a cross indicates that no clusters were observed. The solid line (binodal) marks the separation between the cluster and no-cluster parameter regime. All binodal lines are shown together in (e).
As discussed in the introduction, elongated swimmers align due to steric interactions, which initializes clustering. For squirmers, hydrodynamic interactions might enhance this alignment. We can determine the polar pair correlation $C_1(r)$ (see eq. (11.2)) and the average orientation autocorrelation function $C_e(t)$ (see eq. (11.4)) to quantify alignment. However we have to be careful with our conclusions, since we average over different realizations, which can exhibit different structures, as fig. 11.14 reveals. Furthermore, local alignment is not equivalent to collective swimming: the system in fig. 11.14 (a) has a high local alignment while still being jammed.

The polar pair correlation function $C_1(r)$ for swimmers with $b_z/b_x = 2$ at the global area packing fraction $\phi^{2D} = 0.5$ in fig. 11.15 (a) shows that there is a stronger alignment for squirmers than for active Brownian particles. For neutrals, the alignment is already stronger than for active Brownian particles at $\phi^{2D} = 0.2$ (see fig. 11.15) (b)), although both do not form clusters yet. This could be an indication that hydrodynamically induced alignment is a reason for enhanced phase separation. For pushers however, the polar pair correlation $C_1(r)$ is as large as, or even slightly less than for active Brownian particles at $\phi^{2D} = 0.2$. At $\phi^{2D} = 0.5$ on the other hand, pushers with $\beta = -1$ show stronger alignment and exhibit clustering, which the active Brownian particles do not.

We define the local alignment coefficient $\mathcal{R}$ as

$$\mathcal{R} = \frac{1}{b_x} \int_0^{b_x} dr \, C_1(r),$$

which corresponds to the average alignment $\mathbf{e}_i \cdot \mathbf{e}_j$ of a squirmer $i$ with all its neighbors $j$. Thereby, the squirmers $i$ and $j$ are regarded as neighbors if their surface-to-surface distance $d_s(i, j)$ is less than $b_x$. In fig. 11.16, the local alignment coefficient $\mathcal{R}$ is plotted as a function of the global area packing fraction. Pullers (blue symbols), which show clusters for all plotted area packing fractions, exhibit local alignment, which depends weakly on packing fraction. For pushers, neutrals, and active Brownian particles, the local alignment coefficient increases with increasing global area packing fraction. For neutrals (blue symbols) a plateau can be observed. For the active Brownian particle the local alignment coefficient stays nearly constant for $b_z/b_x = 2$. This agrees with the observation that they are gas-like with $P(\phi_{loc})$ centered near the global packing fraction for all studied parameters (see fig. 11.12). For $b_z/b_x = 3$ however, the active Brownian particles eventually form clusters (see fig. 11.12), which is accompanied by an increase of the local alignment coefficient.

The orientation autocorrelation function of a single swimmer is $C_e(t) = \exp(-D_{R^2} t)$ instead of $C_e(t) = \exp(-2D_{R^2} t)$, due to the quasi-two-dimensional confinement, which we verified in separate simulations. Figure 11.17 shows the orientation autocorrelation function for swimmer suspensions with $b_z/b_x = 2$. For low area packing fraction ($\phi^{2D} = 0.2$) the orientation autocorrelation function decays faster than for a single swimmer, since steric and hydrodynamic interactions reorient swimmers seemingly randomly (see fig. 11.17 (b)). At area packing fraction $\phi^{2D} = 0.5$, where
11.5 Simulation results – spheroidal swimmers

Figure 11.14: Snapshots of spheroidal squirmers with \( b_z/b_x = 2 \) and \( \beta = -1 \) at the area packing fraction \( \phi^{2D} = 0.5 \). Figures (a) and (b) show different realizations that exhibit jamming and aligned swimming. The jammed cluster in figure (a) is rotating.

Figure 11.15: Polar pair correlation function for spheroidal squirmers with \( b_z/b_x = 2 \) at (a) \( \phi^{2D} = 0.5 \) and (b) \( \phi^{2D} = 0.2 \).
Figure 11.16: Local alignment coefficient $N$ as a function of the global area packing fraction $\phi^{2D}$. Red, black, blue, and magenta symbols correspond to simulations of pushers, neutrals, pullers, and active Brownian particles respectively. Circles and triangles correspond to the aspect ratios $b_z/b_x = 2$ and $b_z/b_x = 3$, respectively.

