Numerical simulation of moving rigid bodies in rarefied gases

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In this paper we present a numerical scheme to simulate a moving rigid body with arbitrary shape suspended in a rarefied gas. The rarefied gas is simulated by solving the Boltzmann equation using a DSMC particle method. The motion of the rigid body is governed by the Newton-Euler equations, where the force and the torque on the rigid body is computed from the momentum transfer of the gas molecules colliding with the body. On the other hand, the motion of the rigid body influences the gas flow in its surroundings. We validate the numerical results by testing the Einstein relation for Brownian motion of the suspended particle. The translational as well as the rotational degrees of freedom are taken into account. It is shown that the numerically computed translational and rotational diffusion coefficients converge to the theoretical values.

Key Words `rigid body motion, Boltzmann equation, DSMC, moving body in rarefied gas, Brownian diffusion` MSC 2010 65C05; 65C30; 74F10

1. Introduction

Nanoparticles play a vital role in many industrial processes and natural phenomena, including areas such as chemical engineering, biomedical technology, material science, physics, chemistry, and biology. Nanoparticles are often suspended in fluids during production, handling, and processing, but also after unintentional or unde-

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sired release to the environment. In many cases the suspending fluid is a gas, as in the case of large scale commercial production of nanoparticles, air pollution or clean room technology.

These applications have increased the interest in micro- and nanofluidics and have triggered the development of simulation methods and numerical schemes. For example, Direct Simulation Monte Carlo (DSMC) methods have been used in the modeling of small-scale devices with characteristic dimensions of microns down to nanometers [11]. Furthermore, DSMC is an ideal particle based scheme for the study of hydrodynamic fluctuations [2]. Moving boundary problems for kinetic equations have been recently investigated extensively in connection with MEMS, see [11] for an overview. To simulate such problems, DSMC as well as deterministic methods have been applied. In recent years several works have been reported for moving boundary problems in a rarefied gas, see [9], [20], [22], [7]. To simulate moving rigid bodies in a rarefied gas, not only the influence of the moving boundary on the gas has to be included in the simulation, but also the forces exerted by the gas accelerating the rigid body. See, for example [22,24] for one-dimensional situations with such a two-way coupling. We remark that, on the one hand, using DSMC based approaches for the above time-dependent problems with slow fluid flows requires some control over the large fluctuations inherent in these methods. On the other hand, the deterministic approaches are complicated to extend and computationally costly for higher dimensions. Finally, we note that DSMC methods are especially suited to couple moving rigid objects due to the Lagrangian nature of the gas molecules.

In this paper we present a simulation scheme for moving rigid bodies of arbitrary shape suspended in a rarefied gas suited for three dimensions. As an implementation of the scheme we consider a rigid spherical body contained inside a cube of micron size. The rarefied gas flow is simulated by solving the Boltzmann equation in a time dependent domain of computation using a DSMC particle method. The rigid body motion is given by the Newton-Euler equations, where the forces on the rigid body are calculated from the momentum transfer due to gas molecules impinging on the surface of the rigid body. The resulting motion of the rigid body affects in turn again the gas flow in the surroundings. This means that a two-way coupling has to be modeled. To validate our numerical scheme, we investigate 3D Brownian motion of a spherical rigid body suspended in a gas and compare the numerical results with the Einstein relation. Both the translational and the rotational degrees of freedom of the particle are taken into account. Furthermore, we perform a convergence study comparing the numerically computed translational and rotational diffusion coefficients to existing theoretical values for an increasing number of simulated gas molecules.

The paper is organized as follows. In section 2, we briefly present the Boltzmann equation and a numerical method for solving it. In section 3, we derive the force and the torque exerted on the rigid body due to the surrounding gas medium and also present a scheme based on the interaction of gas molecules and the rigid body

and its implementation to estimate the force and the torque on the rigid body in a DSMC framework. In section 4, we describe the translational and rotational Brownian motions of a rigid spherical particle suspended in a gas based on Langevin's equations and also present the derivation of the corresponding diffusion coefficients. The numerical results are presented in section 5, and some concluding remarks are given in section 6.

2. The Boltzmann equation

2.1. *The equation*

The Boltzmann equation describes the time evolution of a distribution function $f(t, \mathbf{x}, \mathbf{v})$ for particles with velocity $\mathbf{v} \in \mathbb{R}^d, d = 1, 2, 3$ and position $\mathbf{x} \in \mathbb{R}^3$ at time $t > 0$. It is given in nondimensional form as

$$\frac{\partial f}{\partial t} + \mathbf{v} \cdot \boldsymbol{\nabla}_x f = \frac{1}{\epsilon} J(f, f), \tag{2.1}$$

with the initial condition

$$f(t = 0, \mathbf{x}, \mathbf{v}) = f_0(\mathbf{x}, \mathbf{v}), \tag{2.2}$$

where ϵ is the Knudsen number, the ratio of mean free path and characteristic length, $J(f, f)$ is the collision operator which is given for hard-sphere molecules by

$$J(f, f) = \int_{\mathbb{R}^3} \int_{S^2} \beta(||\mathbf{v} - \mathbf{w}||, \mathbf{n})[f(\mathbf{v}')f(\mathbf{w}') - f(\mathbf{v})f(\mathbf{w})]d\omega(\mathbf{n})d\mathbf{w}, \tag{2.3}$$

where S^2 is the unit sphere in $\mathbb{R}^3$, $\mathbf{n} \in S^2$ is the unit vector in the impact direction, β is the collision cross section, $f(\mathbf{v}') = f(t, \mathbf{x}, \mathbf{v}')$, and analogously for $f(\mathbf{v})$ etc. The pair $(\mathbf{v}, \mathbf{w})$ and $(\mathbf{v}', \mathbf{w}')$ are the pre- and post- collisional velocities of two colliding gas molecules, given by

$$\mathbf{v}' = \mathbf{v} - \mathbf{n}[\mathbf{n} \cdot (\mathbf{v} - \mathbf{w})], \mathbf{w}' = \mathbf{w} + \mathbf{n}[\mathbf{n} \cdot (\mathbf{v} - \mathbf{w})]. \tag{2.4}$$

