

Statistical Error in Particle Simulations of Fluid Flow and Heat Transfer

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Abstract— We present predictions for the statistical error due to finite sampling in the presence of thermal fluctuations in molecular simulation algorithms. Specifically, we present predictions for the error dependence on hydrodynamic parameters and the number of samples taken. Expressions for the common hydrodynamic variables of interest such as flow velocity, temperature, density, pressure, shear stress and heat flux are derived using equilibrium statistical mechanics. Both volume-averaged and surface-averaged quantities are considered. Comparisons between theory and computations using direct simulation Monte Carlo for dilute gases, and molecular dynamics for dense fluids, show that the use of equilibrium theory provides accurate results.

I. INTRODUCTION

Recent developments in micro and nano-technology have sparked significant interest in molecular simulation methods such as Molecular Dynamics (MD) [1], [2] and the direct simulation Monte Carlo (DSMC) [3], [4]. Molecular Dynamics is generally used to simulate liquids while DSMC is a very efficient algorithm for simulating dilute gases. In molecular simulation methods the connection to macroscopic observable fields, such as velocity and temperature, is achieved through averaging appropriate microscopic properties. The simulation results are therefore inherently statistical and statistical errors due to finite sampling need to be fully quantified.

Though confidence intervals may be estimated by measuring the variance of these sampled quantities, this additional computation can be burdensome and thus is often omitted. Furthermore, it would be useful to estimate confidence intervals *a priori* so that one could predict the computational effort required to achieve a desired level of accuracy. For example, it is well known that obtaining accurate hydrodynamic fields (e.g., velocity profiles) is computationally expensive in low Mach number flows so it is useful to have an estimate of the computational effort required to reach the desired level of accuracy.

In this paper we present expressions for the magnitude of statistical errors due to thermal fluctuations in molecular simulations for the typical observables of interest, such as velocity, density, temperature, and pressure. We also present expressions for the shear stress and heat flux in the dilute gas limit. Both volume averaging and flux averaging is considered. Although we make use of expressions from equilibrium statistical mechanics, the non-equilibrium

modifications to these results are very small, even under extreme conditions [5]. This is verified by the good agreement between our theoretical expressions and the corresponding measurements in our simulations.

We now give a brief outline of our paper. In section II the theoretical expressions for the statistical error due to thermodynamic fluctuations are presented. These expressions are verified by molecular simulations, as described in section III. Concluding remarks appear in section IV.

II. STATISTICAL ERROR DUE TO THERMAL FLUCTUATIONS

A. Volume-averaged quantities

We first consider the fluid velocity. In a particle simulation, the flow field is obtained by measuring the instantaneous center of mass velocity, \mathbf{u} , for particles in a statistical cell volume. The statistical mean value of the local fluid velocity, $\langle \mathbf{u} \rangle_s$, is estimated over M independent samples. For steady flows, these may be sequential samples taken in time; for transient flows these may be samples from an ensemble of realizations. The average fluid velocity, $\langle \mathbf{u} \rangle$, is defined such that $\langle \mathbf{u} \rangle_s \rightarrow \langle \mathbf{u} \rangle$ as $M \rightarrow \infty$; for notational convenience we also write $\langle \mathbf{u} \rangle = \mathbf{u}_0$. Let us define $\delta u_x \equiv u_x - u_{x0}$ to be the instantaneous fluctuation in the x -component of the fluid velocity; note that all three components are equivalent. From equilibrium statistical mechanics [6],

$$\langle \delta u_x^2 \rangle = \frac{kT_0}{mN_0} = \frac{a^2}{\gamma \text{Ac}^2 N_0} \quad (1)$$

where N_0 is the average number of particles in the statistical cell, T_0 is the average temperature, m is the particle mass, k is Boltzmann's constant, a is the sound speed, and $\gamma = c_P/c_V$ is the ratio of the specific heats. The acoustic number $\text{Ac} = a/a^i$ is the ratio of the fluid's sound speed to the sound speed of a "reference" ideal gas at the same temperature

$$a^i = \sqrt{\gamma kT/m} \quad (2)$$

Note that this reference ideal gas has a ratio of specific heats (γ^i) equal to the *original fluid* specific heat ratio, that is $\gamma^i = \gamma$ as shown in equation (2).

An alternative construction of (1) is obtained from the equipartition theorem [6]

$$\begin{aligned}\frac{3}{2}kT_0 &= \frac{1}{2}m\langle|\mathbf{c} - \mathbf{c}_0|^2\rangle \\ &= \frac{1}{2}m\langle((c_x - c_{x0})^2 + (c_y - c_{y0})^2 + (c_z - c_{z0})^2)\rangle \\ &= \frac{3}{2}m\langle(c_x - c_{x0})^2\rangle\end{aligned}$$

where \mathbf{c} is the translational molecular velocity. For a non-equilibrium system, this expression defines T_0 as the average translational temperature. Note that $\mathbf{u}_0 = \mathbf{c}_0$ and

$$\langle|\delta\mathbf{u}|^2\rangle = \frac{\langle|\mathbf{c} - \mathbf{c}_0|^2\rangle}{N_0} = \frac{3kT_0}{mN_0}. \quad (3)$$

The above expression also reminds us that the expected error in estimating the magnitude of the fluid velocity is $\sqrt{3}$ larger than in estimating a velocity component.

