The Raspberry Model for Hydrodynamic Interactions Revisited, Part II: The Effect of Confinement

Joost de Graaf, Ton Peter, Lukas P. Fischer, and Christian Holm

Institute for Computational Physics (ICP), University of Stuttgart, Allmandring 3, 70569 Stuttgart, Germany
(Dated: March 11, 2015)

The so-called ‘raspberry’ model refers to the hybrid lattice-Boltzmann (LB) and Langevin molecular dynamics scheme for simulating the dynamics of suspensions of colloidal particles, originally developed by V. Lobaskin and B. Dünweg, New J. Phys. 6, 54 (2004), wherein discrete surface points are used to achieve fluid-particle coupling. In this paper, we present a follow up to our study of the effectiveness of the raspberry model in reproducing hydrodynamic interactions in the Stokes regime for spheres arranged in a simple-cubic crystal [L. Fischer, et al., XX XX, xx (2015)]. Here, we consider the accuracy with which the raspberry model is able to reproduce such interactions for particles confined between two parallel plates. To this end, we compare our LB simulation results to established theoretical expressions and finite-element calculations. We show that there is a discrepancy between the translational and rotational diffusion constant, as also found in Part I of our joint publication. We further show that adding internal coupling points to the raspberry, can be used to correct said discrepancy in this geometry as well. Finally, we prove that the use of mixed boundary conditions, raspberry and bounce-back, accurately reproduces hydrodynamic interactions between a spherical colloid and planar walls up to roughly one LB lattice spacing.

I. INTRODUCTION

Here, we continue our investigation into the so-called ‘raspberry’ model, which was first introduced by Lobaskin and Dünweg. The raspberry model allows for the incorporation of a colloidal particle into the lattice-Boltzmann (LB) algorithm by discretizing the surface into point particles and utilizing the coupling scheme of Ahlrichs and Dünweg for these points. The model derives its name from the discretized nature of the colloid’s surface, which resembles a raspberry, when represented by molecular-dynamics (MD) beads, see Fig. 1 (right) for the original ‘hollow’ variant and Fig. 1 (left) for the improved ‘filled’ raspberry introduced in Ref. 3.

There are two geometries that are often used as a benchmarking test for the quality of a hydrodynamic solver, namely: a simple-cubic array of spheres and a sphere between two parallel plates. These geometries can be tackled by semi-analytic calculations in the Stokes limit and thus provide an excellent basis for comparison. The former is of particular interest as a toy model for fluid flow in a porous medium (at small sphere separations), while the latter is relevant to, for example, the field of hydrodynamic chromatography. In Ref. 3 we already demonstrated the raspberry’s ability to accurately capture hydrodynamic interactions in a simple-cubic arrangement. In this paper, we study the confining geometry of two parallel plates. This geometry is of particular interest, since it combines two types of LB boundary conditions: bounce-back boundaries for the no-slip plates and the raspberry model for the no-slip/moving spherical colloid.

For the comparison put forward in this manuscript we use a variety of literature results. The first analytic expressions for the diffusion coefficient of the sphere-plate system were derived by Faxen, who restricted his study to spheres constrained to the mid-plane. Brenner derived an expression for diffusion of a sphere close to a single wall (in the direction perpendicular to the wall), which was later extended to a parallel-plate geometry in Refs. 11 12. The aforementioned publications are but the tip of the iceberg when it comes

FIG. 1. (color online) Representation of the structure of the raspberry models used in our simulations, filled (left) and hollow (right), respectively. The central bead to which all other beads are connected via rigid bonds is shown using a green sphere. The blue spheres represent the beads that form the filled raspberry and the red ones give the surface beads used for the hollow variant. The radius of the beads is chosen to be smaller than the typical effective hydrodynamic radius to help visualize the internal structure.
to the semi-analytic and numerical work that has been carried out on these and comparable geometries. This includes, a.o., semi-analytic calculations, numerical simulations that exploit analytic approximations to capture hydrodynamic behavior, and simulations that employ coarse-grained hydrodynamic solvers to fully resolve hydrodynamic interactions. In addition, numerous experimental studies into the diffusive behavior of spheres under confinement, as well as other shapes, have been undertaken. However, it goes beyond the confines of this introduction to fully list and do justice to all these investigations.

In this paper, we use some of the above literature results to prove that the hollow and filled raspberry reproduce Stokesian hydrodynamic interactions for a sphere confined between two parallel plates. In agreement with Part I and Ref. [35], we find that there is a discrepancy between the effective translational and rotational hydrodynamic radius of the hollow raspberry. As in Part I, filling the raspberry resolves this issue. We also show that hydrodynamic interactions are properly captured by the raspberry by the raspberry-LB method up to one lattice spacing away from either wall. In the right limits, we were able to demonstrate that the mean-square displacement also yields the proper position dependence of the translational diffusion coefficient in a thermalized LB fluid. Finally, we used our high-quality numerical data to establish empirical relations for this position dependence on the effective translational and rotational hydrodynamic behavior.

In this section, we outline the modeling approaches used in our investigation, we refer the reader to Ref. [3].

A. Lattice-Boltzmann Parameters

The raspberry particles, constructed in the same way as in Ref. [3], were coupled to a lattice-Boltzmann (LB) fluid. We used a graphics processing unit (GPU), D3Q19, fluctuating multi-relaxation time (MRT) LB solver, which is attached to the MD software ESPResSo. The particle-fluid interaction proposed by Ahlrichs and Dünweg was used to couple the fluid to embedded MD beads. The particle coordinates were interpolated onto the lattice using a tri-linear scheme. Finally, no-slip walls are incorporated via the bounce-back rule, that is, population densities are reflected at the boundary nodes.