For more details we refer to [5]. We note that in this paper the characteristic length is the diameter of the rigid body. For the investigations in the present paper one has to solve (2.1) in a time dependent domain given by the moving nanoparticle and predefined outer boundaries. We consider diffuse reflection boundary conditions at the outer boundaries of the domain of computation and at the surface of the rigid body. It is worth noting that the diffuse reflection boundary condition at the rigid body has to be evaluated in the co-moving frame of reference.

2.2. *Numerical method for the Boltzmann equation*

We solve the Boltzmann equation using a variant of the DSMC method [4], developed in [17], [1]. The method is based on the time splitting of the Boltzmann equation. Introducing fractional steps one first solves the free transport equation (the collisionless Boltzmann equation) for one time step. During the free flow, boundary conditions

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are taken into account. In this paper we consider a closed cube containing a rigid spherical body. As already mentioned, diffuse reflection boundary conditions apply at all boundaries. In a second step (the collision step), the spatially homogenous Boltzmann equation without the transport term is solved. To solve the homogeneous Boltzmann equation, the key point is to find an efficient particle approximation of the product distribution functions in the Boltzmann collision operator given only an approximation of the distribution function itself. To simulate this equation by a particle method an explicit Euler step is performed. To guarantee positivity of the distribution function during the collision step, a restriction of the time step proportional to the Knudsen number is needed. That means that the method becomes exceedingly expensive for small Knudsen numbers. In such regimes, a special algorithm adapted to the small Knudsen number limit has to be used, see, e.g., [23]. Since here we aim at the large Knudsen number regime, we will not go into details concerning this issue.

The cube is discretized using a uniform grid size along all axes, resulting in a cubic grid. The initial phase space distribution of the gas is a Maxwellian distribution with the initial temperature, density and mean velocity as its parameters. The temperature is kept at the initial temperature throughout the simulations. The initial mean velocity and the initial velocity of the rigid body is zero.

The computational grid is divided into three sets. Gas cells completely filled by gas molecules, rigid body cells completely covered by the rigid body and boundary cells which are partially filled by gas molecules and partially by the rigid body. We note that due to the motion of the body we have to update the volume of the cells occupied by the gas at every time step. This update can be done efficiently by marking the boundary cells and its neighboring cells near the surface of the rigid body. Only the boundary cells and its neighbor cells are the candidate of the boundary cells in the next time step. Some computational efforts are necessary to update the volume of boundary cells occupied by the gas. This is obtained by a Monte Carlo method using particles. Let n_0 be the initial number of gas molecules per cell. We distribute randomly n_0 molecules in boundary cells and we set a counter how many molecules are lying outside the rigid body. We repeat this process for 500 times and then take the average of the counter. The gas volume fraction and the direction of the surface normal of the rigid body in the boundary cells are sufficient for the DSMC simulation near the boundary.

3. Force and torque on the rigid body

Generally, a rigid body suspended in a gas (representing, for example, a nanoparticle) moves under the influence of surface and volume forces. The surface forces are due to collisions of gas molecules with the body. In addition, different types of volume forces could be present, such as electrostatic and gravitational forces. In this work, we only consider the effect of the surface forces onto the rigid body. It should be noted, that via the solution of the Newton-Euler equations, inertial forces are

taken care of automatically.

3.1. *Force, torque and equation of motion*

To compute the surface force exerted on the rigid body by the surrounding gas we proceed as follows. We note that the following process can be applied to an arbitrarily shaped rigid body with obvious modifications. In this paper we consider a rigid spherical particle. Let $S(t) = \{\mathbf{y}(t) : ||\mathbf{y}(t) - \mathbf{X}(t)|| \leq R_{\mathrm{P}}\}$ be the rigid spherical particle with boundary $\partial S(t) = \{\mathbf{y}(t) : ||\mathbf{y}(t) - \mathbf{X}(t)|| = R_{\mathrm{P}}\}$ and center of mass $\mathbf{X}(t)$ at any moment in time t. The force $\boldsymbol{\mathcal{F}}$ and the torque $\boldsymbol{\mathcal{T}}$ exerted on the rigid sphere from the surrounding fluid is given by

$$\boldsymbol{\mathcal{F}} = - \int_{\partial S} \sigma \cdot \mathbf{n}_s dA \tag{3.1}$$

$$\boldsymbol{\mathcal{T}} = - \int_{\partial S} (\mathbf{y} - \mathbf{X}) \times (\sigma \cdot \mathbf{n}_s) dA, \tag{3.2}$$

where σ is the total stress tensor in the fluid and $\mathbf{n}_s$ is the outward normal to the boundary ∂S of the body. Using equations (3.1) and (3.2), the translational and rotational motion of the rigid body is described by the Newton-Euler equations

$$M\frac{d\mathbf{V}}{dt} = \boldsymbol{\mathcal{F}}, \tag{3.3}$$

$$I\frac{d\boldsymbol{\omega}}{dt} = \boldsymbol{\mathcal{T}}, \tag{3.4}$$

where M and I are the mass and moment of inertia of the spherical particle, and $\mathbf{V}$ and $\boldsymbol{\omega}$ are the translational and rotational velocities of the spherical particle, respectively. Equations (3.3) and (3.4) can be solved for $\mathbf{V}$ and $\boldsymbol{\omega}$. The total velocity $\mathbf{U}$ that combines both the translational and rotational motion of the rigid body is given by $\mathbf{U} = \mathbf{V} + (\mathbf{y} - \mathbf{X}) \times \boldsymbol{\omega}, \quad \mathbf{y} \in S$. Correspondingly, the equation of motion for points on the surface is

$$\frac{d\mathbf{y}}{dt} = \mathbf{U}, \quad \mathbf{y} \in S. \tag{3.5}$$

3.2. *Numerical approximation of the force and the torque within the DSMC scheme*