Figure 11.17: Orientation autocorrelation function $C_e(t)$ for spheroidal squirmers with $b_z/b_x = 2$ at (a) $\phi^{2D} = 0.5$ and (b) $\phi^{2D} = 0.2$. 
the squirmers form clusters, while the active Brownian particles do not, the orientation autocorrelation of squirmers has increased, whereas $C_e(t)$ slightly decreases for active Brownian particles (see fig. 11.17 (a)).

### 11.6 Spheroidal squirmers with a rotlet dipole

As a first study, we consider spheroidal pushers ($\beta = -1$) with aspect ratio $b_z/b_x = 2$ and include a rotlet dipole coefficient of $\kappa_{RD} = 4\sqrt{k_B T/ma^3}$. Note that the source dipole coefficient for these parameters is significantly smaller with $\kappa_{SD} = 0.1\sqrt{k_B T/ma^3}$ (see eq. (9.43)). As fig. 11.18 reveals, the result is very similar to that for $\beta = -5$: the cluster disappears. The rotlet dipole decays as $r^{-3}$ for $r \to \infty$ in contrast to the force dipole, which decays like $r^{-2}$. Nevertheless, it shows a strong influence on cluster formation, which is probably due to near field interactions.

![Figure 11.18: Probability density of local packing fraction for different swimmers.](image)

The figure corresponds to fig. 11.11 (g), but includes in addition simulations with $\beta = -1$ and $\kappa_{RD} = 4\sqrt{k_B T/ma^3}$ (dark green line).

Next, we try to match our parameters with those of *E. coli*. The aspect ratio of *E. coli*, including the cell body and the flagella, is around 10 [77]. The map to a spheroid is somewhat crude and it is not clear which aspect ratio to choose. We pick the aspect ratio $b_z/b_x = 4$, which is more than the aspect ratio of the cell body alone but less than the aspect ratio of the whole organism including the flagella. The minor axis of the cell body is around $b_x = 5 \times 10^{-7}$m. Assuming the aspect ratio $b_z/b_x = 4$, we can determine $B_1$ by equating the velocity of *E. coli*...
Figure 11.19: (a) Probability density of local packing fraction, (b) pair correlation function $C_1(r)$, and (c) orientation correlation function for spheroidal squirmers with $b_z/b_x = 4$, $b_x = 3a$, $B_1 = 0.002$, and $\beta = -2$. The red, green, blue, and magenta line correspond to $\kappa^{RD}/(\sqrt{k_B T/ma^3}) = 1, 2, 3$, and 4, respectively.
11.6 Spheroidal squirmers with a rotlet dipole

Figure 11.20: Snapshots of spheroidal squirmers with a rotlet dipole at the global area packing fraction $\phi^{2D} = 0.5$, with $b_x = 3a$, $b_z = 12a$, $B_1 = 0.002\sqrt{k_B T/m}$, $\beta = -2$, and $\kappa^{RD} = 1\sqrt{k_B T/ma^3}$ in (a) and $\kappa^{RD} = 4\sqrt{k_B T/ma^3}$ in (b).

coli $v = 22 \cdot 10^{-6} \text{m/s}$ [77] with the velocity $U_0$ (see eq. (9.30)) of a squirmer with the above major and minor axis. We obtain $B_1 = 2.4 \cdot 10^{-5} \text{m/s}$. The force dipole coefficient $\kappa^{FD}$ of E.coli was measured in ref. [77]. The authors report two forces of magnitude $F = 4.2 \times 10^{-13} \text{N}$ separated by a distance $l = 1.9 \cdot 10^{-6} \text{m}$. We assume the viscosity to be that of water at $25\degree \text{C}$, which is $\eta = 8.9 \cdot 10^{-4} \text{kg/(ms)}$. The resulting force dipole coefficient is thus [3]

$$\kappa^{FD} = \frac{lF}{8\pi \eta} = 3.6 \times 10^{-17} \text{m}^4/\text{s}. \quad (11.6)$$