We may define a ‘‘signal-to-noise’’ ratio as the average fluid velocity over its standard deviation; from the above,

$$\frac{|u_{x0}|}{\sqrt{\langle\delta u_x^2\rangle}} = \text{Ac Ma}\sqrt{\gamma N_0} \quad (4)$$

where $\text{Ma} = |u_{x0}|/a$ is the local Mach number based on the velocity component of interest. This result shows that for fixed Mach number, in a dilute gas simulation ($\text{Ac} = 1$), the statistical error due to thermal fluctuations cannot be ameliorated by reducing the temperature. However, when the Mach number is small enough for compressibility effects to be negligible, favorable relative statistical errors may be obtained by performing simulations at an increased Mach number (to a level where compressibility effects are still negligible).

The one-standard-deviation error bar for the sample estimate $\langle u_x \rangle_s$ is $\sigma_u = \sqrt{\langle\delta u_x^2\rangle}/\sqrt{M}$ and the fractional error in the estimate of the fluid velocity is

$$E_u = \frac{\sigma_u}{|u_{x0}|} = \frac{1}{\sqrt{MN_0}} \frac{1}{\text{Ac Ma}\sqrt{\gamma}}, \quad (5)$$

yielding

$$M = \frac{1}{\gamma \text{Ac}^2 N_0 \text{Ma}^2 E_u^2}. \quad (6)$$

Consider the following example; with $N_0 = 100$ particles in a statistical cell, if a one percent fractional error is desired in a $\text{Ma} = 1$ flow, about $M = 100$ independent statistical samples are required (assuming $\text{Ac} \approx 1$). However, for a $\text{Ma} = 10^{-2}$ flow, about 10^6 independent samples are needed, which quantifies the empirical observation that the resolution of the flow velocity is computationally expensive for low Mach number flows.

Next we turn our attention to the density. From equilibrium statistical mechanics, the fluctuation in the number of particles in a cell is

$$\langle\delta N^2\rangle = -N^2 \frac{kT_0}{V^2} \left(\frac{\partial V}{\partial P}\right)_T = \kappa_T N_0^2 \frac{kT_0}{V} \quad (7)$$

where V is the volume of the statistics cell and $\kappa_T \equiv -V^{-1}(\partial V/\partial P)_T$ is the isothermal compressibility. Note

that for a dilute gas $\kappa_T = 1/P$ so $\langle\delta N^2\rangle = N$ and, in fact, N is Poisson random variable. The fractional error in the estimate of the density is

$$E_\rho = \frac{\sigma_\rho}{\rho_0} = \frac{\sigma_N}{N_0} = \frac{\sqrt{\langle\delta N^2\rangle}}{N_0\sqrt{M}} = \frac{\sqrt{\kappa_T kT_0}}{\sqrt{MV}} = \frac{\sqrt{\kappa_T/\kappa_T^i}}{\sqrt{MN_0}} \quad (8)$$

where $\kappa_T^i = V/N_0 kT_0$ is the isothermal compressibility of the reference dilute gas ($\gamma^i = \gamma$) at the same density and temperature. Since $a \propto 1/\sqrt{\kappa_T}$,

$$E_\rho = \frac{1}{\sqrt{MN_0}} \frac{1}{\text{Ac}} \quad (9)$$

Note that for fixed M and N_0 , the error decreases as the compressibility decreases (i.e., as the sound speed increases) since the density fluctuations are smaller.

Let us now consider the measurement of temperature. First we should remark that the measurement of instantaneous temperature is subtle, even in a dilute gas. But given that temperature is measured correctly, equilibrium statistical mechanics gives the variance in the temperature fluctuations to be

$$\langle\delta T^2\rangle = \frac{kT_0^2}{c_V N_0} \quad (10)$$

where c_V is the heat capacity per particle at constant volume. The fractional error in the estimate of the temperature is

$$E_T = \frac{\sigma_T}{T_0} = \frac{\sqrt{\langle\delta T^2\rangle}}{T_0\sqrt{M}} = \frac{1}{\sqrt{MN_0}} \sqrt{\frac{k}{c_V}} \quad (11)$$

Because the fluctuations are smaller, the error in the temperature is smaller when the heat capacity is large. Note that the temperature associated with various degrees of freedom (translational, vibrational, rotational) may be separately defined and measured. For example, if we consider only the measurement of the translational temperature, then the appropriate heat capacity is that of an ideal gas with three degrees of freedom, i.e. $c_V = \frac{3}{2}k$, corresponding to the three translational components.

Finally, the variance in the pressure fluctuations is

$$\langle\delta P^2\rangle = -kT_0 \left(\frac{\partial P}{\partial V}\right)_S = \frac{\gamma kT_0}{V\kappa_T} \quad (12)$$

so the fractional error in the estimate of the pressure is

$$E_P = \frac{\sigma_P}{P_0} = \frac{\sqrt{\langle\delta P^2\rangle}}{P_0\sqrt{M}} = \frac{P_0^i}{P_0} \frac{\text{Ac}\sqrt{\gamma}}{\sqrt{MN_0}} \quad (13)$$

where $P_0^i = N_0 kT_0/V$ is the pressure of an ideal gas under the same conditions. Note that the error in the pressure is proportional to the acoustic number while the error in the density, eqn. (9), goes as Ac^{-1} .