To keep our result as general as possible, we set the density of the fluid to \( \rho = 1m_0\sigma^{-3} \), the lattice spacing to \( 1\sigma \), the time step to \( \Delta t = 0.005\tau \), the (kinematic) viscosity to \( \nu = 1\sigma^2\tau^{-1} \), the bare particle-fluid friction to \( \zeta_0 = 25\tau^{-1} \), and the strength of the fluctuations to \( k_B T = 0.01\epsilon \), unless stated otherwise. Here, \( \sigma \) is the MD base unit of length, \( \tau \) is the MD base unit of time, and \( \epsilon \) is the MD base unit of energy, which corresponds to \( k_B T \), with \( k_B \) is the Boltzmann constant and \( T \) is the temperature. For further details on the parameters used, including the mass (with MD base unit of mass \( m_0 = \tau^2\epsilon/\sigma^2 \)) and rotational inertia tensors employed, we refer to Ref. [3].

We again switched off the Weeks-Chandler-Anderson (WCA) potential used to construct the raspberry particles, in order not to bias the simulations by introducing non-hydrodynamic interactions. No interactions between the particles and the wall, other than hydrodynamic ones, were present in our simulations.

B. Hydrodynamic Experiments

To assess the quality of the raspberry approximation in modeling the hydrodynamic properties of a colloid we performed several experiments. We use the term ‘quiescent’ to describe an un-thermalized (non-fluctuating) LB fluid. We set up the system of a particle confined between two parallel plates as follows. A channel of height \( H \) was simulated using \( H + 2 \) lattice points along the \( z \)-axis (one layer of boundary cells on either side). In the \( xy \)-plane the box had an extent \( L \) in both directions and periodic boundary conditions were applied in these directions. We used finite-size scaling to remove any dependence on \( L \), typically the results were sufficiently converged for \( L > 5H \). The bounce-back boundary condition was applied to the first and final layer of points, leading to an effective hydrodynamic channel height of \( H \) with a no-slip surface. The height was confirmed by Hagen-Poiseuille flow measurements. For all experiments the raspberry was initialized at a height \( z \), measured with
The following experiments for a spherical particle confined between two parallel plates were performed:

- **A velocity experiment** in a quiescent fluid, see Fig. 2(a). The particle was kept fixed at the initial position. Its velocity was held constant, by resetting the value in every time step; thus an effective moving boundary condition is obtained. The mobility was subsequently determined by measuring the force on the particle in the stationary state. Since the particle does not move, the position dependence of the translational diffusion coefficient parallel and perpendicular to the wall could be accurately assessed.

- **A force experiment** in a quiescent fluid, see Fig. 2(b). The particle was allowed to move freely. The position dependent diffusion coefficient for movement parallel to the walls could be determined via the Einstein-Smoluchowski relation by extracting the terminal velocity. It proved unnecessary to fix the particle in the z direction. The presence of the boundary breaks the symmetry of the system, when the particle is not in the center of the channel. Therefore, there can be rotational and translational cross-coupling terms in the grand hydrodynamic diffusion tensor (GHDT). To examine this we also measured the angular velocity of the particle.

- **A torque experiment** in a quiescent fluid, see Fig. 2(c). A torque was applied to the particle about an axis parallel (Fig. 2(c), left) or perpendicular (Fig. 2(c), right) to the walls and the angular velocity was measured. Again it proved unnecessary to fix the particle in the z direction.

- **An MSD experiment** in a thermalized fluid, see Fig. 2(d). The system was equilibrated until the particle fluctuated with the proper imposed fluid temperature. Then over many integration cycles (typically $10^8$ per run) and for several runs, we kept track of the position of the particle. The positions were binned according to the following procedure. Each bin was made half a lattice spacing ($0.5\sigma$) in width for the direction of interest. All sub-trajectories of our runs that originated in a specific bin were assigned to that bin and followed until they exited the bin. To ensure a minimum smearing out of the position dependence, we selected only those parts of the trajectories that went through a region $0.1\sigma$ in diameter around the center of the bin. This eliminated the trajectories that moved in and out of the bin but never crossed the center. For each selected trajectory, we determined the MSD and averaged this over all the relevant trajectories. The MSD was obtained by examining a trajectory of 50,000 integration steps from the starting point. Because we used a low temperature ($k_B T = 0.01\epsilon$), we were able to access the long-time diffusion regime without the particle traveling beyond the confines of the relevant bin, despite the long MSD sampling length.

---

**FIG. 2.** (color online) Visualization of the various hydrodynamic experiments carried out between two parallel plates, separated by a distance $H$. Here, a two-dimensional (2D) cross-section of the setups is shown. The blue arrows and symbols denote quantities applied to the fluid and raspberry, the red arrows and symbols indicate measured quantities, the green arrows indicate a spatially fixed raspberry. The black arrows signify a thermalized fluid. We refer to the text for a description of the experiments and applied and measured quantities.

- A velocity experiment in a quiescent fluid, see Fig. 2(a). The particle was kept fixed at the initial position. Its velocity was held constant, by resetting the value in every time step; thus an effective moving boundary condition is obtained. The mobility was subsequently determined by measuring the force on the particle in the stationary state. Since the particle does not move, the position dependence of the translational diffusion coefficient parallel and perpendicular to the wall could be accurately assessed.

- **A force experiment** in a quiescent fluid, see Fig. 2(b). The particle was allowed to move freely. The position dependent diffusion coefficient for movement parallel to the walls could be determined via the Einstein-Smoluchowski relation by extracting the terminal velocity. It proved unnecessary to fix the particle in the z direction. The presence of the boundary breaks the symmetry of the system, when the particle is not in the center of the channel. Therefore, there can be rotational and translational cross-coupling terms in the grand hydrodynamic diffusion tensor (GHDT). To examine this we also measured the angular velocity of the particle.