To compute the force $\boldsymbol{\mathcal{F}}$ and the torque $\boldsymbol{\mathcal{T}}$ on the spherical particle, at first we have to compute the stress tensor σ in the fluid domain and finally insert the value of σ in the equations (3.1) and (3.2). For a dilute gas, the flow is modeled by kinetic theory and given by the Boltzmann equation (2.1). The stress tensor σ can be computed as a moment of the phase-space distribution function. However, for moving boundary problems the correct numerical approximation of the stress tensor in the DSMC cells which are partially covered by the rigid body is not very accurate, because of the small number of simulated molecules in that cell. Thus, we compute the force and the torque from the interaction of the fluid molecules and the rigid

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particle. This is similar to a microscopic approach where the force and the torque would be computed by the collision of the spherical particle with the simulated gas molecules. When the gas molecules collide with the spherical particle, they transfer momentum and energy. Therefore, the total force and the total torque exerted on the spherical particle are computed by accumulating the increments of the linear and angular momentum imparted by all the colliding molecules to the rigid body. This leads to the following procedure to approximate numerically the force and the torque exerted on the spherical particle.

Let us discretize the boundary ∂S of the spherical particle by a uniformly distributed pointset $\partial S_h = \{\mathbf{y}_i, i = 1, \ldots, N_1\}$. Let us consider a gas molecule hitting the particle surface at a point $\mathbf{y} \in \partial S$ with momentum $\mathbf{p}$, being reflected with momentum $\mathbf{p}'$. In the following we assume that gas molecule is reflected diffusively from the boundary of the moving spherical particle. Then we find the closest neighbor $\mathbf{y}_i$ of $\mathbf{y}$ in the pointset ∂S_h to store pre- and post- collision momenta of that molecule. During the time interval Δt, there could be a number of such gas molecules impinging at a position whose closest neighbor is $\mathbf{y}_i$. The total pre- and post- collision momenta at $\mathbf{y}_i$ are calculated by taking the sum of pre-collision momenta $\mathbf{p}$ and post-collision momenta $\mathbf{p}'$ of those molecules which impinge in the neighborhood of $\mathbf{y}_i$. Let $\mathbf{p}_i$ and $\mathbf{p}'_i$ be the total pre- and post-collision momenta at the point $\mathbf{y}_i$. Then, the force $\boldsymbol{\mathcal{F}}_i$ and the torque $\boldsymbol{\mathcal{T}}_i$ exerted on the spherical particle at $\mathbf{y}_i$ during the time interval Δt are given by

$$\boldsymbol{\mathcal{F}}_i = \frac{\mathbf{p}_i - \mathbf{p}'_i}{\Delta t}, \quad i = 1, \ldots, N_1 \tag{3.6}$$

$$\boldsymbol{\mathcal{T}}_i = (\mathbf{y}_i - \mathbf{X}) \times \boldsymbol{\mathcal{F}}_i, \quad i = 1, \ldots, N_1, \tag{3.7}$$

where $\mathbf{X}$ is the center of mass of the spherical particle. Hence, the total force $\boldsymbol{\mathcal{F}}$ and the total torque $\boldsymbol{\mathcal{T}}$ on the spherical particle are given by

$$\boldsymbol{\mathcal{F}} = \sum_i \boldsymbol{\mathcal{F}}_i = \sum_i \frac{\mathbf{p}_i - \mathbf{p}'_i}{\Delta t} \tag{3.8}$$

$$\boldsymbol{\mathcal{T}} = \sum_i \boldsymbol{\mathcal{T}}_i = \sum_i (\mathbf{y}_i - \mathbf{X}) \times \boldsymbol{\mathcal{F}}_i. \tag{3.9}$$

Each DSMC molecule represents a large number of physical gas molecules. Therefore, we need to determine the mass of each DSMC simulated molecule to be used in the momentum transfer calculation. We employ the ideal gas law

$$pV = n\Re T \quad \text{rewritten as} \tag{3.10}$$

$$p = \rho R T, \tag{3.11}$$

where n is the number of moles and V is the system volume. $\Re$ and R are the universal and specific gas constants, respectively. From (3.10) and (3.11), we can write

$$n = \frac{\rho V}{m_{\mathrm{g}} N_{\mathrm{A}}}, \tag{3.12}$$

where N_{A} is the Avogadro number. The total number of physical gas molecules in the system is

$$N = \frac{\rho V}{m_{\mathrm{g}}}, \tag{3.13}$$

where m_{g} is the mass of a physical gas molecule.