B. Shear stress and heat flux for dilute gases

The thermodynamic results in the previous section are general; in this section we consider transport quantities

and restrict our analysis to dilute gases. In a dilute gas, the shear stress and heat flux are defined as

$$\tau_{xy0} = \langle \tau_{xy} \rangle = \langle \rho c_x c_y \rangle \quad (14)$$

and

$$q_{x0} = \langle q_x \rangle = \left\langle \frac{1}{2} \rho c_x c^2 \right\rangle \quad (15)$$

respectively. Under the assumption of a dilute gas, the fluctuation in the (equilibrium) shear stress and heat flux in a volume V containing N_0 particles can be calculated using the Maxwell-Boltzmann distribution. Note that in equilibrium, the expected values of the shear stress and the heat flux are zero.

Using the definitions of shear stress and heat flux in terms of moments of the velocity distribution, direct calculation of the variance of the x - y component of the stress tensor based on a single-particle distribution function gives

$$\langle \tau_{xy}^2 \rangle = \langle (\rho c_x c_y)^2 \rangle \quad (16)$$

$$= \rho_0^2 \langle (c_x c_y)^2 \rangle \quad (17)$$

$$= \frac{1}{4} \rho_0^2 \left(\frac{2kT_0}{m} \right)^2 = P_0^2 \quad (18)$$

In obtaining the second equation we assumed $\langle c_x \rangle = \langle c_y \rangle = \langle (\rho - \rho_0) c_x c_y \rangle = 0$. For the x component of the heat flux vector, we find

$$\langle q_x^2 \rangle = \left\langle \left(\frac{1}{2} \rho c_x c^2 \right)^2 \right\rangle \quad (19)$$

$$= \rho_0^2 \left\langle \left(\frac{1}{2} c_x c^2 \right)^2 \right\rangle \quad (20)$$

$$= \frac{35}{32} \rho_0^2 \left(\frac{2kT_0}{m} \right)^3 = \frac{35}{8} c_m^2 P_0^2 \quad (21)$$

where $c_m = \sqrt{2kT_0/m}$ the most probable particle speed and we have assumed $\langle c_x \rangle = \langle c_y \rangle = \langle (\rho - \rho_0) c^2 \rangle = 0$. Note that in equilibrium for a cell containing N_0 particles, the variance of the mean is given by $\langle \delta \tau_{xy}^2 \rangle = \langle \tau_{xy}^2 \rangle / N_0$ and $\langle \delta q_x^2 \rangle = \langle q_x^2 \rangle / N_0$ for the shear stress and heat flux respectively.

In order to derive expressions for the relative fluctuations we need expressions for the magnitude of the fluxes. We are only able to provide closed form expressions for the latter in the continuum regime where

$$\tau_{xy} = \mu \left(\frac{\partial u_x}{\partial y} + \frac{\partial u_y}{\partial x} \right) \quad (22)$$

and

$$q_x = -\kappa \frac{\partial T}{\partial x} \quad (23)$$

where μ is the coefficient of viscosity and κ is the thermal conductivity. Above Knudsen numbers of $\text{Kn} \approx 0.1$, it is known that these continuum expressions are only approximate and better results are obtained using more elaborate formulations from kinetic theory (e.g., Burnett's formulation). Here, the Knudsen number is defined as $\text{Kn} = \lambda/\ell$ and the mean free path as

$$\lambda = \frac{8}{5\sqrt{\pi}} \frac{c_m \mu}{P_0} \quad (24)$$

Note that this expression for the mean free path simplifies to the hard sphere result when the viscosity is taken to be that of hard spheres.

Using (22) and (23) we find that in *continuum flows*, the relative fluctuations in the shear stress and heat flux are given by

$$E_\tau = \frac{\sqrt{\langle \delta \tau_{xy}^2 \rangle}}{|\tau_{xy0}| \sqrt{M}} = \frac{16}{5\sqrt{2\pi}\gamma} \frac{1}{\text{Kn Ma}_*} \frac{1}{\sqrt{N_0 M}} \left| \frac{\partial u_x^*}{\partial y^*} + \frac{\partial u_y^*}{\partial x^*} \right|^{-1} \quad (25)$$

and

$$E_q = \frac{\sqrt{\langle \delta q_x^2 \rangle}}{|q_{x0}| \sqrt{M}} = \frac{8\sqrt{35}}{5\sqrt{2\pi}} \frac{\text{Pr}(\gamma-1)}{\gamma} \frac{T}{\Delta T} \frac{1}{\text{Kn}} \frac{1}{\sqrt{N_0 M}} \left| \frac{\partial T^*}{\partial x^*} \right|^{-1} \quad (26)$$

respectively. Here, stars denote non-dimensional quantities: $u^* = u_0/\tilde{u}$, $T^* = T/\Delta T$ and $x^* = x/\ell$, where \tilde{u} , ΔT , and ℓ are characteristic velocity, temperature variation and length. The Mach number Ma_* is defined with respect to the characteristic velocity \tilde{u} rather than the local velocity used to define Ma , and Pr is the gas Prandtl number.

If viscous heat generation is responsible for the temperature differences characterized by ΔT , then it is possible to express equation (26) in the following form

$$E_q = \frac{\sqrt{\langle \delta q_x^2 \rangle}}{|q_{x0}| \sqrt{M}} = \frac{8\sqrt{35}}{5\gamma\sqrt{2\pi}} \frac{\text{Br}}{\text{Kn Ma}_*^2} \frac{1}{\sqrt{N_0 M}} \left| \frac{\partial T^*}{\partial x^*} \right|^{-1} \quad (27)$$

The Brinkman number

$$\text{Br} = \frac{\mu \tilde{u}^2}{\kappa \Delta T}, \quad (28)$$

is the relevant non-dimensional group that compares temperature differences due to viscous heat generation to the characteristic temperature differences in the flow. (It follows that if viscous heat generation is responsible for the temperature changes, $\text{Br} \approx 1$.)