- **A torque experiment** in a quiescent fluid, see Fig. 2(c). A torque was applied to the particle about an axis parallel (Fig. 2(c), left) or perpendicular (Fig. 2(c), right) to the walls and the angular velocity was measured. Again it proved unnecessary to fix the particle in the z direction.

- **An MSD experiment** in a thermalized fluid, see Fig. 2(d). The system was equilibrated until the particle fluctuated with the proper imposed fluid temperature. Then over many integration cycles (typically $10^8$ per run) and for several runs, we kept track of the position of the particle. The positions were binned according to the following procedure. Each bin was made half a lattice spacing ($0.5\sigma$) in width for the direction of interest. All sub-trajectories of our runs that originated in a specific bin were assigned to that bin and followed until they exited the bin. To ensure a minimum smearing out of the position dependence, we selected only those parts of the trajectories that went through a region $0.1\sigma$ in diameter around the center of the bin. This eliminated the trajectories that moved in and out of the bin but never crossed the center. For each selected trajectory, we determined the MSD and averaged this over all the relevant trajectories. The MSD was obtained by examining a trajectory of 50,000 integration steps from the starting point. Because we used a low temperature ($k_B T = 0.01\epsilon$), we were able to access the long-time diffusion regime without the particle traveling beyond the confines of the relevant bin, despite the long MSD sampling length.
In all of the above experiments, care was taken to ensure that the particle remained in the low translational and rotational Reynolds number regime

\[ Re^T = \frac{vR}{\nu} \ll 1; \quad (1) \]
\[ Re^R = \frac{\omega R^2}{\nu}, \quad (2) \]

with \( v \) and \( \omega \) the maximum/typical (angular) velocity, respectively.

C. Finite-Elements Calculations

In order to compare the quality of our results for the raspberry particles to reference curves, we utilized literature values from analytic and numerical studies whenever possible. However, for the sphere confined between two plates, literature results were often not available, to the best of our knowledge. Therefore, we performed finite-element calculations using the COMSOL 4.4 MultiPhysics Modeling Software to establish reference data.

We set up a system with a colloid of radius \( R = 0.5 \) \( \mu m \) and solved the Navier-Stokes equations for an incompressible fluid with density \( \rho = 10^3 \) \( kg \cdot m^{-3} \) and dynamic viscosity \( \eta = 10^{-3} \) \( kg \cdot m^{-1} \cdot s^{-1} \). We neglected the non-linear term, since we are interested in \( Re^X \ll 1 \) (laminar flow), where \( X = T \) or \( R \). The plate separation was varied between \( H = 1.1 \) \( \mu m \) and \( 10.0 \) \( \mu m \). We used a cylindrical domain with open boundary conditions (zero normal stress) for the fluid and no-slip conditions for the plates (the bases of the cylinder). The diameter of the plates was varied to remove finite-size effects, but we found that a diameter of \( 10H \) was sufficient to guarantee convergence.

To ensure that the laminar flow condition is satisfied, we used (angular) velocity boundary conditions on the sphere with low values of the (angular) velocity: \( v = 10^{-6} \) \( m \cdot s^{-1} \) and \( \omega = 10^{-6} \) \( s^{-1} \). We solved for the force and torque exerted by the fluid and plates on the sphere and used the Einstein-Smoluchowski relations to derive the relevant diffusion coefficients. The sphere was located on the symmetry axis of the cylinder, and we varied the position with respect to the plates along this axis keeping the sphere’s position fixed in the other directions. Similarly, we examined the plate-separation dependence of the diffusion coefficient by keeping the sphere fixed in the center of the cylinder and moving the plates outward.

Whenever possible, we exploited the rotational symmetry of the system to minimize the number of elements required, i.e., for perpendicular movement and rotation about an axis perpendicular to the wall. For the full three-dimensional (3D) geometries, constructing a sufficiently refined mesh that contained a manageable number of elements proved difficult. We used local refinement with elements 1 nm in size over the surface of the particle and in the disks with radius \( R \) around the center of the plates. The size of the elements was allowed to expand radially outward. We verified that integration over the surface yielded the surface area to within a fractional deviation of \( 10^{-5} \). We therefore expect that similar integration of the force and torque over the surface would show limited deviation between the true solution of the boundary value problem and our numerical result. To further demonstrate the quality of our approximate solution, we performed mesh refinement for a selected number of configurations and found no significant change.

D. Notations Used throughout this Manuscript

In this section, we summarize the notations used throughout this manuscript. This will aid in the understanding of our results, as many of the notations are necessarily similar.