Define

$$\nu := \frac{N}{N_0}. \tag{3.14}$$

Here, N_0 is the total number of DSMC simulated molecules, hence $\nu \geq 1$ is the number of physical gas molecules representing a single DSMC simulated molecule. This is also known as the statistical weight of the simulated DSMC molecule. Thus, the mass of a DSMC molecule is given by

$$M = m_{\mathrm{g}}\nu. \tag{3.15}$$

Equations (3.13), (3.14) and (3.15) finally yield

$$M = \frac{\rho V}{N_0}. \tag{3.16}$$

Thus, the force and the torque (3.8) and (3.9) are rewritten as

$$\boldsymbol{\mathcal{F}} = \sum_i \frac{M(\mathbf{v}_i - \mathbf{v}_i')}{\Delta t}, \tag{3.17}$$

$$\boldsymbol{\mathcal{T}} = \sum_i (\mathbf{y}_i - \mathbf{X}) \times \frac{M(\mathbf{v}_i - \mathbf{v}_i')}{\Delta t}, \tag{3.18}$$

where $\mathbf{v}_i$ and $\mathbf{v}_i'$ are the pre- and post- collisional velocities. With these expressions we can determine the translational and rotational motion of the spherical particle, solving (3.3), (3.4) and (3.5). For the time integration we use an explicit Euler scheme with given initial velocities $\mathbf{V}(t = 0) = \mathbf{V}_0$, $\boldsymbol{\omega}(t = 0) = \boldsymbol{\omega}_0$ and initial configuration $S(0) = S_0$ of the spherical particle. We have considered the same time step Δt for both Newton-Euler equations as well as the DSMC method.

4. Brownian motion

The theory of translational Brownian motion is concerned with the calculation of the probability density for the position of a rigid particle in a fluid. It is usually based on Langevin's equation, which is Newton's second law with the assumption that the force acting on the rigid particle is the sum of a viscous retarding force proportional to the velocity of the rigid particle and a rapidly fluctuating force whose statistical properties are such that the the velocity distribution approaches a Maxwell-Boltzmann distribution [10,16]. Analogously, rotational Brownian motion is concerned with the calculation of the probability density of the orientation of a body in a fluid. The specification of the orientation of a body requires three

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coordinates, such as Euler's angles. Here, we consider the simple case where the rigid particle rotates about a fixed axis through its center of mass. The theory of rotational Brownian motion is again based on a Langevin equation.

4.1. *Translational Brownian motion of a rigid particle*

Consider a system composed of N monoatomic gas molecules occupying a volume V and having an absolute temperature T. Let us consider a rigid particle suspended in a rarefied gas. The particle undergoes a random motion due to the impacting gas molecules. Let $\mathbf{V} = \frac{d\mathbf{X}(t)}{dt}$ denote the velocity of the particle relative to the gas. The theory of Brownian motion states that the velocity of the particle follows the Maxwell-Boltzmann velocity distribution which is given by [26]

$$f_{\mathbf{V}} = (\frac{M}{2\pi k_{\mathrm{B}}T})^{3/2}\exp\Big(-\frac{M\,||\mathbf{V}||^2}{2k_{\mathrm{B}}T} \Big), \tag{4.1}$$

where M is the mass of the particle and k_{B} is the Boltzmann constant. The mean square velocity can be calculated by taking the second moment of the velocity distribution (4.1) and is given by

$$< ||\mathbf{V}||^2 > = \int_{\mathbb{R}^3} ||\mathbf{V}||^2 \, f_{\mathbf{V}} d\mathbf{V} = \frac{3k_{\mathrm{B}}T}{M}. \tag{4.2}$$

Hence, the mean translational kinetic energy of the particle is given by

$$\frac{1}{2}M < ||\mathbf{V}||^2 > = \frac{3}{2}k_{\mathrm{B}}T. \tag{4.3}$$

The random impact of the surrounding gas molecules generally causes two kinds of effects: firstly, they act as a random driving force on the Brownian particle to maintain its irregular motion, and, secondly, they give rise to a friction force.

A simple model for the Brownian motion of a particle with mass M and center of mass $\mathbf{X}$ is the phenomenological stochastic equation [12]

$$M\frac{d^2\mathbf{X}(t)}{dt^2} = -\gamma\frac{d\mathbf{X}(t)}{dt} + \varsigma\mathbf{F}(t), \tag{4.4}$$

denoted as Langevin equation. The frictional force exerted by the medium is represented by the first term on the right-hand side, where γ is the translational friction coefficient, assumed to be independent of the particle velocity. The second term, $\varsigma\mathbf{F}(t)$, is the random force due to collisions with the surrounding gas molecules. For the sake of simplicity and idealization, the random force is usually assumed to be a white noise process [12] with autocorrelation function

$$< \mathbf{F}(t_1)\mathbf{F}(t_2) > = \delta(t_1 - t_2)\mathbb{I}. \tag{4.5}$$

At long times, the explicit solution of equation (4.4) yields a variance for the velocity $\mathbf{V} = \frac{d\mathbf{X}(t)}{dt}$ given by

$$\frac{3\varsigma}{2\gamma M}.$$

Assuming that the distribution of velocites follows a Maxwell-Boltzmann distribution, we obtain (see Nelson [16])

$$\varsigma = 2\gamma k_{\mathrm{B}} T. \tag{4.6}$$

For long times, the corresponding variance of $\mathbf{X}(t)$ is given by $6 D_T t$, with the translational diffusion coefficient

$$D_T = \frac{k_{\mathrm{B}} T}{\gamma}, \tag{4.7}$$

resulting in

$$< \|\mathbf{X}(t) - \mathbf{X}_0\|^2 > = \frac{6 k_{\mathrm{B}} T}{\gamma} t, \tag{4.8}$$

where $\mathbf{X}_0 = \mathbf{X}(t = 0)$ is the initial position of the particle. This is the same variance as for a classical diffusion process with diffusion constant D_T. (4.8) is usually referred to as the Einstein equation. We note that relation (4.6) is an explicit manifestation of the fluctuation-dissipation theorem [12]. It sets a constraint to the random force whose power spectrum is determined by the level of friction.