It is very instructive to extend the above analysis to equation (11). If we define the relative error in temperature with respect to the temperature changes rather than the absolute temperature, we obtain

$$E_{\Delta T} = \frac{\sigma_T}{\Delta T} = \frac{\sqrt{\langle \delta T^2 \rangle}}{\Delta T \sqrt{M}} = \frac{T_0}{\Delta T \sqrt{M N_0}} \sqrt{\frac{k}{c_V}} \quad (29)$$

$$= \frac{1}{\text{Pr}(\gamma-1)} \frac{\text{Br}}{\text{Ma}_*^2} \frac{1}{\sqrt{M N_0}} \sqrt{\frac{k}{c_V}} \quad (30)$$

where again, if viscous heat generation is the only source of heat, $\text{Br} \approx 1$. The above development shows that resolving the temperature *differences* or heat flux due to viscous heat generation is very computationally inefficient for low speed flow since for a given expected error $E_{\Delta T}$ we find that the number of samples scales as $M \propto \text{Ma}_*^{-4}$.

Comparison of equations (5) and (25) and equations (30) and (27) reveals that

$$E_\tau \sim \frac{E_u}{\text{Kn}} \quad (31)$$

and

$$E_q \sim \frac{E_{\Delta T}}{\text{Kn}} \quad (32)$$

since the non-dimensional gradients will be of order one. As the above equations were derived for the continuum regime ($\text{Kn} < 0.1$), it follows that the relative error in these moments is significantly higher. This will also be shown to be the case in the next section when the shear stress and heat flux are evaluated as fluxal (surface) quantities. This has important consequences in hybrid methods [7], [8], as coupling in terms of state (Dirichlet) conditions is subject to less variation than coupling in terms of flux conditions.

C. Fluxal quantities

In this section we give expressions for the relative errors in the components of the stress tensor and the heat flux vector, when calculated as fluxes across a reference surface. Our analysis is based on the assumption of an infinite, ideal gas in equilibrium. The following expressions

$$\sqrt{\langle \delta(\tau_{xy}^f)^2 \rangle} = \frac{\rho c_m^2}{2\sqrt{\pi}} \frac{1}{\sqrt{N^+}} \quad (33)$$

$$\sqrt{\langle \delta(q_x^f)^2 \rangle} = \sqrt{\frac{3}{\pi}} \frac{\rho c_m^3}{2} \frac{1}{\sqrt{N^+}} \quad (34)$$

were obtained in [9] and are quoted here without proof. Here the superscript f denotes fluxal measurement and N^+ denotes the number of particles crossing the surface from left to right. The above relations also hold in the case where a mean flow in directions parallel to the measuring surface exists.

Following the development in the previous section, we derive expressions for the relative expected error in the continuum regime in which models exist for the shear stress and heat flux. In this regime we find

$$\begin{aligned} E_{\tau}^f &= \frac{\sqrt{\langle \delta(\tau_{xy}^f)^2 \rangle}}{|\tau_{xy0}^f| \sqrt{M}} \\ &= \frac{16}{5\pi\sqrt{2}\gamma} \frac{1}{\text{Kn}} \frac{1}{\text{Ma}_*} \frac{1}{\sqrt{MN^+}} \left| \frac{\partial u^*}{\partial y^*} + \frac{\partial v^*}{\partial x^*} \right|^{-1} \end{aligned} \quad (35)$$

and

$$\begin{aligned} E_q^f &= \frac{\sqrt{\langle \delta(q_x^f)^2 \rangle}}{|q_{x0}^f| \sqrt{M}} \\ &= \frac{16\sqrt{3}}{5\pi} \frac{\text{Pr}(\gamma-1)}{\gamma} \frac{T}{\Delta T} \frac{1}{\text{Kn}} \frac{1}{\sqrt{MN^+}} \left| \frac{\partial T}{\partial x^*} \right|^{-1} \end{aligned} \quad (36)$$

$$= \frac{16\sqrt{3}}{5\gamma\pi} \frac{\text{Br}}{\text{Kn}} \frac{1}{\text{Ma}_*^2} \frac{1}{\sqrt{MN^+}} \left| \frac{\partial T}{\partial x^*} \right|^{-1} \quad (37)$$

Comparing (35) with the corresponding expressions for volume-averaged stress tensor, (25), one finds that, aside from the numerical coefficients, the expressions differ only in the number of particles used, either N^+ or N_0 ; one finds a similar result for the heat flux.

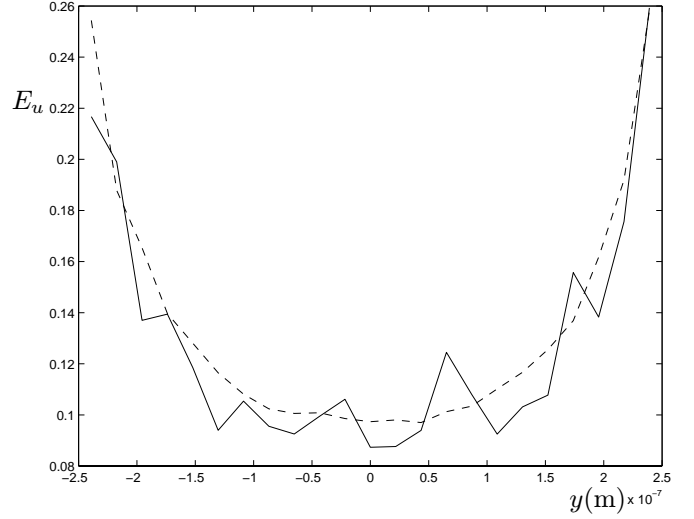


Fig. 1. Fractional error in velocity for Poiseuille flow in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (5) and the solid line denotes DSMC simulation results.