- \( H \), the plate separation for the confined channel system, see Fig. 2(a).
- \( R^h \), the effective hydrodynamic radius obtained by extrapolating translational diffusion measurements, see Figs. 2(a,b), for the limit of plate separation \( H \uparrow \infty \). The subscript \( h \) is used to differentiate \( R_h \) from the raspberry radius \( R \).
- \( R^R \), the effective hydrodynamic radius obtained by extrapolating rotational diffusion measurements, see Fig. 2(c), for the limit of plate separation \( H \uparrow \infty \).
- \( z \), the vertical position of the sphere with respect to the center of the channel \( (z = 0) \), see Fig. 2(a).
- \( z_{\text{max}} \), the maximum distance that a sphere can move from the center \( (z = 0) \) before making contact with a wall: \( z_{\text{max}} = H/2 - R^h \) and \( z_{\text{max}} = H/2 - R^R \), respectively, see Fig. 2(a).
- \( D^T_0 \), the bulk translational diffusion coefficient.
- \( D^R_0 \), the bulk rotational diffusion coefficient.
- \( D^T \), the translational diffusion coefficient for movement along an axis perpendicular to the plates as a function of \( z \) for fixed plate separation, see Fig. 2(a).
- \( D^T_{z,\text{perp}} \), the translational diffusion coefficient for movement along an axis parallel to the plates and as a function of \( z \) for fixed plate separation, see Fig. 2(b).
- \( D^T_{z,\text{para}} \), the translational diffusion coefficient for movement along an axis parallel to the plates as a function of \( z \) for fixed plate separation, see Fig. 2(c) left.
- \( D^R \), the rotational diffusion coefficient for rotation about an axis perpendicular to the plates as a function of \( z \) for fixed plate separation, see Fig. 2(c) right.
- \( D^R_{z,\text{perp}} \), the rotational diffusion coefficient for rotation about an axis parallel to the plates as a function of \( z \) for fixed plate separation, see Fig. 2(c) right.
- $D_{H,\text{perp}}^T$, the translational diffusion coefficient for movement along an axis perpendicular to the plates as a function of the plate separation $H$ for a sphere in the center of the channel $z = 0$, see Fig. 2(a).
- $D_{H,\text{para}}^T$, the translational diffusion coefficient for movement along an axis parallel to the plates as a function of the plate separation $H$ for a sphere in the center of the channel $z = 0$, see Fig. 2(b).
- $D_{H,\text{perp}}^R$, the rotational diffusion coefficient for rotation about an axis perpendicular to the plates as a function of the plate separation $H$ for a sphere in the center of the channel $z = 0$, see Fig. 2(c) left.
- $D_{H,\text{para}}^R$, the rotational diffusion coefficient for rotation about an axis parallel to the plates as a function of the plate separation $H$ for a sphere in the center of the channel $z = 0$, see Fig. 2(c) right.
- $f$, the fractional deviation between two results.

One final point of possible confusion is the use of the terms ‘parallel’ and ‘perpendicular’ to the plates. By parallel, we mean motion in a direction that is along the plates (orthogonal to the normal that defines the plates). By perpendicular motion, we mean in the direction parallel to this normal, i.e., away from the plates. For rotation experiments these terms are used to describe the orientation of the axis of rotation with respect to the plates. Please refer to Fig. 2 and the above list if there is any confusion in reading the text.

III. RESULTS

In this section, we discuss the results that we obtained by performing the simulations and numerical calculations outlined in Section II.

A. Perpendicular Translational Motion between Two Plates

Let us begin by considering motion perpendicular to the plates, see Fig. 2(a). That is, motion along the normal vector that defines the plate orientation. This is the simplest case, since there are no translation-rotation cross-coupling terms in the GHDT for this direction. Moreover, there are analytic expressions for the positional dependence $z$ of the perpendicular translational diffusion coefficient $D_{z,\text{perp}}^T$. These follow from the result derived by Brenner for perpendicular diffusion near a single wall [10], see Refs. [11, 12] for the extension to two plates. N.B. Both Refs. [11, 12] contain a typo in formulating the expression in Ref. [10]. The results shown in this section are for a filled raspberry model. However, we also considered a hollow raspberry and we will comment on the agreement between the two models at the end of this section.

![Figure 3](attachment:fig3.png)

**FIG. 3.** (color online) The translational diffusion coefficient perpendicular to the channel walls $D_{z,\text{perp}}^T$ for a sphere of radius $R$ moving through a channel of height $H = 14\sigma$. The diffusion coefficient is normalized by the bulk translational diffusion coefficient $D_0^T$ and is given as a function of the position in the channel $z$, which itself is normalized by $z_{\text{max}}$. The parameter $z_{\text{max}}$ is the maximum distance that the sphere can move from the center ($z = 0$) before making contact with a wall, see the main text for further details. (a) The blue circles show the results of quiescent LB simulations, the green squares with error bars the results of thermalized LB simulations, and the dashed red curve the semi-analytic result. [10, 12] The dashed vertical line (thick black) indicates the position for which the sphere and the wall are separated by one LB lattice spacing. (b) The fractional deviation $f$ of the quiescent LB result from the theoretical result. The horizontal gray line shows a fractional deviation of 2.5%.

Figure 3(a) shows the result of our quiescent velocity and thermalized MSD experiments, Figs. 2(a) and 2(d), respectively, compared to the semi-analytic value of $D_{z,\text{perp}}^T$. Here, we used a channel height of $H = 14\sigma$, in which we placed a spherical raspberry with radius $R = 3\sigma$. As can be seen in Fig. 3(b) the agreement between our quiescent simulations and the corrected series expression is quite excellent. There is a fractional devia-
tion $f$ of less than 1% throughout the channel, up to the point where the raspberry and the channel walls are separated by less than $1\sigma$. For such small separations there is a systematic deviation between the theory and our LB results, see Fig. 3(a). This deviation can be attributed to the break-down of the description in terms of an effective hydrodynamic radius in proximity of a wall (within one lattice spacing). We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing). We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing). We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing). We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing). We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing).

We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing). We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing). We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing). We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing). We will come back to the effective hydrodynamic radius in proximity of a wall (within one lattice spacing).

In order to determine the value of the effective hydrodynamic radius, we varied the distance between the plates and measured $D_{z,\text{perp}}^T$ at $z = 0$ (the middle of the channel), where the diffusion coefficient attains its highest value. We denote this plate-separation dependent diffusion coefficient by $D_{H,\text{perp}}^T$. The result of our simulations, compared to the theoretical expression of Refs. [10–12] is shown in Fig. 3.

Note that agreement between the two data sets is quite excellent for $H > 2R_h^L + \sigma$. Figure 3(b) shows systematic deviation between the theory and numerical results, but it is small. The structure in $f$ originates from the fitting procedure used to extract the bulk diffusion coefficient and its subsequent use as a normalization factor. Furthermore, the model breaks down for smaller separations, as can be clearly seen in Fig. 3(a), since sub-lattice changes in the plate separation cannot be obtained using the bounce-back boundaries. Finally, the effective radii that we obtained for the filled and hollow raspberry data were close to the ones that we obtained from our simple-cubic geometry, see Table II as we will detail in Section IV. For both the hollow and the filled raspberry model, the dependence of $D_{z,\text{perp}}$ and $D_{H,\text{perp}}^T$ on $z$ and $H$, respectively, is the same within the error bar (not shown here).