In Stokes flow regime, assuming small Knudsen and Reynolds numbers, the friction coefficient of a spherical particle has the following form [21]

$$\gamma_{\mathrm{C}} = 6 \pi \mu R_{\mathrm{P}}, \tag{4.9}$$

where μ is the viscosity of the fluid and R_{P} is the radius of the spherical particle. This equation is valid under the assumption that the fluid satisfies a no-slip boundary condition, meaning that the relative velocity of the fluid at the solid surface is zero. This assumption holds in the continuum regime where the Knudsen number $Kn << 1$. Substituting the value of the friction coefficient γ_{C} from (4.9) to (4.7), we get

$$D_{\mathrm{T,C}} = \frac{k_{\mathrm{B}} T}{6 \pi \mu R_{\mathrm{P}}}. \tag{4.10}$$

This is known as the Stokes-Einstein equation for diffusion of spherical particles in a fluid.

In the case of a very large Knudsen number $Kn >> 1$, an expression for the friction coefficient was derived by Epstein[8] using kinetic theory

$$\gamma_{\mathrm{FM}} = \frac{8}{3} R_{\mathrm{P}}^2 \rho \sqrt{\frac{2 \pi k_{\mathrm{B}} T}{m_{\mathrm{g}}}} \left(1 + \frac{\pi \alpha}{8}\right), \tag{4.11}$$

where ρ is the density of the gas. The coefficient α represents the fraction of gas molecules that are reflected diffusively, $(1 - \alpha)$ is the fraction of molecules with specular reflection.

The translational diffusion coefficient of a spherical particle in a rarefied gas is obtained by combining equations (4.7) and (4.11) and is given by

$$D_{\mathrm{T,FM}} = \frac{3}{8} \sqrt{\frac{m_{\mathrm{g}} k_{\mathrm{B}} T}{2 \pi}} \frac{1}{(1 + \frac{\alpha \pi}{8}) R_{\mathrm{P}}^2 \rho}. \tag{4.12}$$

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4.2. *Rotational Brownian motion and Brownian diffusion*

There also exists rotational diffusion, the change of the orientation of a colloidal particle due to the random torque exerted on it by the surrounding molecules. Rotational diffusion is important, for example, for the study of dielectric relaxation, fluorescence depolarization, or the line width in nuclear magnetic resonance measurements [15]. The dynamics of rotational diffusion of aspherical particle rotating about a fixed axis can be derived from the rotational analog of Langevin's equation based on the Euler equation [10]. This is given by

$$I\frac{d^2\Theta}{dt} = -\Upsilon\omega_3 + \Gamma,\tag{4.13}$$

where Θ is the angular displacement about a fixed axis parallel to the z-axis through the center of mass of the particle, $\omega_3 = \frac{d\Theta}{dt}$ is the third component of the angular velocity $\boldsymbol{\omega} = (\omega_1, \omega_2, \omega_3)$, and $I = \frac{2}{5}MR^2$ is the moment of inertia of the spherical particle. When the body rotates only about a fixed axis, the first two components ω_1, ω_2 of angular velocity $\boldsymbol{\omega}$ are zero. The first term on the right hand side of (4.13) is the friction torque with rotational friction coefficient Υ. The second term is the stochastic torque. This random torque has a correlation function given by [10]

$$< \Gamma(t_1)\Gamma(t_2) > = 2\pi k_{\mathrm{B}}T\Upsilon\delta(t_1 - t_2).\tag{4.14}$$

The amplitude $2\pi k_{\mathrm{B}}T\Upsilon$ of the stochastic torque is given by similar considerations as before: By virtue of equipartition of energy, the distribution of the angular velocity of the spherical particle has to be a Maxwell-Boltzmann distribution given by [10], [26]

$$f\boldsymbol{\omega} = \left(\frac{I}{2\pi k_{\mathrm{B}}T}\right)^{3/2}\exp\left(\frac{-I\,||\boldsymbol{\omega}||^2}{2k_{\mathrm{B}}T}\right).\tag{4.15}$$

Therefore, the mean square angular velocity of the particle is given by

$$< ||\boldsymbol{\omega}||^2 > = \int_{\mathbb{R}} ||\boldsymbol{\omega}||^2 f\boldsymbol{\omega}\,d\boldsymbol{\omega} = \frac{k_{\mathrm{B}}T}{I}.\tag{4.16}$$

Hence, the rotational kinetic energy of the particle is

$$\frac{1}{2}I < ||\boldsymbol{\omega}||^2 > = \frac{3}{2}k_{\mathrm{B}}T.\tag{4.17}$$

Since the components $\omega_1, \omega_2, \omega_3$ of the rotational velocity of the Brownian particle are mutually independent, the rotational kinetic energy in each component is

$$\frac{1}{2}I < \omega_i^2 > = \frac{1}{2}k_{\mathrm{B}}T,\ i = 1, 2, 3.\tag{4.18}$$