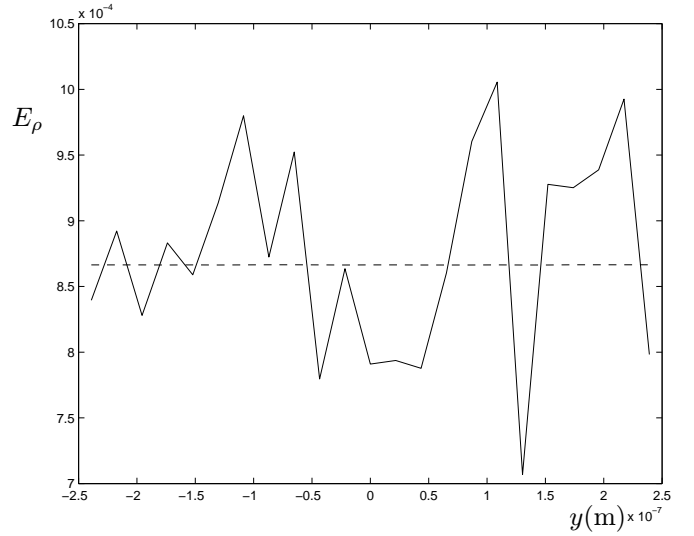


Fig. 2. Fractional error in density for Poiseuille flow in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (9) and the solid line indicates DSMC simulation results.

III. SIMULATIONS

A. Dilute Gases

We performed DSMC simulations to verify the validity of the expressions given above. Standard DSMC techniques [3], [4] were used to simulate flow of gaseous argon (molecular mass $m = 6.63 \times 10^{-26}$ kg, hard sphere diameter $\sigma = 3.66 \times 10^{-10}$ m) in a two-dimensional channel (length L and height H). The simulation was periodic in the x direction (along the channel axis). The two walls at $y = -H/2$ and $y = H/2$ were fully accommodating and flat. The simulation was also periodic in the third (homogeneous) direction.

The average gas density was $\rho_0 = 1.78 \text{ kg/m}^3$ and in all

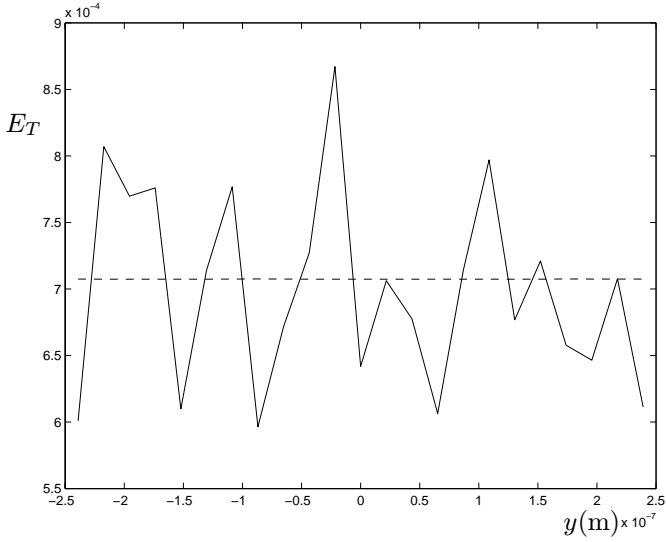


Fig. 3. Fractional error in temperature for Poiseuille flow in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (11) and the solid line indicates DSMC simulation results.

calculations over 40 particles per cell were used. The cell size was $\Delta x = \lambda_0/3$ where λ_0 is the reference mean free path. The time step was $\Delta t = \lambda_0/(7.5c_m)$. For a discussion of the errors resulting from finite cell sizes and time steps see [10], [11], [12]. The fractional error in the simulations is obtained from the standard deviation of cell values in the x and z directions. To ensure that the samples were independent, samples were taken only once every 250 time steps. To ensure that the system was in its steady state the simulation was run for 10^6 time steps before sampling was started.

A constant acceleration was applied to the particles to produce Poiseuille flow in the x direction with maximum velocity at the centerline $u_0^{max} \approx 2$ m/s. Figures 1, 2, and 3 show good agreement between the theoretical expressions from section II and simulation measurements for the fractional error in velocity, density and temperature, respectively. The fractional error in the velocity measurement is minimum at the centerline since the Poiseuille velocity profile is parabolic and maximum at the centerline, (see Fig. 1). The density and temperature were nearly constant across the system so the fractional errors in these quantities are also nearly constant.