### B. Parallel Translational Motion between Two Plates

We continued our investigation into the quality of the raspberry model under confinement by examining the diffusion coefficient in the direction parallel to the plates, see Fig. 2(b). That is, motion perpendicular to the normal vector that defines the plate orientation. For such motion there are translation-rotation cross-coupling terms in the GHDT. [40] Only when the sphere moves in the plane centered between the two plates is there no coupling, due to symmetry.

For parallel motion in this center plane there are analytic expressions for height dependence of the parallel translational diffusion coefficient $D_{H,\text{para}}^T$ derived by Fuxen [8]. These are purportedly accurate up to fifth order in the plate separation, see Eq. (3). However, the fifth order coefficient was only provided in a private communication [9] and no account of its derivation exist. In order to further verify our results we compared our raspberry data to finite-element calculations, the data points of which we fitted using a fourth-order polynomial. Again, we only show results for the filled raspberry model in this section.

Figure 5(a) shows the result of our quiescent velocity
FIG. 5. (color online) The translational diffusion coefficient parallel to the channel walls \( D_{T,\text{para}} \) for a sphere (\( R = 3\sigma \)) moving through a channel of height \( H = 14\sigma \), plotted as a function of the position \( z \). The notation is otherwise the same as in Fig. 3. (a) Quiescent LB simulations (blue circles), thermalized LB simulations (green squares with error bars), and COMSOL results (dashed red curve). (b) The fractional deviation \( f \) between the quiescent LB and COMSOL result (gray line \( f = 2.5\% \)).

FIG. 6. (color online) The translational diffusion coefficient parallel to the channel walls in the center of the channel \( D_{T,\text{para}} \) for a sphere of radius \( R = 3\sigma \) moving through a channel of height \( H \). The notation is otherwise the same as in Fig. 4. (a) The blue circles show the data from quiescent LB simulations, the dashed green curve the theoretical approximation by Faxen \([8, 9]\), and the dashed red curve the result of finite-element calculations performed using COMSOL. (b) The fractional deviation \( f \) of the quiescent LB result from the theoretical result by Faxen (green squares) and the COMSOL result (red circles). The horizontal gray line shows a fractional deviation of 2.5%.

To determine the bulk diffusion coefficient and the associated effective hydrodynamic radius, we examined \( D_{T,\text{para}} \) for parallel motion in the center plane. The result of our simulations is shown in Fig. 8, which shows a comparison between our data, the theoretical result by Faxen, \([8, 9]\) and the COMSOL finite-element calculations. The result obtained using the raspberry model agrees perfectly with the finite-element data over the entire range of plate separations considered here. As ex-
expected there is a significant deviation between Faxen’s result for small plate separations, since the theory ignores higher-order contributions in \( H \) to the diffusion constant. We only find acceptable agreement between our data and Faxen’s expression for \( H \gtrsim 4R^2_h \). From our numerical results a modified empirical form of Faxen’s law can be derived which fits the data better over the range \( H > 2R^2_h + \sigma \)

\[
\frac{D_{R,\text{para}}^T}{D_0^T} = A + Bx + Cx^2 + Dx^3 + Ex^4 + Fx^5
\]

\( x \equiv \frac{2R^2_h}{H} \);

(3)

\[
A = +0.997 \pm 0.001; \quad A = +1.000;
\]

(4)

\[
B = -0.949 \pm 0.009; \quad B = -1.004;
\]

(5)

\[
C = -0.215 \pm 0.034; \quad C = +0.000;
\]

(6)

\[
D = -0.542 \pm 0.025; \quad D = +0.418;
\]

(7)

\[
E = +0.895 \pm 0.051; \quad E = +0.210;
\]

(8)

\[
F = +0.000 \pm 0.000; \quad F = -0.169,
\]

(9)

where the first column of numbers with error bars gives our numerically obtained coefficients, and the second column the coefficients determined by Faxen. Note that we used a different form of polynomial expansion (leaving in the \( x^2 \) term) to achieve a compact expression. The first coefficient \( A \) can be taken to be 1 without significantly changing the result of the fitting procedure.

Again we found that for both the hollow and the filled raspberry model the dependence of \( D_{R,\text{para}}^T \) and \( D_{H,\text{para}}^T \) on \( z \) and \( H \), respectively, is the same within the error bar (not shown here). The value of the effective radii for the two models differed, though, see Table I. This is in agreement with our findings for the perpendicular rotation of our knowledge no analytic expressions for rotational coupling, due to symmetry considerations. To the best of our knowledge there are no analytic expressions for rotational diffusion coefficients between two plates exist. Therefore, we compare to finite-element calculations, the data points of which we fitted using a fourth-order polynomial. We only show results for the filled raspberry model for the position \( z \) and height \( H \) dependence of the diffusion coefficients.

