Analysis of transport properties determined by Langevin dynamics using Green–Kubo formulae

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HIGHLIGHTS

- We employ Green–Kubo formulae to investigate the transport properties of Langevin model.
- Langevin velocity model predicts the Prandtl number to be 3/2 for monatomic gas.
- Langevin acceleration model could adjust the Prandtl number with an additional time scale.

ARTICLE INFO

ABSTRACT

Recently, the Langevin dynamics method has been applied to simulate gas flows. It is very crucial to evaluate whether the Langevin dynamics could correctly predict transport properties of gas or not. In this paper, the transport properties of Langevin velocity model and acceleration model are analyzed by using Green–Kubo formulae. For the Langevin velocity model, the time correlation functions have the exact exponent forms, and the Prandtl number for monatomic gas is predicted to be 3/2. For the Langevin acceleration model with an additional time scale, the molecular movements change from Markovian process to Non-Markovian process, and the Prandtl number could be adjusted to some extent. In the limit of equilibrium, there is a minimum about 1.298 for the Prandtl number of monatomic gas when the two time scales are equal in Langevin acceleration model. Besides theoretical analyses, molecular simulations according to the Langevin velocity model and acceleration model are performed, and the simulation results validate our analytical solutions.

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1. Introduction

A stochastic algorithm based on Langevin equation, has been proposed to simulate rarefied gas flows recently [1–3]. Here we call it Langevin simulation Monte Carlo (LSMC) method. Comparing with the direct simulation Monte Carlo (DSMC) method [4,5], which has been very popular in the simulation of rarefied gas flows, the LSMC method is more efficient for simulating small Knudsen number flows. It is known that in DSMC method, the molecular movements and inter-molecular collisions are assumed uncoupled during small time intervals. Molecular motions are modeled deterministically, while inter-molecular collisions are treated statistically. For accurate DSMC applications, the sizes of cell within which molecular collision partners are selected should be less than mean free path of molecules, and the time steps should be less than mean collision time. Therefore, DSMC would become computationally very expensive for the simulation of small Knudsen number flows. On the other hand, the Langevin equation uses drift term and diffusion term to describe molecule movements, and no direct molecular collisions have to be modeled. This allows the LSMC method to proceed with much larger time step than that used in DSMC method. Using Langevin simulation, very good agreement of molecular stresses and mean velocity in comparison with DSMC, linearized Boltzmann and experiment has been achieved [1,3].

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According to the principle of stochastic process, the Langevin equation is closely related to the Fokker–Planck equation [6], and the LSMC method could be regarded as a stochastic solution of the Fokker–Planck equation. The relation between LSMC method and Fokker–Planck equation is similar to the relation between DSMC method and Boltzmann equation. Based on gas kinetic theory, the Fokker–Planck equation could be considered as an approximation of Boltzmann equation [7]. Many researchers have discussed the Fokker–Planck description as a model equation for gas dynamics. For example, Lebowitz et al. [8] and Pawula [9] tried to approximate the Boltzmann equation by a Fokker–Planck equation with a simple drift model, which results in a wrong Prandtl number for monatomic gas molecules. Since the Prandtl number is very important for the heat transport phenomena, many efforts have been made for the Fokker–Planck model to provide correct Prandtl number. Heinz [10] introduced an acceleration model with an additional time scale, which could be used to adjust the Prandtl number. With the acceleration model, the molecule movements change from Markovian process to Non-Markovian process. Yano et al. [11] proposed a Fokker–Planck equation with a source term to correct Prandtl number. Recently, Gorji et al. [2] introduced a cubic non-linear drift term in Fokker–Planck equation, and this model leads to the correct Prandtl number for monatomic gas. In the equilibrium limit, the non-linear drift model automatically regresses to be linear.

In this paper, we employed the Green–Kubo formulae [12] to analyze the transport properties of Langevin equation, including velocity model and acceleration model proposed by Heinz [10]. The Green–Kubo relations give the exact mathematical formulae for transport coefficients in terms of integrals of time correlation functions of some specific microscopic flux. Based on the Langevin model and Ito calculus [13], we could directly obtain the analytical solution of velocity for each molecule, and then the time correlation functions are determined. Using Green–Kubo formula, we obtain the analytical solutions of transport coefficients, and the corresponding results are presented in Sections 2 and 3 for the Langevin velocity model and acceleration model, respectively. In order to validate our analytical results, molecular simulations under equilibrium condition are performed, and the corresponding results are shown in Section 4. The discussions and conclusions are presented in Section 5.

2. Analysis of Langevin velocity model

Let us consider simple Langevin model for the velocity u_x of one molecule in x direction,

$$\frac{du_x}{dt} = -\frac{1}{\tau}(u_x - \langle u_x \rangle) + \left(\frac{4e_s}{3\tau}\right)^{1/2} \frac{dw}{dt}, \tag{1}$$

where $\langle u_x \rangle$ is the mean, or macroscopic velocity of molecules, τ is the characteristic relaxation time scale of molecular velocities, e_s refers to the specific kinetic energy of molecules, i.e., $e_s = \frac{3}{2}\langle u_x^2 \rangle$, and $w(t)$ is a Wiener process with the following properties

$$\left\langle \frac{dw}{dt}(t) \right\rangle = 0, \tag{2}$$

$$\left\langle \frac{dw}{dt}(t) \frac{dw}{dt}(t') \right\rangle = \delta(t - t'). \tag{3}$$

Note that our analysis is performed under equilibrium condition, and hence $\langle u_x \rangle$ equals zero. Meanwhile, the kinetic energy is constant and equal to $\frac{3}{2}RT$, where R is the gas constant, and T is the temperature of system.