The expressions for shear stress and heat flux were verified using Couette (walls at equal temperature with different velocities) and “temperature” Couette (walls at zero velocity with different temperatures) calculations respectively. In these calculations, very small cell sizes ($\Delta x = \lambda/6$) and time steps ($\Delta t = \lambda/(30c_m)$) were used. The momentum and energy fluxes de-correlate faster than the conserved hydrodynamic variables, such as density, so independent samples are obtained after fewer time steps: the system was equilibrated for 10^6 time steps and samples were taken every 50 time steps. Good agreement is found between the theoretical results and simulation measurements

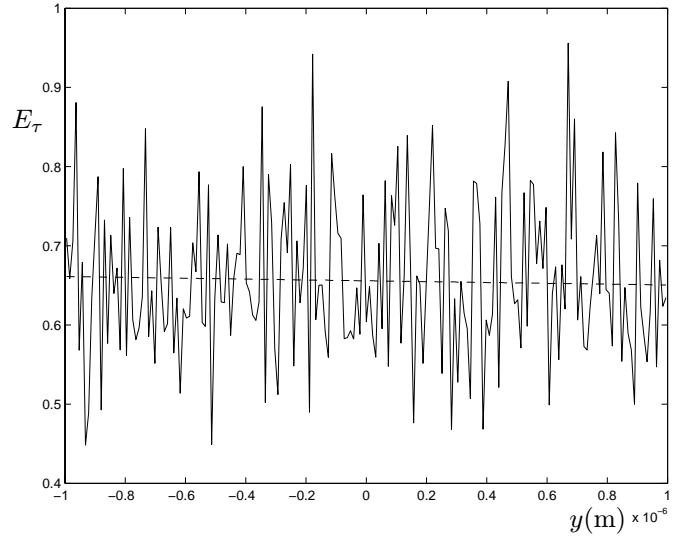


Fig. 4. Fractional error in the shear stress τ_{xy} for Couette flow in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (25) and the solid line indicates DSMC simulation results.

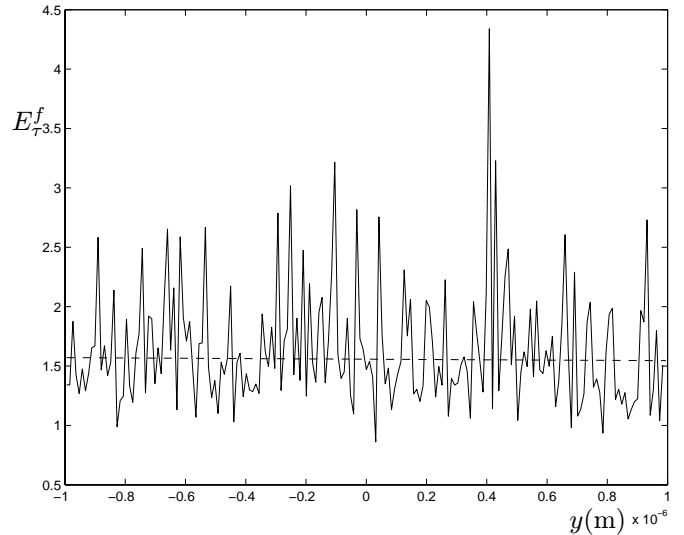


Fig. 5. Fractional error in the shear stress for Couette flow in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (35) and the solid line indicates DSMC simulation results.

for volume averaged and fluxal quantities, as shown in figures 4, 5, 6, 7.

A final note: In DSMC simulations one considers each particle as “representing” a large number of molecules in the physical system. In all the expressions given above, N_0 and N^+ relates to the number of particles used by the simulation so the fluctuations can be reduced by using larger numbers of particles (i.e., using a lower molecule-to-particle ratio).

B. Dense fluids

We performed molecular dynamics simulations to test the validity of equations (5), (9), (11) for dense fluids. A

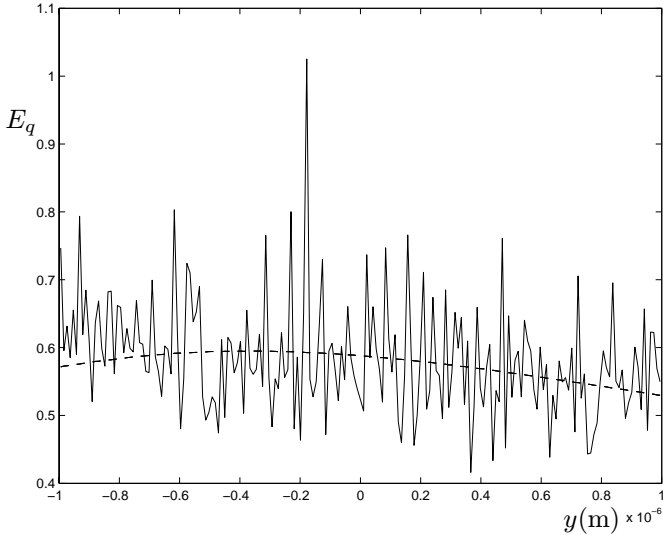


Fig. 6. Fractional error in the heat flux for “Temperature Couette” in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (26) and the solid line indicates DSMC simulation results.

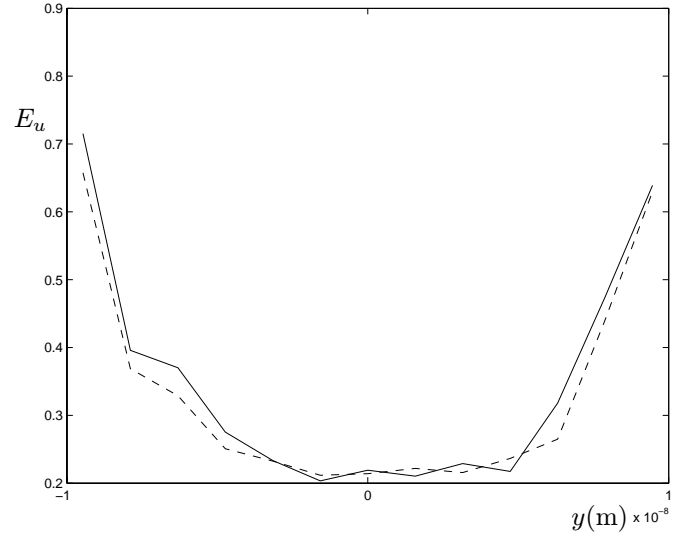


Fig. 8. Fractional error in velocity for dense-fluid Poiseuille flow in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (5) and the solid line denotes MD simulation results.