Figure 7(a) shows the result of our quiescent torque experiments compared the data obtained using COMSOL for both forms of rotation. Here, we used a channel height of \( H = 14\sigma \) in which we placed a spherical raspberry with radius \( R = 3\sigma \). As can be seen in Fig. 7(b) the agreement between our quiescent simulations and the finite-element calculations is quite excellent. The fractional deviation \( f \) is less than 1% throughout the channel for rotation about an axis parallel to the wall and even better for rotation about an axis perpendicular to the wall. We can see significant deviation for the former when the raspberry and the channel walls are separated by less than 1\( \sigma \), but

\[
D_{z,\text{perp}}^R = B x^2 + C x^3 + D x^4 + E x^5 + F x^6
\]

\( x \equiv \frac{2R^2_h}{H} \);

(11)

\[
A = +0.994 \pm 0.009; \quad A = +1.000;
\]

(12)

\[
B = +0.949 \pm 0.009; \quad B = -1.004;
\]

(13)

\[
C = -0.215 \pm 0.034; \quad C = +0.000;
\]

(14)

\[
D = +0.542 \pm 0.025; \quad D = +0.418;
\]

(15)

\[
E = +0.895 \pm 0.051; \quad E = +0.210;
\]

(16)

\[
F = +0.000 \pm 0.000; \quad F = -0.169,
\]

(17)

where the first column of numbers with error bars gives our numerically obtained coefficients, and the second column the coefficients determined by Faxen. Note that we used a different form of polynomial expansion (leaving in the \( x^2 \) term) to achieve a compact expression. The first coefficient \( A \) can be taken to be 1 without significantly changing the result of the fitting procedure.

Again we found that for both the hollow and the filled raspberry model the dependence of \( D_{z,\text{perp}}^R \) and \( D_{z,\text{para}}^R \) on \( z \) and \( H \), respectively, is the same within the error bar (not shown here). The value of the effective radii for the two models differed, though, see Table I. This is in agreement with our findings for the perpendicular motion between plates and in the simple-cubic geometry. We will discuss this further in Section IV.

C. Rotational Motion between Two Plates

Finally, we considered the accuracy of the raspberry model under confinement for the case of rotation about an axis in the direction parallel and perpendicular to the plates, see Fig. 2(c). For rotation about an axis parallel to the wall (perpendicular to the normal, see Fig. 2(c) (left)) there are rotation-translation cross-coupling terms in the GHDT. Only when \( z = 0 \) is there no such coupling, due to symmetry considerations. To the best of our knowledge no analytic expressions for rotational diffusion coefficients between two plates exist. Therefore, we compare to finite-element calculations, the data points of which we fitted using a fourth-order polynomial. We only show results for the filled raspberry model for the position \( z \) and height \( H \) dependence of the diffusion coefficients.

Figure 7(a) shows the result of our quiescent torque experiments compared the data obtained using COMSOL for both forms of rotation. Here, we used a channel height
this can be explained by the small particle-wall separation compared to the lattice spacing. Again, we did not observe translation-rotation cross coupling in our data.

Note that there is some structure to the deviation between the parallel-rotation raspberry data and the finite element calculations. This is related to the fitting procedure used to extract the bulk rotational diffusion coefficient \( D_R^R \) by varying \( H \). A small deviation in the effective hydrodynamic radius \( R_H^R \) can have significant impact on the shape of the curve, as we will explain shortly. For the rotation about an axis perpendicular to the wall there is a similar increase when the wall-raspberry separation is less than 1\( \sigma \), but the agreement is still quite reasonable.

To determine the bulk diffusion coefficient and the associated effective hydrodynamic radius, we examined \( D_{H,\perp}^R \) and \( D_{H,\parallel}^R \) (rotation in the mid-plane with varying \( H \)), see Fig 8. Again we find quite acceptable agreement between the data obtained using the raspberry model and the finite-element calculations. However, we can clearly see that there is structure to the fractional deviation \( f \) between the two results in Fig. 8(c). This can be explained as follows.

In determining the effective hydrodynamic radius, we used a quartic polynomial to fit our data, which was given as a function of \( 2R_H^R/H \) and normalized by the value of \( D_R^R \) corresponding to that \( R_H^R \). That is, the data which we fitted is a function of \( R_H^R \) and we applied a root-finding algorithm to ensure that the fitted value for \( H \) equals 1 of \( D_{H,\perp}^R/D_0^R \) and \( D_{H,\parallel}^R/D_0^R \) equals 1. We thus solved for the value \( R_H^R \).

The above procedure, however, strongly favors the criterion imposed on the extrapolated data point, while it sacrifices overall matching to the finite-element calculations – which we assume to be more accurate. Hence, there can be a systematic deviation between the two data sets, as is indeed found in Fig. 8(c). However, our fitting procedure is reasonable, since it does not implicitly assumes the finite-element data to be superior to that obtained using the raspberry model and it only uses internal fitting parameters. The agreement between the two results, despite the systematic deviations, can therefore be considered excellent.

From our numerical results we can formulate modified empirical forms for the rotational diffusion coefficient’s dependence on the channel height \( H \), for mid-channel rotation about an axis parallel and perpendicular to the walls. These expressions are as follows

\[
\frac{D_{H,i}^R}{D_0^R} = 1 + A_i x^2 + B_i x^3 + C_i x^4; \tag{11}
\]

\[
x = \frac{2R_H^R}{H}; \tag{12}
\]

\[
A_{\perp} = -0.10 \pm 0.01; \quad A_{\parallel} = -0.09 \pm 0.01; \tag{13}
\]

\[
B_{\perp} = +0.09 \pm 0.03; \quad B_{\parallel} = -0.17 \pm 0.04; \tag{14}
\]

\[
C_{\perp} = -0.26 \pm 0.02; \quad C_{\parallel} = -0.38 \pm 0.03. \tag{15}
\]

where \( i \) is a subscript that can be either ‘perp’ for rotation about an axis perpendicular to the walls and ‘para’ for rotation about an axis parallel to the walls. We ignored