Using Ito calculus [13], the exact solution of Eq. (1) could be written as

$$u_x(t) = u_x(0)\, e^{-\frac{t}{\tau}} + \left(\frac{4e_s}{3\tau}\right)^{\frac{1}{2}} \int_0^t e^{-\frac{t-s_x}{\tau}} \frac{dw}{ds_x}(s_x)\, ds_x. \tag{4}$$

Similarly, the evolutions of velocities in y and z direction are

$$u_y(t) = u_y(0)\, e^{-\frac{t}{\tau}} + \left(\frac{4e_s}{3\tau}\right)^{\frac{1}{2}} \int_0^t e^{-\frac{t-s_y}{\tau}} \frac{dw}{ds_y}(s_y)\, ds_y, \tag{5}$$

$$u_z(t) = u_z(0)\, e^{-\frac{t}{\tau}} + \left(\frac{4e_s}{3\tau}\right)^{\frac{1}{2}} \int_0^t e^{-\frac{t-s_z}{\tau}} \frac{dw}{ds_z}(s_z)\, ds_z. \tag{6}$$

Having these expressions of velocity evolutions, the transport coefficients can be obtained by using Green–Kubo relations [12], which give the diffusion, viscosity and thermal conductivity coefficients as follows:

$$D = \int_0^{+\infty} \langle u_x(0)u_x(t) \rangle\, dt, \tag{7}$$

$$\mu = \frac{1}{k_B VT} \int_0^{+\infty} \langle P_{xy}(0)P_{xy}(t) \rangle\, dt, \tag{8}$$

$$\kappa = \frac{1}{k_B VT^2} \int_0^{+\infty} \langle J_x(0)J_x(t) \rangle\, dt, \tag{9}$$

where V is the volume of system, T is the temperature, $P_{xy}(t) = \sum_{i=1}^{N} m u_{i,x} u_{i,y}$ and $J_x(t) = \sum_{i=1}^{N} \left(\frac{1}{2} m u_i^2 - \frac{5}{2} k_B T \right) u_{i,x}$ are the microscopic stress and heat flux, respectively, N is the total number of particles in the system, k_B is the Boltzmann constant, and the $\langle \ \rangle$ denotes an ensemble average. In the following analysis, we firstly calculate the time correlation functions as $\langle u_x(0) u_x(t) \rangle$, $\langle P_{xy}(0) P_{xy}(t) \rangle$ and $\langle J_x(0) J_x(t) \rangle$, and then obtain the transport coefficients by integrating the corresponding time correlation functions.

2.1. Diffusion coefficient

By taking the average of $u_x(0)$ multiplied by $u_x(t)$ as Eq. (4), we could obtain

$$\langle u_x(0) u_x(t) \rangle = \langle u_x^2(0) \rangle \, e^{-\frac{t}{\tau}} + \left(\frac{4e_s}{3\tau} \right)^{\frac{1}{2}} \left\langle u_x(0) \int_0^t e^{-\frac{t-s_x}{\tau}} \frac{dw}{ds_x}(s_x) \, ds_x \right\rangle. \tag{10}$$

Because $\frac{dw}{ds_x}$ is uncorrelated to $u_x(0)$, the ensemble average makes the last term in Eq. (10) zero. The time correlation function of velocity is simplified as

$$\langle u_x(0) u_x(t) \rangle = \langle u_x^2(0) \rangle \, e^{-\frac{t}{\tau}} = \frac{2e_s}{3} \, e^{-\frac{t}{\tau}}. \tag{11}$$

Consequently, the diffusion coefficient could be obtained using Green–Kubo formula as Eq. (7),

$$D = \frac{2e_s}{3} \int_0^{+\infty} e^{-\frac{t}{\tau}} \, dt = \frac{2e_s}{3} \tau. \tag{12}$$

2.2. Viscosity coefficient

Due to the independence of particles in Langevin dynamics, the ensemble average terms of inter-particles equal zero. And thus the time correlation function of microscopic stress can be simplified as

$$\langle P_{xy}(0) P_{xy}(t) \rangle = \left\langle \sum_{i=1}^{N} m u_{i,x}(0) u_{i,y}(0) \cdot \sum_{j=1}^{N} m u_{j,x}(t) u_{j,y}(t) \right\rangle$$
$$= N m^2 \langle u_x(0) u_y(0) u_x(t) u_y(t) \rangle. \tag{13}$$

On substituting Eqs. (4) and (5) into Eq. (13), we obtain

$$\langle P_{xy}(0) P_{xy}(t) \rangle = N m^2 \, e^{-\frac{2t}{\tau}} \langle u_x^2(0) u_y^2(0) \rangle + N m^2 \left(\frac{4e_s}{3\tau} \right)^{\frac{1}{2}} \left\langle u_x(0) u_y^2(0) \int_0^t e^{-\frac{t-s_x}{\tau}} \frac{dw}{ds_x}(s_x) \, ds_x \right\rangle$$
$$+ N m^2 \left(\frac{4e_s}{3\tau} \right)^{\frac{1}{2}} \left\langle u_x^2(0) u_y(0) \int_0^t e^{-\frac{t-s_y}{\tau}} \frac{dw}{ds_y}(s_y) \, ds_y \right\rangle$$
$$+ N m^2 \frac{4e_s}{3\tau} \left\langle u_x(0) u_y(0) \int_0^t e^{-\frac{t-s_x}{\tau}} \frac{dw}{ds_x}(s_x) \, ds_x \int_0^t e^{-\frac{t-s_y}{\tau}} \frac{dw}{ds_y}(s_y) \, ds_y \right\rangle. \tag{14}$$