This allows us to estimate $B_2$, such that our squirmer features the same force dipole coefficient (see eq. (9.42)). We obtain $B_2 = -5.1 \times 10^{-5} \text{m/s}$, which yields $\beta = B_2/B_1 = -2$. For the rotlet dipole, we are not aware of experimental measurements of $\kappa^{RD}$ via the flow field. In the experiments of [6], the radius of the circles that E. coli traces above a no-slip surface is reported to be around an order of magnitude larger than its body’s minor axis. In section 9.6 we found similar results, when choosing $\kappa^{RD} = O(1)\sqrt{k_B T/ma^3}$. Hence we vary $\kappa^{RD}$ from $\kappa^{RD} = 1\sqrt{k_B T/ma^3}$ to $\kappa^{RD} = 4\sqrt{k_B T/ma^3}$. Simulation results for 200 spheroidal squirmers at the global area packing fraction $\phi^{2D} = 0.5$, with $b_x = 3a$, $b_z = 12a$, $B_1 = 0.002\sqrt{k_B T/m}$, $\beta = -2$, and different $\kappa^{RD}$, are shown in fig. 11.19. We observe that an increasing $\kappa^{RD}$ leads to decreased alignment and finally suppresses cluster formation at $\kappa^{RD} = 4\sqrt{k_B T/ma^3}$ (see fig. 11.19 (a)). These results are in agreement to those for squirmers of aspect ratio 2 (see fig. 11.18). The disappearance of clustering is accompanied by a decrease of the polar pair correlation function and a faster decay of the orientation autocorrelation function (see fig. 11.19 (b) and (c)). Apparently the hydrodynamic interactions due to the rotlet dipole result in torques that reorient...
swimmers. This can be interpreted as a mechanism to yield an effectively decreased Péclet number, which would explain the disappearance of clusters. Figure 11.20 shows snapshots of the simulations exhibiting a cluster for $\kappa_{RD} = 1\sqrt{\frac{k_B T}{ma^3}}$ and a gas-like phase for $\kappa_{RD} = 4\sqrt{\frac{k_B T}{ma^3}}$. Note that the velocities in the gas-phase for high $\kappa_{RD}$ (see fig. 11.20 (b)) is quite high, which is due to the squirmers pushing each other around, which can be seen in videos of the simulation.

11.7 Summary and conclusions

We studied the collective dynamics of spherical and spheroidal squirmers confined in a narrow slit by MPC simulations. To elucidate the role of hydrodynamic interactions, we complemented our studies by simulations of spherical and spheroidal active Brownian particles, which do not interact hydrodynamically.

Contrary to ref. [30], but in accordance with ref. [29], we found that hydrodynamics suppresses motility-induced phase separation for spherical microswimmers. We attribute the contradiction with ref. [30] to a simulation artifact in that work. In ref. [29], where two-dimensional squirmers were studied, it was argued that hydrodynamics suppresses phase separation, since hydrodynamic torques during swimmer-swimmer collisions lead to a strong reorientation and hence an effectively decreased Péclet number. Our analysis of the orientation autocorrelation function suggests that the same argument holds for the quasi-two-dimensional system studied here.

Interestingly, for spheroidal squirmers, we found that hydrodynamics enhances – rather than suppresses – motility-induced phase separation. This result is surprising at first glance. However, we have to keep in mind that the mechanisms for cluster formation of isotropic and anisotropic swimmers are different. For anisotropic squirmers, alignment due to steric interactions is a key mechanism. Hydrodynamics may enhance this alignment for elongated microswimmers, and hence enhance their cluster formation. Our studies on the cooperative swimming of two pullers in a narrow slit in chapter 10 already suggested that hydrodynamics can lead to a persistent alignment of elongated microswimmers. In fact, we found that spheroidal pullers show the strongest tendency to form clusters. In ref. [31], simulations of dumbbell swimmers in three dimensions were performed, and it was reported that hydrodynamics enhances cluster formation. Since dumbbell swimmers are anisotropic, this finding matches with ours for spheroidal squirmers. However, we have to be careful with a too general conclusion. Our studies for very strong pushers ($\beta = -5$) and for swimmers with a high rotlet dipole mode show that certain hydrodynamic properties of elongated microswimmers can also suppress phase separation.

In our simulation studies, we did not observe the phenomenon of mesoscale turbulence reported for self-propelled rods in refs. [20, 209]. This is probably related to the fact that we considered around $O(10^3)$ swimmers, while ref. [20] reported turbulence for $O(10^4)$ or more self-propelled rods. Hence, the question how hydro-
dynamic interactions influence mesoscale turbulence remains open, until simulation results for larger systems of squirmers are available.
Bibliography


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Bibliography


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List of publications

In accordance with §5(3) of the doctoral regulations, extracts of this thesis have been published in the following articles:


Thereby, publication [A] corresponds to chapter 5, publication [B] corresponds to chapter 6, publication [C] corresponds to chapter 7, and publication [D] corresponds to chapters 9 and 10. In accordance with §5(3) of the doctoral regulations, I state my contribution to these publications:

[A] I did the research under the supervision of R. G. Winkler. The writing was a collaborative effort.