![FIG. 8. (color online) The perpendicular (a) and parallel (b) rotational diffusion coefficient \((D_{H,\perp}^R, D_{H,\parallel}^R)\) for a sphere of radius \( R = 3\sigma \) moving through a channel of height \( H \). The diffusion coefficients are normalized by the bulk coefficient \( D_R^R \) and plotted as a function of \( 1/H \), expressed in terms of twice the hydrodynamic radius \( R_H^R \). The notation is otherwise the same as in Figs. 4 and 6. Quiescent LB simulations are given by blue circles and COMSOL results using dashed red curves. (c) The fractional deviation \( f \) of the quiescent LB results from the COMSOL data (gray line \( f = 2.5\% \)).](image-url)
TABLE I. Summary of the various effective radii determined in the experiments described in Section III and Ref. [3]. There is a \( \pm 0.01\sigma \) error bar for the translational experiments and \( \pm 0.02\sigma \) for the rotational experiments. The bare friction coefficient used to generate this data is \( \zeta_0 = 25\tau^{-1} \).

|               | filled          | hollow         |
|---------------|-----------------|----------------|
|               | \( R^0 \)      | \( R^0 \)      |
| simple-cubic  | 3.53\(\sigma\) | 3.50\(\sigma\) |
|               | 3.47\(\sigma\) | 3.38\(\sigma\) |
| walls \( \perp \) | 3.53\(\sigma\) | 3.50\(\sigma\) |
|               | 3.48\(\sigma\) | 3.44\(\sigma\) |
| walls \( \parallel \) | 3.53\(\sigma\) | 3.51\(\sigma\) |
|               | 3.47\(\sigma\) | 3.37\(\sigma\) |

The linear coefficient due to the functional form of our result.

Again we found that for both the hollow and the filled raspberry model the dependence of \( D^R_z, D^R_{\text{para}}, D^R_{H,\text{perp}}, \) and \( D^R_{H,\text{para}} \) on \( z \) and \( H \), respectively, is the same within the error bar (not shown here). The value of the effective radii for the two models differed, see Table I in agreement with our findings for the perpendicular and parallel motion between plates, as well as in the simple-cubic geometry. [3] We will discuss this further in Section IV.

IV. DISCUSSION

In Section III we have shown that there is excellent agreement between established theoretical and numerical results for the Stokesian hydrodynamic behavior of spherical particles compared to their raspberry equivalent in the confining geometry of two parallel plates. This extends our findings in Part I [3] to geometries with walls. The discrepancy between the translational and rotational hydrodynamic radius of the original hollow raspberry – first observed by Ollila et al. [35] – is present for the parallel plate system as well. In this section we discuss this discrepancy in more detail.

In Table I we summarize the results for the hydrodynamic experiments obtained in our experiments. Note that within the error bar the data for the filled raspberry is fully consistent, whereas the data from the hollow raspberry shows a clear discrepancy between \( R^0 \) and \( R^0 \). The \( R^0 \) obtained from the experiment with rotation about an axis perpendicular to the wall (Fig. 2(c), right) seems to be an outlier for both the filled and hollow raspberry, but it is still within two standard deviations of the mean for the filled raspberry. As explained in Section III C our fitting procedure ensured that for the box length \( L \) \( \perp \) \( \infty \) in the simple-cubic geometry [3] and for the plate separation \( H \) \( \perp \) \( \infty \) in the confined geometry, the fitted result converges to the expected bulk diffusion coefficient for a given hydrodynamic radius, for which we subsequently solved. Therefore, the rotational data has a higher standard deviation in general, as taking the cubic root to establish the effective radius leads to stronger error propagation.

From our results, we conclude that the level of disagreement between the effective radii of the hollow raspberry model seems to be unaffected by the confinement. We have also shown that filling the raspberry model again significantly improves the agreement between the effective hydrodynamic radius obtained by translational and rotational experiments. There is agreement between the effective radii that are found for the filled raspberry using the various experiments, within the numerical error. A similar conclusion cannot be reached for the hollow raspberry, indicating that this model is more sensitive to the details of its surroundings.

V. CONCLUSION AND OUTLOOK

Summarizing, we have examined the properties of the raspberry model using a classic fluid dynamics experiments, wherein a sphere is subjected to the confining geometry of parallel plates. This so-called ‘raspberry’ model refers to a hybrid lattice-Boltzmann (LB) and Langevin molecular dynamics (MD) scheme for simulating the dynamics of suspensions of colloids originally developed by Lobaskin and Dünweg. [1] The particle is represented by a set of points on its surface that couple to the fluid through a frictional force acting both on the solvent and on the solute, which depends on the relative velocity. Our results show that the proper hydrodynamic behavior in this limit is reproduced to a surprising degree of accuracy over a wide range of length scales. We have also used our numerical data to determine empirical relations for the position dependence of the translational and rotational diffusion coefficients of a sphere confined between two plates.

From our combined data we can draw the following additional conclusions, as an extension of the conclusions already presented in Ref. [3], concerning the quality of the raspberry model.

- To determine the bulk translational diffusion coefficient of a particle, a force or velocity experiment performed in the center of two plates is more suitable than in a simple-cubic geometry. This type of experiment does not have the disadvantage of a back force/velocity density, [3] which could interfere with the fitting procedure required to extrapolate results to bulk (infinite plate separation).
- The raspberry point-coupling can be effectively combined with bounce-back boundary conditions. The combination yields accurate results over a large range of particle-boundary separations, as verified for the specific geometry of two parallel plates.
- We estimate the raspberry to accurately reproduce hydrodynamics interactions for particle-particle and particle-surface separations greater than \( \sigma \),
with $\sigma$ the LB lattice spacing. The separation is determined by the effective radii. Here, we consider the result of our rotational diffusion experiments and translational diffusion experiments in confinement significant, as there are no back-torques or back-forces applied to the fluid. Our estimate does not seem unreasonable, since the near-field effects of discretizing the surface into a finite number of MD beads with an effective hydrodynamic radius of approximately $\sigma$ should become noticeable at comparable separations. There is one caveat, however. In the case of a rotating raspberry the motion and structure of the raspberry are such that the fluid is constantly exposed to different fluid-coupling points (the MD beads). This may remove artifacts caused by the discretization of the surface (or volume) that are more noticeable in the translational experiment, where the orientation is fixed.