Considering that the inter-particles and inter-directions are uncorrelated, and the Wiener process is uncorrelated to initial velocities, the last three terms in Eq. (14) equal zero. Hence Eq. (14) reduces to

$$\langle P_{xy}(0) P_{xy}(t) \rangle = N m^2 \, e^{-\frac{2t}{\tau}} \langle u_x^2(0) \rangle \langle u_y^2(0) \rangle = \frac{4e_s^2}{9} N m^2 \, e^{-\frac{2t}{\tau}}. \tag{15}$$

Integrating the time correlation function of microscopic stress, we obtain the viscosity coefficient

$$\mu = \frac{1}{k_B V T} \frac{4e_s^2}{9} N m^2 \int_0^{+\infty} e^{-\frac{2t}{\tau}} \, dt = \frac{P\tau}{2}, \tag{16}$$

where P is the pressure, i.e., $P = n k_B T$, and n is the number density, i.e., $n = N/V$.

2.3. Thermal conductivity coefficient

Similarly, considering the independence of particles, the time correlation function of microscopic heat flux reduces to

$$\langle J_x(0) J_x(t) \rangle = N \left\langle \left(\frac{1}{2} m u^2(0) - \frac{5}{2} k_B T \right) u_x(0) \cdot \left(\frac{1}{2} m u^2(t) - \frac{5}{2} k_B T \right) u_x(t) \right\rangle, \tag{17}$$

where $u^2(0) = u_x^2(0) + u_y^2(0) + u_z^2(0)$ and $u^2(t) = u_x^2(t) + u_y^2(t) + u_z^2(t)$.

On substituting Eqs. (4)–(6) into Eq. (17) and making some simplifications, we obtain

$$\langle J_x(0)J_x(t)\rangle = N\left\langle \left(\frac{1}{2}mu^2(0) - \frac{5}{2}k_BT\right)^2 u_x^2(0)\, e^{-\frac{3t}{\tau}}\right\rangle. \tag{18}$$

By taking the moments of the Maxwellian distribution function under equilibrium condition, we could obtain $\langle u_x^2(0)\rangle = \frac{2e_s}{3}$, $\langle u_x^4(0)\rangle = \frac{4e_s^2}{3}$ and $\langle u_x^6(0)\rangle = \frac{40e_s^3}{9}$. And thus Eq. (18) could be written as

$$\langle J_x(0)J_x(t)\rangle = \frac{20e_s^3}{27}Nm^2\, e^{-\frac{3t}{\tau}}. \tag{19}$$

Integrating the time correlation function of microscopic heat flux, we obtain the thermal conductivity coefficient

$$\kappa = \frac{1}{k_BVT^2}\frac{20e_s^3}{27}Nm^2\int_0^{+\infty} e^{-\frac{3t}{\tau}}\, \mathrm{d}t = \frac{5PR\tau}{6}. \tag{20}$$

2.4. Prandtl number

The Prandtl number is a dimensionless number approximating the ratio of momentum diffusivity and thermal diffusivity and can be expressed as

$$\mathrm{Pr} = \frac{C_p\mu}{\kappa}, \tag{21}$$

where C_p is the specific heat capacity. For monatomic gas, $C_p = \frac{5}{2}R$. On substituting Eqs. (16) and (20) into Eq. (21), the Prandtl number equals 3/2 for monatomic gas. This analytical result is the same as that presented in Jenny et al.'s paper [1], where they derived the Prandtl number by taking moments of the Fokker–Planck equation. It is known that the Prandtl number is 2/3 for monatomic gas, so the simple Langevin velocity model could not predict correct Prandtl number.

3. Analysis of Langevin acceleration model

Since the simple Langevin velocity model cannot predict correct Prandtl number for monatomic gas, some developments have been made for adjusting Prandtl number. Heinz [10] proposed the acceleration model with an additional time scale τ_a for describing molecular movements,

$$\frac{\mathrm{d}u_x}{\mathrm{d}t} = a_x, \tag{22}$$

$$\frac{\mathrm{d}a_x}{\mathrm{d}t} = \frac{1}{\tau_a}\left[-\left(1+\frac{\tau_a}{\tau}\right)a_x - \frac{1}{\tau}u_x + \left(\frac{4e_s}{3\tau}\left(1+\frac{\tau_a}{\tau}\right)\right)^{1/2}\frac{\mathrm{d}w}{\mathrm{d}t}\right], \tag{23}$$

where a_x is the acceleration in x direction. The velocity model as Eq. (1) can be recovered by multiplying Eq. (23) by τ_a and taking the limit $\tau_a \to 0$. For the sake of derivation easily, letting $r_1 = -\frac{1}{\tau}$ and $r_2 = -\frac{1}{\tau_a}$, Eq. (23) changes to simple form as

$$\frac{\mathrm{d}a_x}{\mathrm{d}t} = (r_1+r_2)a_x - r_1r_2u_x + \left(-\frac{4e_s}{3}r_1r_2(r_1+r_2)\right)^{1/2}\frac{\mathrm{d}w}{\mathrm{d}t}. \tag{24}$$