The theory of rotational diffusion can be derived in analogy to the theory of translational diffusion. For large times the solution of equation (4.13) can be approximated by the solution of a diffusion equation with rotational diffusion coefficient

$$D_{\mathrm{R}} = \frac{k_{\mathrm{B}}T}{\Upsilon}.\tag{4.19}$$

Thus, if at time $t = 0$ the orientation of the particle is $\Theta = \Theta_0$, the mean square angular displacement of the particle is given by

$$< |\Theta - \Theta_0|^2 > = 2D_{\mathrm{R}}t. \tag{4.20}$$

The rotational friction coefficient is the proportionality constant between the drag torque on the particle and its angular velocity about a fixed axis. In the continuum regime ($Kn << 1$), the rotational friction coefficient of a spherical particle is given by [13]

$$\Upsilon_{\mathrm{C}} = 8\pi\mu R_{\mathrm{P}}^3. \tag{4.21}$$

It depends on the viscosity μ of the fluid and the radius of the particle R_{P}.
In free molecular regime ($Kn >> 1$), the expression for the rotational friction coefficient is [13,8]

$$\Upsilon_{\mathrm{FM}} = \frac{2\pi}{3}\sqrt{\frac{8k_{\mathrm{B}}T}{\pi m_{\mathrm{g}}}}\rho R_{\mathrm{P}}^4. \tag{4.22}$$

It depends, among others, on the temperature T, and density ρ of the gas. Correspondingly, in the free molecular regime, the rotational diffusion coefficient is given by

$$D_{\mathrm{R,FM}} = \frac{3}{4\pi}\sqrt{\frac{\pi m_{\mathrm{g}}k_{\mathrm{B}}T}{2}}\rho R_{\mathrm{P}}^4. \tag{4.23}$$

5. Numerical results

In this section, we present results for the above described numerical algorithm. The numerical method is validated by comparing the numerical approximation of the translational and rotational diffusion coefficients with the explicit formulas given in the previous section. The cube-shaped computational domain is of size $10^{-6} \times 10^{-6} \times 10^{-6}$ m^3, the gas is kept at a uniform temperature of $T = 300$ K. A uniform Cartesian grid of $30 \times 30 \times 30$ cells is defined inside the computational domain. We have considered different numbers n_0 of gas molecules (such that $\nu \geq 1$) to compute the numerical value of the diffusion coefficients and compare the results with the theoretical values. The radius of the spherical particle is taken to be 10^{-7} m. The gas is argon, a monoatomic gas with mass $m_{\mathrm{g}} = 6.63 \times 10^{-26}$ kg, the Boltzmann constant is $k_{\mathrm{B}} = 1.38 \times 10^{-23}$ J/K, and the specific gas constant is $R = k_{\mathrm{B}}/m_{\mathrm{g}} = 208$ J/(kgK) [24]. We use a hard-sphere collision model with diameter $d = 3.68 \times 10^{-10}$ m. The numerical computation is performed for a Knudsen number $Kn = 11$, using the the particle diameter as length scale. The spherical particle is initially kept at rest in the center of the computational domain, with its center of mass at $(5 \times 10^{-7}\mathrm{m}, 5 \times 10^{-7}\mathrm{m}, 5 \times 10^{-7}\mathrm{m})$. Hard collisions are performed between gas molecules and the spherical particle. In our simulations, diffuse boundary conditions are applied at the boundary of the particle as well as at the walls of the domain. The force and the torque are computed using (3.17) and (3.18). The
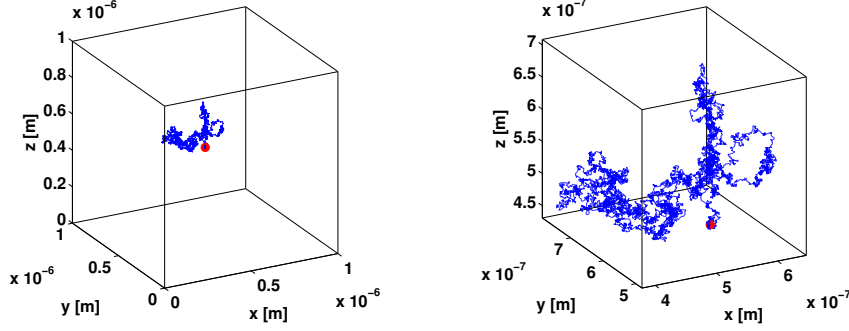
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Fig. 1: Brownian motion of the spherical particle until a time of 3.7879×10^{-6} seconds.Left: normal view, right: zoom view.

trajectory of the spherical particle is computed using (3.5). The trajectory of the center of mass of the particle up to a time of 3.7879×10^{-6} seconds is shown in Fig. 1.

To compute the numerical translational diffusion coefficient of the particle, we have performed the experiment a number of times under similar physical conditions. The displacement of the center of particle is sampled at $t = 2.617 \times 10^{-8}$ seconds. The corresponding distribution of endpoints of the particle trajectories is shown in figure 2 (left). The corresponding distribution of the x-component of the displacement of the center of mass is displayed in Fig. 2 (right). The other components show a similar behaviour. The center of mass of the particle follows a Gaussian distribution [25] with zero mean displacement and a variance in each direction approximately equal to $2D_\text{T}t$.

We have performed numerical experiments for an increasing number of simulated gas molecules and sampled the data to obtain the center of mass of the spherical particles at time $t = 2.617 \times 10^{-8}$ seconds and the translational diffusion coefficient, computed by using (4.8). The theoretical value of the translational diffusion coefficient is given by (4.12). Fig. 3 shows that the numerical approximation of the translational diffusion coefficient of the particle converges to the theoretical value for an increasing number of simulated molecules.