From the above and our examination in Part I, it becomes clear that the raspberry model is an excellent way to approximate fluid-particle coupling in an LB algorithm even in confining geometries where no-slip moving raspberry and bounce-back boundary conditions are mixed.

ACKNOWLEDGEMENTS

J.d.G. acknowledges financial support by a “Nederlandse Organisatie voor Wetenschappelijk Onderzoek” (NWO) Rubicon Grant (#680501210). We thank the “Deutsche Forschungsgemeinschaft” (DFG) for financial funding through the SPP 1726 “Microswimmers – from single particle motion to collective behavior”. We are also grateful to L. Helden and O. Hickey for useful discussions.

[1] V. Lobaskin and B. Dünweg, New J. Phys. 6, 54 (2004).
[2] P. Ahlrichs and B. Dünweg, J. Chem. Phys. 111, 8225 (1999).
[3] L. Fischer, T. Peter, C. Holm, and J. de Graaf, J. Chem. Phys. submitted (2015).
[4] J. Hofman, H. Clercx, and P. Schram, Physica 268, 353 (1999).
[5] J. Giddings, Separ. Sci. Technol. 13, 241 (1978).
[6] R. Noel, K. Gooding, F. Regnier, C. Orr, and M. Mullins, J. Chromatogr. A 166, 373 (1978).
[7] B. Dünweg and A. Ladd, Lattice boltzmann simulations of soft matter systems, in Advanced Computer Simulation Approaches for Soft Matter Sciences III, edited by C. Holm and K. Kremer, volume 221 of Adv. Polymer Sci., pages 89–166, Springer (Berlin/Heidelberg), 2009.
[8] H. Faxen, Ann. Phys. (Leipzig) 68, 89 (1922).
[9] J. Happel and H. Brenner, Low Reynolds Number Hydrodynamics, Springer (Berlin/Heidelberg), 2nd edition, 1983.
[10] H. Brenner, Chem. Eng. Sci. 16, 242 (1961).
[11] L. Lobry and N. Ostrowsky, Phys. Rev. B 53, 12050 (1996).
[12] B. Lin, J. Yu, and S. Rice, Phys. Rev. E 62, 3909 (2000).
[13] M. O’Neill, Mathematika 11, 67 (1964).
[14] N. Liron and S. Mochon, J. Eng. Math. 10, 287 (1976).
[15] B. Cichocki and R. Jones, Physica A 258, 273 (1998).
[16] P. Kalinay, J. Chem. Phys. 141, 144101 (2014).
[17] E. Dufresne, D. Altman, and D. Grier, Euro. Phys. Lett. 53, 264 (2001).
[18] J. Swan and J. Brady, J. Fluid Mech. 687, 254 (2011).
[19] L. Pasol et al., Chem. Eng. Sci. 66, 4078 (2011).
[20] Y. von Hansen, M. Hinczewski, and R. Netz, J. Chem. Phys. 134, 235102 (2011).
[21] J. Bleibel, A. Dominguez, F. Gunther, J. Harting, and M. Oettel, Soft Matter 10, 2945 (2014).
[22] W. Mitchell and S. Spagnolie, arXiv 1409.8208, 1 (2014).
[23] J. Padding and W. Briels, J. Chem. Phys. 132, 054511 (2010).
[24] A. Nikoubashman, C. Likos, and G. Kahl, Soft Matter 9, 2603 (2013).
[25] R. Tatsumi and R. Yamamoto, J. Chem. Phys. 138, 184905 (2013).
[26] A. Dubov, S. Schmieschek, E. Asmolov, J. Harting, and O. Vinogradova, J. Chem. Phys. 140, 034707 (2014).
[27] G. Volpe, L. Helden, T. Brettschneider, J. Wehr, and C. Bechinger, Phys. Rev. Lett. 104, 170602 (2010).
[28] T. Brettschneider, G. Volpe, L. Helden, J. Wehr, and C. Bechinger, Phys. Rev. E 83, 041113 (2011).
[29] S. Dettmer, S. Pagliara, K. Misiumas, and U. Keyser, Phys. Rev. E 89, 062305 (2014).
[30] S. Dettmer, U. Keyser, and S. Pagliara, Rev. Sci. Instr. 85, 023708 (2014).
[31] M. Lisicki, B. Cichocki, S. Rogers, J. Dhont, and P. Lang, Soft Matter 10, 4312 (2014).
[32] A. Chakrabarty, F. Wang, C.-Z. Fan, K. Sun, and Q.-H. Wei, Langmuir 29, 14396 (2013).
[33] J. Jones, J. van der Maarel, and P. Doyle, Phys. Rev. Lett. 110, 068101 (2013).
[34] A. Chakrabarty et al., Langmuir 30, 13844 (2014).
[35] S. Ollila, C. Smith, T. Ala-Nissila, and C. Denniston, Multiscale Model. Simul. 11, 213 (2013).
[36] D. Roehm and A. Arnold, Eur. Phys. J. ST 210, 73 (2012).
[37] H. J. Limbach, A. Arnold, B. A. Mann, and C. Holm, Comp. Phys. Comm. 174, 704 (2006).
[38] A. Arnold et al., ESPResSo 3.1 — Molecular Dynamics Software for Coarse-Grained Models, in Meshfree Methods for Partial Differential Equations VI, edited by M. Griebel and M. A. Schweitzer, volume 89 of Lecture Notes in Computational Science and Engineering, page 7, Springer, 2013.
[39] S. Frisch, B. Hasslacher, and Y. Pomeau, Phys. Rev. Lett. 56, 1505 (1986).
[40] A. Goldman, R. Cox, and H. Brenner, Chem. Eng. Sci. 22, 637 (1967).