The formal solution of Eq. (24) gives

$$a_x(t) = a_x(0)\, e^{(r_1+r_2)t} + \int_0^t e^{(r_1+r_2)(t-s)}\left[-r_1r_2u_x + \left(-\frac{4e_s}{3}r_1r_2(r_1+r_2)\right)^{1/2}\frac{\mathrm{d}w}{\mathrm{d}s}(s)\right]\mathrm{d}s. \tag{25}$$

It can be seen from Eq. (25) that the acceleration model describes a non-Markovian process because the future velocity is calculated in terms of the velocity history between zero and t. This is the main difference between the acceleration model and velocity model.

Combining Eq. (24) with Eq. (22), we could obtain the exact solution of velocity

$$u_x(t) = a_x(0)\frac{e^{r_2t} - e^{r_1t}}{r_2 - r_1} + u_x(0)\frac{r_2\, e^{r_1t} - r_1\, e^{r_2t}}{r_2 - r_1}$$

$$+ \frac{1}{r_2 - r_1}\left(-\frac{4e}{3}r_1r_2(r_1+r_2)\right)^{1/2}\int_0^t (e^{r_2(t-s)} - e^{r_1(t-s)})\frac{\mathrm{d}w}{\mathrm{d}s}(s)\, \mathrm{d}s. \tag{26}$$

By differentiating Eq. (26) with respect to t, the expression of acceleration follows as

$$a_x(t) = a_x(0) \frac{r_2 \, e^{r_2 t} - r_1 \, e^{r_1 t}}{r_2 - r_1} + u_x(0) \frac{r_1 r_2 (e^{r_1 t} - e^{r_2 t})}{r_2 - r_1}$$
$$+ \frac{1}{r_2 - r_1} \left(-\frac{4e}{3} r_1 r_2 (r_1 + r_2) \right)^{1/2} \int_0^t (r_2 \, e^{r_2(t-s)} - r_1 \, e^{r_1(t-s)}) \frac{dw}{ds}(s) \, ds. \tag{27}$$

The velocities and accelerations in y and z directions have the same forms as Eqs. (26) and (27), respectively. Considering that energy is conserved under equilibrium condition, these relations $\langle u_x^2(t) \rangle = \frac{2e_s}{3}$, $\langle a_x^2(t) \rangle = \frac{2e_s}{3} r_1 r_2$ and $\langle u_x(t) a_x(t) \rangle = 0$ are necessarily satisfied. Using these expressions, the transport coefficients could be obtained by employing Green–Kubo formulae.

3.1. Diffusion coefficient

By taking the average of $u_x(0)$ multiplied by $u_x(t)$ as Eq. (26), we obtain

$$\langle u_x(0) u_x(t) \rangle = \langle u_x(0) a_x(0) \rangle \frac{e^{r_2 t} - e^{r_1 t}}{r_2 - r_1} + \langle u_x^2(0) \rangle \frac{r_2 \, e^{r_1 t} - r_1 \, e^{r_2 t}}{r_2 - r_1}$$
$$+ \frac{1}{r_2 - r_1} \left(-\frac{4e}{3} r_1 r_2 (r_1 + r_2) \right)^{1/2} \left\langle u_x(0) \int_0^t (e^{r_2(t-s)} - e^{r_1(t-s)}) \frac{dw}{ds}(s) \, ds \right\rangle. \tag{28}$$

Because $\langle u_x(t) a_x(t) \rangle = 0$ and $u_x(0)$ is uncorrelated to $\frac{dw}{ds}$, both of the first and last terms in Eq. (28) are zero. And thus the time correlation function of velocity is simplified as

$$\langle u_x(0) u_x(t) \rangle = \frac{2e_s}{3} \frac{r_2 \, e^{r_1 t} - r_1 \, e^{r_2 t}}{r_2 - r_1}. \tag{29}$$

Consequently, the diffusion coefficient could be obtained using Green–Kubo formula as Eq. (7),

$$D = \frac{2e_s}{3} \int_0^{+\infty} \frac{r_2 \, e^{r_1 t} - r_1 \, e^{r_2 t}}{r_2 - r_1} \, dt = \frac{2e_s}{3} \frac{-(r_1 + r_2)}{r_1 r_2} = \frac{2e_s}{3} (\tau + \tau_a). \tag{30}$$

3.2. Viscosity coefficient

On substituting Eqs. (26) and (27) into Eq. (13), we obtain the time correlation function of microscopic stress,

$$\langle P_{xy}(0) P_{xy}(t) \rangle = \frac{4e_s^2}{9} Nm^2 \frac{(r_2 \, e^{r_1 t} - r_1 \, e^{r_2 t})^2}{(r_2 - r_1)^2}. \tag{31}$$