The rotational diffusion coefficient of the spherical particle in equation (4.23) is derived for a non-moving particle which is rotating only about a fixed axis through its center of mass. Thus, in our numerical simulation, we compute the value of the rotational diffusion coefficient of the spherical particle by fixing its center of mass and letting the spherical particle rotate only around a single axis parallel to the $z-$ axis by putting first and second components of the angular velocity equal to zero. The torque is computed by using (3.18). As a result, one obtains the angular velocity $\boldsymbol{\omega} = (0, 0, \omega_3)$. Finally, the angular displacement Θ is computed using the
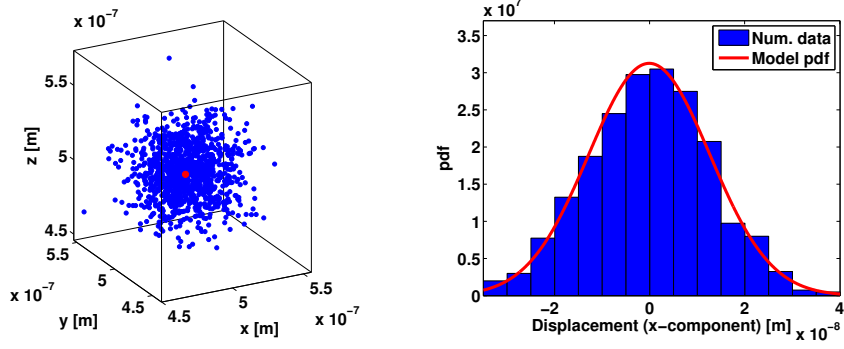
Fig. 2: Endpoints of the Brownian trajectories of the spherical particles (left). The histograms represent the simulation data and the solid line is the Gaussian distribution of the x-displacements (right), both at time 2.2617×10^{-8} seconds.
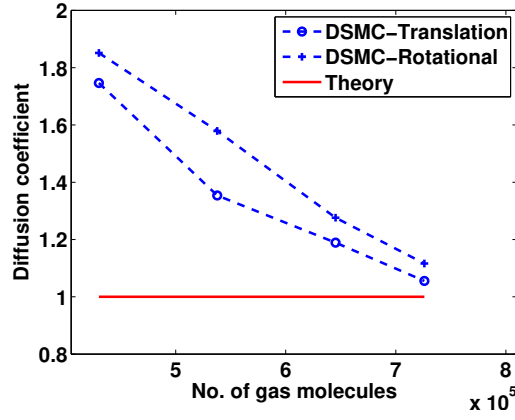

Fig. 3: Convergence of the normalized translational and rotational diffusion coefficients with increasing number of gas molecules. The solid line is the theoretical value, lines with - - o - - and - - + - - represent the translational and the rotational diffusion coefficients, respectively.

equation of angular motion

$$\frac{d\Theta}{dt} = \omega_3, \quad \text{with initial angle} \quad \Theta_0 = 0. \tag{5.1}$$

The angular displacement of the spherical particle is sampled at time $t = 2.2617 \times 10^{-8}$ seconds. The corresponding distribution function is shown in Fig. 4. It is well described by a Gaussian distribution with zero mean angular displacement and a variance of $2D_{\mathrm{R}}t$.

We have also computed the rotational diffusion coefficient of the spherical parti-
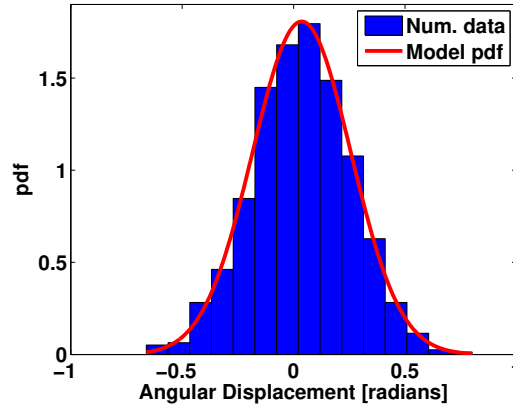
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Fig. 4: The histograms represents the simulation data and the solid line is the Gaussian distribution function of the angular displacement about the z-axis at time 2.2617×10^{-8} seconds.

cle for different numbers of simulated molecules by using (4.20). The approximated values are compared with the theoretical value of the rotational diffusion coefficient (4.23). The comparison of the numerically computed values of the rotational diffusion coefficient with increasing number of simulated gas molecules and the theoretical value is again shown in Figure 3.

For the Brownian motion of the colloidal particle the translation and rotational velocities must follow the Maxwellian distributions (4.1) and (4.15). To test this, the numerical experiment has been run for a long time at a given temperature of $T = 300$ K, and the translational and rotational velocities have been sampled at each time step. Figs. (5) show the distribution of the translational velocity in x-direction and the distribution of the rotational velocity. The histograms represent the simulated data, and the solid lines are the model Gaussian curves. It can be concluded that the numerically computed probability density functions agree with the velocity distributions (4.1) and (4.15).