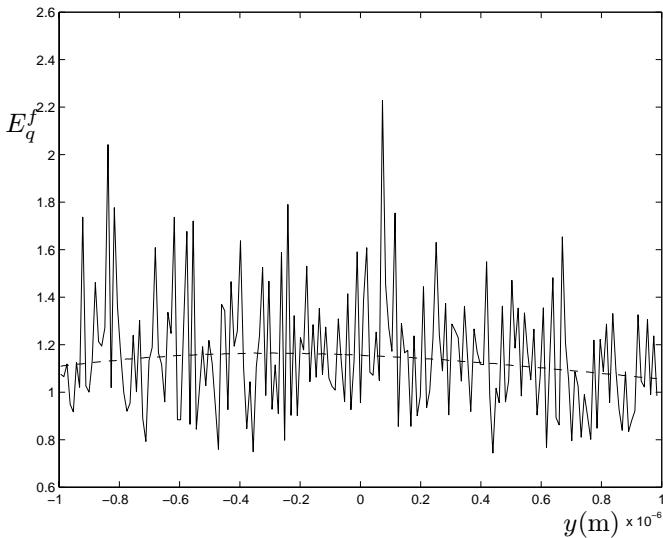


Fig. 7. Fractional error in the heat flux for “Temperature Couette” in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (37) and the solid line indicates DSMC simulation results.

similar geometry to the dilute gas simulations described above was used but at a significantly higher density. In particular, we simulated liquid argon ($\sigma_{LJ} = 3.4 \times 10^{-10}$ m, $\varepsilon_{LJ} = 119.8k_b$) at $T = 240K$ and $\rho = 860Kg/m^3$ in a two-dimensional channel with the x and z directions periodic. The channel height was $H = 69.7\sigma_{LJ}$. The wall molecules were connected to fcc lattice sites through springs and interacted with the fluid through a Lennard-Jones potential with the same parameters. The spring constant $k_s = 460\varepsilon\sigma^{-2}$ was chosen in such a way that the root mean square displacement of wall atoms around their equilibrium position at the simulated temperature was well below the Lindermann criterion for the melting point of a

solid. The length and depth of the system was $28\sigma_{LJ}$ and $29.1\sigma_{LJ}$ in the x and z directions respectively. A constant force $f = 8 \times 10^{-5}\varepsilon/\sigma_{LJ}$ per particle was used to generate a velocity field with a maximum velocity of approximately 13 m/s.

In order to calculate the fluctuation of density, temperature and velocity, we divided the simulation cell into 13 layers in the y -direction with a height $\Delta y = 4.6396\sigma$. We further divided each layer into 49 cells, 7 in each of the x and z directions. The density, temperature and velocity in each cell were calculated every 2000 timesteps; the timestep was taken to be equal to $(0.005t_{LJ})$, where $t_{LJ} = \sqrt{m_{LJ}\sigma_{LJ}^2/\varepsilon_{LJ}}$. We have checked that this time interval is longer than the system’s correlation time such that samples taken between such intervals are independent. For each cell, 200 samples are used to calculate the average density, temperature and velocity. The fluctuation was calculated for each layer using the 49 equivalent cells in the $x - z$ plane.

Due to the sensitivity of the compressibility κ_T on the interaction cutoff r_c , a rather conservative value of $r_c = 4.0\sigma_{LJ}$ was used. We also introduced a correction for the still-finite cutoff which used the compressibility predictions of the Modified Benedict-Webb-Rubin equation of state [13]. The agreement between the theoretical predictions and the simulations is good (see Figures 8, 9 and 10).

IV. CONCLUSIONS

We have presented expressions for the statistical error in estimating the velocity, density, temperature and pressure in molecular simulations. These expressions were validated for flow of a dilute gas and dense liquid in a two-dimensional channel using the direct simulation Monte Carlo and Molecular Dynamics respectively. Despite the non-equilibrium nature of the validation experiments, good

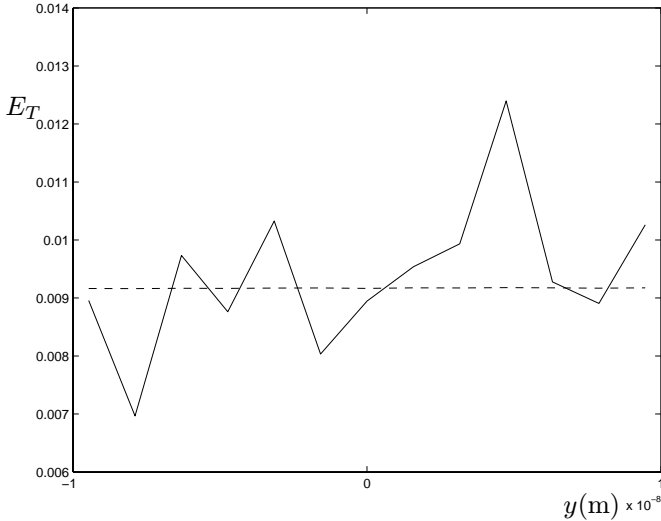


Fig. 9. Fractional error in temperature for dense-fluid Poiseuille flow in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (11) and the solid line indicates MD simulation results.