Integrating the time correlation function of microscopic stress, we obtain the viscosity coefficient,

$$\mu = \frac{Nm^2}{k_B V T} \frac{4e_s^2}{9} \int_0^{+\infty} \frac{(r_2 \, e^{r_1 t} - r_1 \, e^{r_2 t})^2}{(r_2 - r_1)^2} \, dt$$
$$= \frac{Nm^2}{k_B V T} \frac{4e_s^2}{9} \frac{-((r_1 + r_2)^2 + r_1 r_2)}{2 r_1 r_2 (r_1 + r_2)}$$
$$= \frac{p(\tau^2 + 3\tau \tau_a + \tau_a^2)}{2(\tau + \tau_a)}. \tag{32}$$

3.3. Thermal conductivity coefficient

On substituting Eqs. (26) and (27) into Eq. (17) and making some simplifications, we obtain the time correlation function of microscopic heat flux,

$$\langle J_x(0) J_x(t) \rangle = \frac{20 e_s^3}{27} Nm^2 \frac{(r_1 \, e^{r_2 t} - r_2 \, e^{r_1 t})^3}{(r_2 - r_1)^3}. \tag{33}$$

Integrating the time correlation function of microscopic heat flux, we obtain the thermal conductivity coefficient,

$$\kappa = \frac{Nm^2}{k_B V T^2} \frac{20 e_s^3}{27} \int_0^{+\infty} \frac{(r_2 \, e^{r_1 t} - r_1 \, e^{r_2 t})^3}{(r_2 - r_1)^3} \, dt$$
$$= \frac{Nm^2}{k_B V T^2} \frac{20 e_s^3}{27} \frac{-(r_1 + r_2)(2 r_1^2 + 9 r_1 r_2 + 2 r_2^2)}{3 r_1 r_2 (2 r_1 + r_2)(r_1 + 2 r_2)}$$
$$= \frac{5 p R}{6} \frac{(\tau + \tau_a)(2 \tau_a^2 + 9 \tau_a \tau + 2 \tau^2)}{(2 \tau_a + \tau)(\tau_a + 2 \tau)}. \tag{34}$$
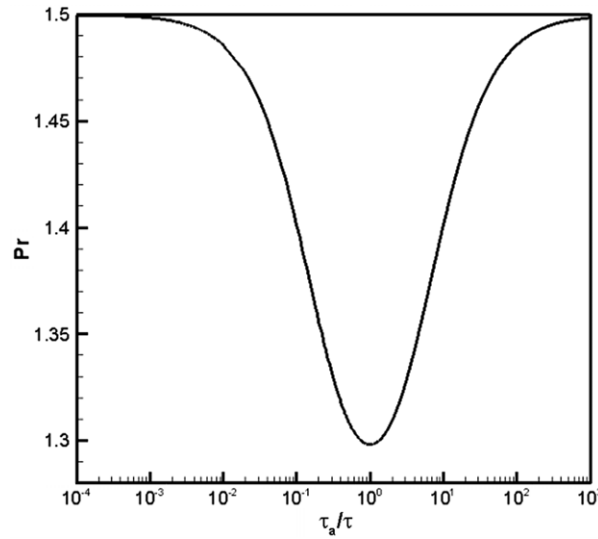
Fig. 1. The Prandtl number versus τ_a/τ for the Langevin acceleration model.

3.4. Prandtl number

On substituting Eqs. (32) and (34) into Eq. (21), we obtain the Prandtl number determined by Langevin acceleration model for monatomic gas,

$$\mathrm{Pr} = \frac{3(2\tau_a + \tau)(\tau_a + 2\tau)(\tau^2 + 3\tau\tau_a + \tau_a^2)}{2(\tau + \tau_a)^2(2\tau_a^2 + 9\tau_a\tau + 2\tau^2)}. \tag{35}$$

It is obvious that the Prandtl number varies with the ratio of two time scales, i.e., τ_a/τ. As shown in Fig. 1, when $\tau_a/\tau \to 0$ or $\tau_a/\tau \to +\infty$, the Prandtl number has the maximum value 1.5; when the two time scales are equal, the Prandtl number has the minimum value, which is about 1.298. Comparing with the Langevin velocity model, the Langevin acceleration model could adjust the Prandtl number to some extent, but cannot approach to the correct value 2/3 for monatomic gas. Note that our analysis is performed in the limit of equilibrium, τ and τ_a are assumed constant. Under non-equilibrium condition, τ and τ_a may vary in space via temperature dependence, and it is possible to adjust Prandtl number to the correct value 2/3 for monatomic gas. More details have been presented in Heinz's paper [10].

4. Molecular simulations

To validate our analysis results, molecular simulations for Argon gas under standard state are performed, i.e., the temperature and pressure of the system are 273 K and 1.01×10^5 Pa, respectively. The computational domain is a three-dimensional box, and each boundary condition of which is assumed periodic. The introduction of periodic boundaries is equivalent to considering an infinite, space-filling array of identical copies of the simulation region. In the process of simulation, a molecule that leaves the simulation region through a particular boundary immediately reenters the region through the opposite boundary. As the DSMC method, each simulation molecule in the LSMC method also represents a large number of real molecules. The total number of simulation molecules is about 800 thousands. At the initial time, molecules are uniformly distributed in the simulation domain, and their velocities and accelerations are randomly selected from Normal distributions. The calculating time step is 0.1τ, where $\tau = 4.179 \times 10^{-10}$ s, which is determined according to Eq. (16) to ensure the viscosity predicted by Langevin velocity model is the same as the experimental value, 2.117×10^{-5} Nsm^{-2}. For Langevin acceleration model, we choose $\tau = 8.039 \times 10^{-11}$ s and $\tau_a = 2.752 \times 10^{-10}$ s, which make the viscosity predicted by acceleration model is the same as experimental value and the Prandtl number equals 4/3. In each calculating time step, the update of molecular velocities for Langevin velocity model is based on Eq. (4), while the update of molecular velocities and accelerations for Langevin acceleration model is based on Eqs. (26) and (27). For both models, the evolutions of molecular positions are performed according to