[B] In this publication, the relevance of angular momentum conservation in MPC is discussed based on the simulation of colloids and Couette flow. The simulations and analytical calculations regarding colloids are due to M. Yang, J. Hu, G. Gompper and M. Ripoll. I verified the analytical calculations, assisted in the interpretation of measured friction coefficients, and in writing under the supervision of R. G. Winkler. For the Couette flow, I carried out the analytical calculations and simulations under the supervision of G. Gompper, M. Ripoll, and R. G. Winkler. The writing was a collaborative effort.

[C] I did the research under the supervision of R. G. Winkler and G. Gompper. Elmar Westphal translated my simulation code to CUDA (Compute Unified
Device Architecture, a parallel computing platform for GPUs). The writing was a collaborative effort.

[D] I did the research under the supervision of R. G. Winkler and G. Gompper. Elmar Westphal translated my simulation code to CUDA and conceived several ideas to optimize the code. The writing was a collaborative effort.
Summary

In this thesis, we present multiparticle collision dynamics simulations of systems ranging from a single colloid to the collective dynamics of microswimmers. In multiparticle collision dynamics (MPC), point particles undergo streaming and subsequent collision steps, which mimics the microscopic behavior of fluids. While the shear viscosity of MPC has been extensively studied, the bulk viscosity has received little attention. We determine the bulk viscosity of different MPC algorithms analytically and by means of simulations. Moreover, we discuss the importance of angular momentum conservation in hydrodynamic simulations including slip boundary conditions – a pitfall that has been overlooked in the literature.

We study the implementation of colloids in MPC and discuss several method-related properties. The transition from a short time scale characterized by molecular chaos to the hydrodynamic time scale is studied on the basis of autocorrelation functions. In particular, we disprove the claim that the total diffusion coefficient is obtained as the sum of the short-time and hydrodynamic contributions. Furthermore, we perform simulations corresponding to a recent experiment of a colloid in a harmonic trap near a wall with no-slip boundary conditions. The background of this experiment is the idea that a colloid could function as a sensor for surface properties. Our simulations, together with the analytical results of another publication, falsify the theoretical analysis of said experiment.

The squirmer model is widely applied in analytical and numerical investigations on the dynamics of biological and artificial microswimmers. We extend the model to account for prolate spheroidal shapes, preserving the leading order hydrodynamic singularities and the property of being analytically solvable. Furthermore, we present an implementation of such a spheroidal squirmer in MPC. Subsequently, we study the dynamics of squirmers in a narrow slit, which corresponds to a quasi-two dimensional setup. For two pullers, which are squirmers with a negative force dipole coefficient, we find a stable fixed-point of cooperative swimming. For many squirmers we observe motility induced cluster formation. We find that hydrodynamics suppresses cluster formation for spherical squirmers while it enhances cluster formation for elongated swimmers. Furthermore, a negative force dipole coefficient (pullers) is advantageous for cluster formation, while a positive coefficient (pushers) is disadvantageous. Aside from that, further hydrodynamic singularities are important, e.g., the rotlet dipole, which suppresses cluster formation.
Acronyms

**AMC** Angular momentum conservation, see page 59

**AT** Anderson Thermostat, a MPC variant (MPC-AT), see page 55

**FACF** Autocorrelation function of the random force, see page 46

**LD** Langevin dynamics, see page 51

**LVACF** Longitudinal velocity autocorrelation function, see page 83

**MPC** Multiparticle collision dynamics, see page 8

**MPC+a** MPC with angular momentum conservation, see page 55

**MPC-a** MPC without angular momentum conservation, see page 55

**PSD** Power spectral density, see page 121

**SRD** Stochastic-rotation dynamics, a MPC variant (MPC-SRD), see page 55

**VACF** Velocity autocorrelation function, see page 46
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I thank all my colleagues at the Forschungszentrum Jülich for interesting discussions and the stimulating working atmosphere. In particular, I want to mention my past and present office mates Anoop Varghese, Chien-Cheng Huang, and Sebastian Sohn for interesting discussions about physics and our research projects, but also for off-topic conversations. I am grateful to Anoop Varghese and Adam Wysocki for kindly sharing their knowledge about physics and the MPC method. I enjoyed the cooperation with all scientists with whom I co-authored papers, including Mingcheng Yang, Jinglei Hu, Marisol Ripoll, Gerhard Gompper, and Elmar Westphal. I fondly remember the cooperation with Elmar Westphal on writing a GPU code for the simulation of spheroidal squirmers in MPC.

More than anyone else, I thank my partner Lena, whose love and support kept me going.
Curriculum Vitae

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Publications
since 2015 See List of publications on page 203.