6. Conclusion and outlook

We have presented a numerical method for simulating rigid bodies with arbitrary shape in a rarefied gas. The rarefied gas is modeled by the Boltzmann equation, and the motion of the particle is described by the Newton-Euler equations. The Boltzmann equation is solved by a DSMC type of particle method with a hard-sphere collision term. The forces acting on the particle are computed from the momentum transfer due to the molecules colliding with the surface. Translational and rotational diffusions of a spherical particle was investigated numerically. The obtained probability density functions for the particle's center-of-mass position, the
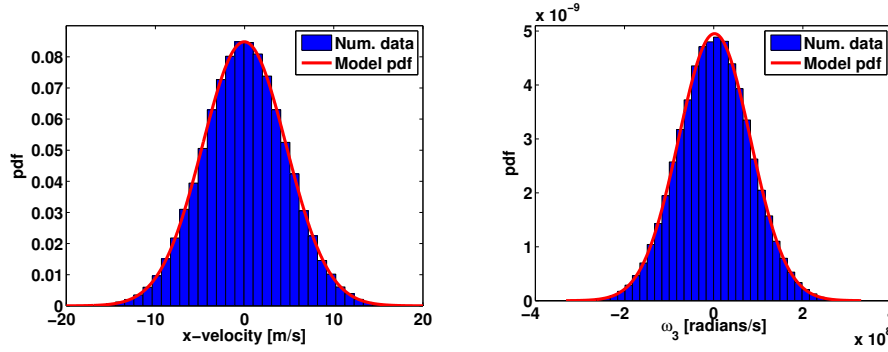
Fig. 5: x-component of the translational velocity (left), rotational velocity ω_3 of the spherical particle (right). In both figures the histograms represent the simulation data and the solid line is the Gaussian distribution function at T = 300 K.

translational and rotational velocities, and the corresponding diffusion coefficients were compared with results from the theory of Brownian motion. The convergence of the diffusion coefficients to their theoretical values with increasing number of simulated molecules was demonstrated. Future work will concentrate on computing thermophoretic transport processes of (non) spherical particles at different Knudsen numbers using the approach presented here.

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References

1. H. Babovsky, R. Illner, A Convergence Proof for Nanbu's Simulation Method for the Boltzmann Equation, *SIAM J. Numer. Anal.*, 26 (1989) 45-64.
2. K. Balakrishnan, J. B. Bell, A. Donev and A. L. Garcia Fluctuating Hydrodynamics and Direct Simulation Monte Carlo, 28th International Symposium on Rarefied Gas Dynamics, *AIP Conf. Proc.*, 1501(2012), 695-704.
3. G. K. Batchelor, An Introduction to Fluid Dynamics, Cambridge University Press, 1967.
4. G. A. Bird, Molecular Gas Dynamics and Direct Simulation of Gas Flows, Clarendon, Oxford, 1994.
5. C. Cercigani, The Boltzmann Equation and its Applications, Springer, Berlin, 1988.
6. C. Cercignani, M. Lampis, Kinetic Model of Gas-surface Interactions, *Transp. Th. and Stat. Phys.*, 1 (2) (1971) 101-114.
7. G. Dechriste', L. Mieussens, Numerical simulation of micro flows with moving obstacles, *J. Phys.: Conf. Series* 362 (2012) 012030.
8. P. S. Epstein, On the Resistance Experienced by Spheres in Their Motion Through Gases, *Phys. Rev.* 23 (1924) 710.

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9. M. A. Gallis, D. J. Rader, and J. R. Torczynski, Thermophoresis in Rarefied Gas Flows, *Aerosol Science and Technology*, 36 (2002) 1099-1117.
10. P. S. Hubbard, Rotational Brownian Motion, *Phys. Rev. A*, 6 (1972) 2421.
11. G. Karniadakis, A. Beskok, N. Aluru, Microflows and Nanoflows: Fundamentals and Simulations. Springer., New York, 2005.
12. R. Kubo, The Fluctuation-dissipation Theorem, *Rep. Prog. Phys.*, 29 (1966) 255.
13. S. K. Loyalka, Motion of a Sphere in a Gas: Numerical Solution of the Linearized Boltzmann Equation, *Phys. Fluid A*, 4 (1992) 1049.
14. L. Mädler, S.K. Friedlander, Transport of Nanoparticles in Gases: Overview and Recent Advances, *Aerosol and Air Quality Research*, 7(3) (2007) 304-342.
15. R. M. Mazo, Brownian Motion: Fluctuations, Dynamics, and Applications, Oxford University Press, 2002.
16. E. Nelson Dynamical theories of Brownuan motion, Princeton University Press, 2nd Ed. , 2001.
17. H. Neunzert, J. Struckmeier, Particle Methods for the Boltzmann Rquation, *Acta Numerica*, 4 (1995), 417-457.
18. A.P. Philipse, Notes on Brownian Motion, Utrecht University, Debye Institute, Van 't Hoff Laboratory, August 2011.
19. W.B. Russel, Small Particles Suspended in Liquid, *Ann. rev. Fluid Mech.*, 13 (1981) 425-55.
20. G. Russo, F. Filbet, Semi-Lagrangian schemes applied to moving boundary problems for the BGK model of rarefied gas dynamics, *Kinetic and Related Models* 2 (2009), 231-252.
21. G. G. Stokes, On the Effect of Fluids on the Motion of Pendulums, *Trans. Cambridge Philos. Soc.*, 9, 8 (1851). Reprinted in Mathematical and Physical Papers III (Cambridge University Press, Cambridge, MA).
22. T. Tsuji, K. Aoki, Moving boundary problems for a rarefied gas: Spatially one dimensional case, *J. Comp. Phys.* 250 (2013), 574-600.
23. S. Tiwari, A. Klar, S. Hardt, A particle-particle hybrid method for kinetic and continuum equations, *J. Comput. Phys.* 228 (2009) 7109-7124.
24. S. Tiwari, A. Klar, S. Hardt, A. Donkov, Simulation of a Moving Liquid Droplet Inside a Rarefied Gas Region, *Computers & Fluids.* 71 (2013) 283-196.
25. G. E. Uhlenbeck, L. S. Ornstein, On the Theory of Brownian Motion, *Physical Rev.*, 36 (1930).
26. N. Wax, Selected Papers on Noise and Stochastic Processes, edited by N. Wax, Dover, New York , 1954.