$$\Delta x(t) = \int_0^t u(t') \, dt'. \tag{36}$$

For numerical simulations, the terms containing Wiener process in Eqs. (4), (26), (27) and (36) are replaced by sampling from a normal distribution. Jenny et al. [1] and Ermak and Buckholz [14] have developed this type of numerical schemes. It is crucial that the first and second conditional moments and joint conditional moments predicted by numerical solutions should be consistent with that predicted by the original evolution equations.

Particularly, for the Langevin velocity model, the numerical solution for each calculating time step Δt follows as

$$x(\Delta t) = x(0) + u(0)\tau(1 - e^{-\Delta t/\tau}) + \sqrt{B}\varepsilon_1, \tag{37}$$

$$u(\Delta t) = u(0)\, e^{-\Delta t/\tau} + \sqrt{\frac{C^2}{B}}\varepsilon_1 + \sqrt{A - \frac{C^2}{B}}\varepsilon_2, \tag{38}$$

where ε_1 and ε_2 are independent, normal distributed random variables, and A, B and C are related to the moments of velocities and displacements,

$$A = \frac{2e_s}{3}\left(1 - e^{-2\Delta t/\tau}\right), \tag{39}$$

$$B = \frac{2e_s\tau^2}{3}\left(\frac{2\Delta t}{\tau} - \left(1 - e^{-\Delta t/\tau}\right)\left(3 - e^{-\Delta t/\tau}\right)\right), \tag{40}$$

$$C = \frac{2e_s\tau}{3}\left(1 - e^{-\Delta t/\tau}\right)^2. \tag{41}$$

For the Langevin acceleration model, the numerical solution for each calculating time step Δt follows as

$$x(\Delta t) = x(0) + a(0)\frac{r_1(e^{r_2 t} - 1) - r_2(e^{r_1 t} - 1)}{(r_2 - r_1)r_1 r_2} - u(0)\frac{r_1^2(e^{r_2 t} - 1) - r_2^2(e^{r_1 t} - 1)}{(r_2 - r_1)r_1 r_2} + \sqrt{A}\varepsilon_1, \tag{42}$$

$$u(\Delta t) = a(0)\frac{e^{r_2 t} - e^{r_1 t}}{r_2 - r_1} + u(0)\frac{r_2 e^{r_1 t} - r_1 e^{r_2 t}}{r_2 - r_1} + \sqrt{\frac{D^2}{A}}\varepsilon_1 + \sqrt{B - \frac{D^2}{A}}\varepsilon_1, \tag{43}$$

$$\begin{aligned}
a(\Delta t) &= a(0)\frac{r_2 e^{r_2 t} - r_1 e^{r_1 t}}{r_2 - r_1} + u(0)\frac{r_1 r_2(e^{r_1 t} - e^{r_2 t})}{r_2 - r_1} \\
&\quad + \sqrt{\frac{E^2}{A}}\varepsilon_1 + \sqrt{\frac{(AF - DE)^2}{A(AB - D^2)}}\varepsilon_2 + \sqrt{C - \frac{E^2}{A} - \frac{(AF - DE)^2}{A(AB - D^2)}}\varepsilon_3,
\end{aligned} \tag{44}$$

where ε_1, ε_2 and ε_3 are independent, normal distributed random variables, and A, B, C, D, E and F are related to the moments of velocities, accelerations and displacements,

$$\begin{aligned}
A &= -\frac{2e}{3}\frac{r_2^2(3 - 4e^{r_1 t} + e^{2r_1 t})}{(r_2 - r_1)^2 r_1^2} + \frac{2e}{3}\frac{(4 - 4e^{r_1 t} - 4e^{r_2 t} + 4e^{(r_1 + r_2)t} + 2r_2 t)}{(r_2 - r_1)^2} \\
&\quad - \frac{2e}{3}\frac{r_2(-1 + e^{2r_1 t} + 2r_2 t)}{(r_2 - r_1)^2 r_1} + \frac{2e}{3}\frac{r_1(1 - e^{2r_2 t} + 2r_2 t)}{(r_2 - r_1)^2 r_2} - \frac{2e}{3}\frac{r_1^2(3 - 4e^{r_2 t} + e^{2r_2 t} + 2r_2 t)}{(r_2 - r_1)^2 r_2^2},
\end{aligned} \tag{45}$$

$$B = \frac{2e}{3}\frac{(r_2 - r_1)^2 - r_1 r_2(e^{r_2 t} - e^{r_1 t})^2 + (r_1 e^{r_2 t} - r_2 e^{r_1 t})^2}{(r_2 - r_1)^2}, \tag{46}$$

$$C = \frac{2e}{3}r_1 r_2\frac{(r_2 - r_1)^2 - (r_1 + r_2)(r_1 e^{2r_1 t} + r_2 e^{2r_2 t}) - 4r_1 r_2 e^{(r_1 + r_2)t}}{(r_2 - r_1)^2}, \tag{47}$$