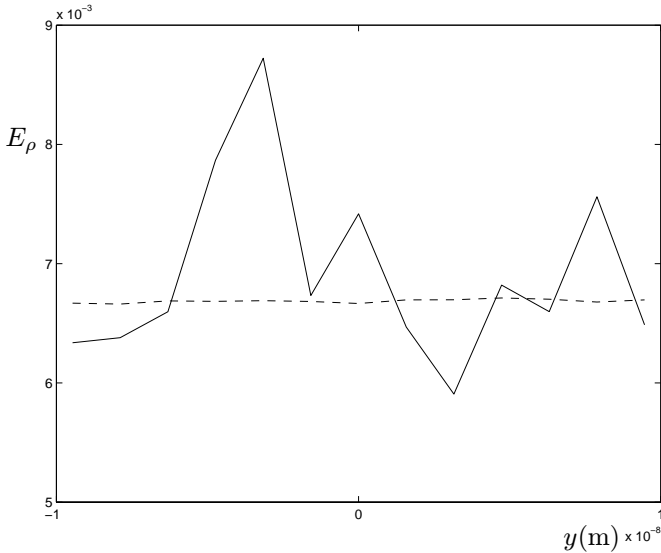


Fig. 10. Fractional error in density for dense-fluid Poiseuille flow in a channel as a function of the transverse channel coordinate, y . The dashed line denotes equation (9) and the solid line indicates MD simulation results.

agreement is found between theory and simulation, verifying that modifications to non-equilibrium results are very small. The agreement with equilibrium theory is particularly remarkable in the dense fluid case where significant non-equilibrium due to a shear of the order of $5.5 \times 10^8 \text{ s}^{-1}$ exists. We thus expect these results to hold for general non-equilibrium applications of interest.

Predictions were also presented for the statistical error in estimating the shear stress and heat flux in dilute gases through cell averaging and surface averaging. Comparison with direct Monte Carlo simulations shows that the equilibrium assumption is justified.

It was found that the fluctuation in state variables is significantly smaller compared to flux variables in the continuum regime $\text{Kn} \rightarrow 0$. This is important for the development of hybrid methods. Although a direct comparison was only presented between volume averaged quantities, we find that the fluxal measurements for the shear stress and heat flux perform similarly to the volume averaged counterparts (regarding scaling with the Knudsen number).

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REFERENCES

- [1] M. P. Allen, D. J. Tildesley. *Computer Simulation of Liquids*, Clarendon Press, Oxford, 1987.
- [2] D. Frenkel and B. Smit, “Understanding Molecular Simulation, From Algorithms to Applications”, Academic Press, San Diego, 2002.
- [3] G. A. Bird. *Molecular Gas Dynamics and the Direct Simulation of Gas Flows*, Clarendon Press, Oxford, 1994.
- [4] F. J. Alexander and A. L. Garcia, “The Direct Simulation Monte Carlo”, *Computers in Physics*, **11**, 588-593, 1997.
- [5] M. Malek-Mansour, A. L. Garcia, G. C. Lie, E. Clementi, “Fluctuating Hydrodynamics in a Dilute Gas”, *Physical Review Letters*, **58**, 874-877, 1987; A. L. Garcia, M. Malek-Mansour, G. Lie, M. Mareschal, E. Clementi, “Hydrodynamic fluctuations in a dilute gas under shear” *Physical Review A*, **36**, 4348-4355, 1987.
- [6] Landau LD, Lifshitz EM. *Statistical Mechanics*. Oxford: Pergamon Press, 1980.
- [7] A.L. Garcia, J.B. Bell, Wm.Y. Crutchfield, and B.J. Alder, “Adaptive Mesh and Algorithm Refinement using Direct Simulation Monte Carlo”, *J. Comp. Phys.* **154** 134 (1999).
- [8] N. G. Hadjiconstantinou, “Hybrid Atomistic-Continuum Formulations and the Moving Contact-Line Problem”, *Journal of Computational Physics*, **154**, 245-265 (1999).
- [9] N. G. Hadjiconstantinou, A. L. Garcia, M. Z. Bazant, and G. He, “Statistical Error in Particle Simulations of Hydrodynamic Phenomena”, submitted to the *Journal of Computational Physics*, July 2002.
- [10] F. J. Alexander, A. L. Garcia and B. J. Alder, “Cell Size Dependence of Transport Coefficients in Stochastic Particle Algorithms”, *Phys. Fluids*, **10** 1540 (1998); Erratum: *Phys. Fluids*, **12** 731 (2000).
- [11] N. G. Hadjiconstantinou, “Analysis of discretization in the direct simulation Monte Carlo”, *Phys. Fluids*, **12**, 2634, 2000.
- [12] A. L. Garcia and W. Wagner, “Time step truncation error in direct simulation Monte Carlo”, *Phys. Fluids*, **12**, 2621, 2000.
- [13] J. K. Johnson, J. A. Zollweg and K. E. Gubbins, “The Lennard-Jones equation of state revisited”, *Molecular Physics*, **78**, 591-618, 1993.