$$D = -\frac{2e}{3}\frac{(r_1 + r_2)(r_1(-1 + e^{r_2 t}) + r_2(1 - e^{r_1 t}))^2}{r_1 r_2(r_2 - r_1)^2}, \tag{48}$$

$$E = \frac{2e}{3}\frac{(1 + 2e^{r_1 t} - e^{r_2 t})(e^{r_2 t} - 1)r_1^2}{(r_2 - r_1)^2} - \frac{2e}{3}\frac{(e^{2r_1 t} - e^{2r_2 t} - 2)r_1 r_2}{(r_2 - r_1)^2} + \frac{2e}{3}\frac{(1 + 2e^{2r_2 t} - e^{r_1 t})(e^{r_1 t} - 1)r_2^2}{(r_2 - r_1)^2}, \tag{49}$$

$$F = -\frac{2e}{3}\frac{r_1 r_2(r_1 + r_2)(e^{r_2 t} - e^{r_1 t})^2}{(r_2 - r_1)^2}. \tag{50}$$

Using the numerical schemes as Eqs. (37)–(41) for the Langevin velocity model and as Eqs. (42)–(50) for the Langevin acceleration model, molecular simulations are proceeded up to 1 million time steps, and then ensemble average are performed to obtain the time correlation functions. Figs. 2 and 3 show the time correlation functions of microscopic stress and heat flux, respectively. The time scale is normalized by the parameter τ in Langevin velocity model. It is obvious that the time correlation functions for the velocity model have the exponent forms. At the initial stage the time correlation functions for acceleration model are larger than that for velocity model, and after no longer than τ, the time correlation functions for acceleration model become less than that for velocity model. Note that the parameter τ in velocity model and the parameters τ and τ_a in acceleration model are determined by the viscosity coefficient, so the integrations of time correlation function of microscopic stress for the velocity model and acceleration model are the same, which predict correctly the viscosity coefficient. However, the integration of time correlation function of microscopic heat flux for acceleration model is larger
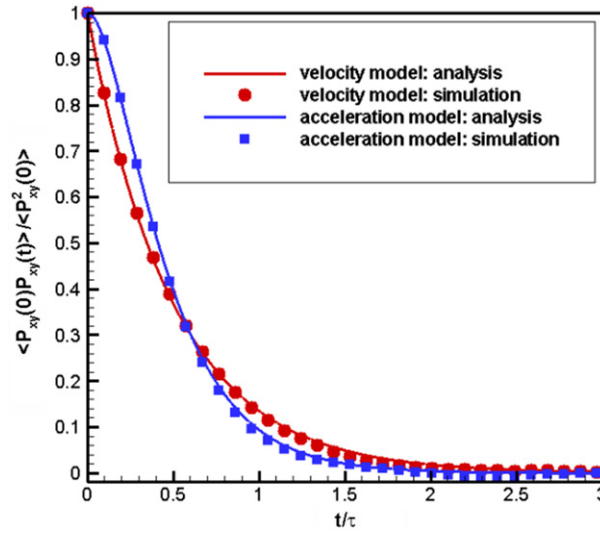
Fig. 2. Time correlation function of microscopic stress versus time.
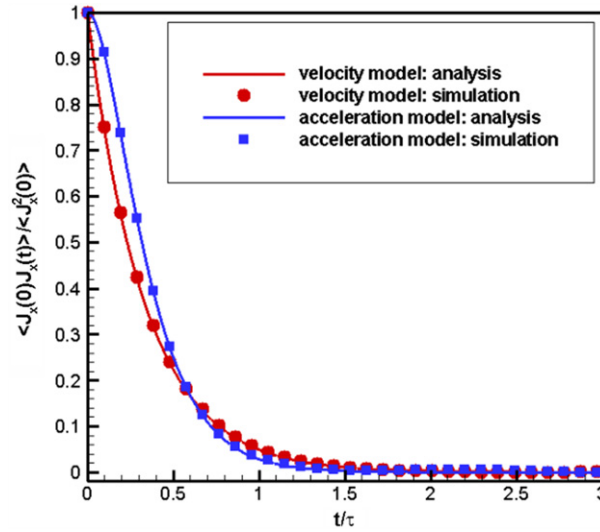

Fig. 3. Time correlation function of microscopic heat flux versus time.

than that for velocity model, and it means that the thermal conductivity coefficient predicted by acceleration model is larger than that for velocity model. In this way, acceleration model could decrease the Prandtl number to some extent.

Fig. 4 shows the acceleration correlation function for acceleration model. After about 0.3τ the correlation becomes minus, then approaches to the minimum at about 0.6τ, and finally goes back to zero at about 3τ. The integration of acceleration correlation function is zero, and it means that the acceleration has a zero integral correlation time [13].

5. Discussions

In this paper, transport properties of Langevin velocity model and acceleration model are investigated using Green–Kubo formulae. The analytical solutions of transport coefficients, including diffusion, viscosity, and thermal conductivity coefficients are obtained. For the Langevin velocity model, the time correlation functions have the exact exponent forms, and the predicted Prandtl number is 3/2 for monatomic gas. For the Langevin acceleration model with an additional time scale, the time correlation functions are somewhat different from that of velocity model, and the Prandtl number could be adjusted to some extent, but the minimum of Prandtl number predicted by acceleration model is still larger than the correct value 2/3 for monatomic gas. Recently, a non-linear velocity model has been proposed by Gorji et al. [2], and the cubic non-linear drift term leads to the correct Prandtl number of 2/3 for monatomic gas under nonequilibrium state. However, in the limit of equilibrium, the non-linear velocity model automatically reduces to the simple velocity model, which predicts the Prandtl
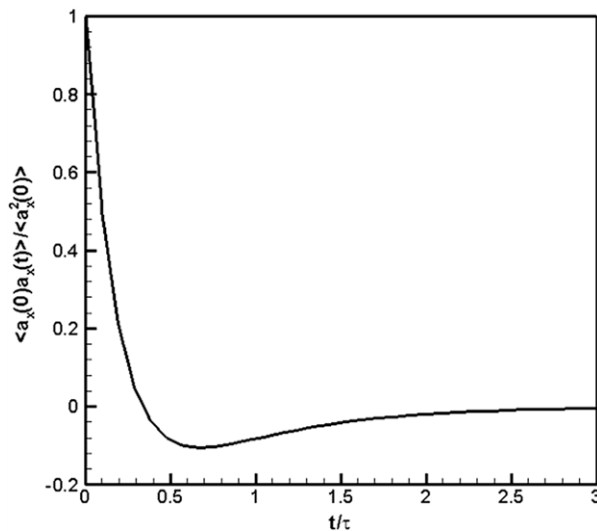
Fig. 4. Acceleration correlation function for the Langevin acceleration model.

number as 3/2. To the best our knowledge, none of Langevin models could predict correct Prandtl number for monatomic gas in the limit of equilibrium by far. How to develop a reasonable Langevin model for predicting correct transport properties is still an open question. Note that we only consider ideal particle systems in this paper, especially the ideal gas molecules. For the nonideal dissipative system, inter-particle interactions play an important role in the transport properties. The problems associated with mass-transfer processes, i.e., diffusion processes, in dissipative systems of interacting particles [15–18] have been widely investigated in various fields, such as in plasmas of combustion products and in polymer colloid suspensions. Yet the research of viscosity and thermal conductivity coefficients of these systems is still rare. It is worthwhile to study on this subject in the future.

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References

 [1] P. Jenny, M. Torrilhon, S. Heinz, A solution algorithm for the fluid dynamic equations based on a stochastic model for molecular motion, J. Comput. Phys. 229 (2010) 1077–1098.
 [2] M.H. Gorji, M. Torrilhon, P. Jenny, Fokker–Planck model for computational studies of monatomic rarefied gas flows, J. Fluid Mech. 680 (2011) 574–601.
 [3] F. Fei, J. Fan, A diffusion information preservation method for small Knudsen number flows, J. Comput. Phys. 243 (2013) 179–193.
 [4] G.A. Bird, Molecular Gas Dynamics and Direct Simulation of Gas Flows, Clarendon, Oxford, 1994.
 [5] C. Shen, Rarefied Gas Dynamics: Fundamentals, Simulations and Micro Flows, Springer, Berlin, 2005.
 [6] H. Risken, The Fokker–Planck Equation, Springer-Verlag, Berlin, 1984.
 [7] C. Cercignani, The Boltzmann Equation and its Application, Springer-Verlag, New York, 1988.
 [8] J.L. Lebowitz, H.L. Frisch, E. Helfand, Nonequilibrium distribution functions in a fluid, Phys. Fluids 3 (1960) 325–338.
 [9] R.F. Pawula, Approximation of the linear Boltzmann equation by the Fokker–Planck equation, Phys. Rev. 162 (1967) 186–188.
[10] S. Heinz, Molecular to fluid dynamics: the consequences of stochastic molecular motion, Phys. Rev. E 70 (2004) 036308.
[11] R. Yano, K. Suzuki, H. Kuroda, Analytical and numerical study on the nonequilibrium relaxation by the simplified Fokker–Planck equation, Phys. Fluids 21 (2009) 047104.
[12] R. Kubo, M. Toda, N. Hashitsume, Statistical Physics II: Nonequilibrium Statistical Mechanics, Springer-Verlag, Berlin, 1985.
[13] S. Heinz, Mathematical Modeling, Springer-Verlag, Berlin, 2011.
[14] D.L. Ermak, H. Buckholz, Numerical-integration of the langevin equation: Monte Carlo simulation, J. Comput. Phys. 35 (1980) 169–182.
[15] O.S. Vaulina, X.G. Adamovich, O.F. Petrov, V.E. Fortov, Evolution of the mass-transfer processes in nonideal dissipative systems. I. Numerical simulation, Phys. Rev. E 77 (2008) 066403.
[16] O.S. Vaulina, X.G. Adamovich, O.F. Petrov, V.E. Fortov, Evolution of the mass-transfer processes in nonideal dissipative systems II: experiments in dusty plasma, Phys. Rev. E 77 (2008) 066404.
[17] V.E. Fortov, O.F. Petrov, O.S. Vaulina, K.G. Koss, Brownian motion of dust particles in a weakly ionized plasma, JETP Lett. 97 (2013) 322–326.
[18] O.S. Vaulina, X.G. Koss, The dynamics of mass transfer in equilibrium correlated systems, Phys. Scr. 88 (2013) 055502.