

Molecular-Dynamics Simulations of Nonequilibrium Heat and Momentum Transport in Very Dilute Gases

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Molecular-dynamics simulations were used to study the heat and momentum transport phenomena in very dilute gases flowing through a two-dimensional channel. Aside from pronounced slips in velocity and temperature at the wall, Navier-Stokes equations seem to be valid up to the maximum Knudsen number (Kn), 0.27, studied. It is further shown that the viscosity of a very dilute gas decreases monotonically with the decreasing density. For Kn less than ~ 0.05 , the slip coefficient, 1.16, calculated from the simulation is in good agreement with theoretical results, ranging from 1.134 to 1.230. However, at higher Kn, the slip coefficient is predicted to depend on the Knudsen number.

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The kinetic theory of a dilute gas, based on the Chapman-Enskog solution of the Boltzmann equation, has been very successful in describing nonequilibrium transport phenomena in a dilute gas.¹ On the basis of this theory one cannot only derive the field equations describing the nonequilibrium heat and momentum transport at the macroscopic level, but also obtain the temperature dependence of the transport coefficients. One of the important features of this theory is its surprising and successful prediction that for a dilute gas, various transport coefficients do not depend on the density.

However, there exist a significant number of experiments which show that when the pressure is reduced significantly, the thermal conductivity and the shear viscosity of a dilute gas are not longer density independent, but monotonically decrease with the decreasing density. The very first such experiment was done in 1875 by Knudt and Warburg.² Similarly Smoluchowski³ and Knudsen⁴ showed that when the gas is sufficiently dilute, the velocity and temperature profiles differ significantly from the predictions of continuum hydrodynamics inasmuch that there are pronounced velocity and temperature slips at the wall which increase with increasing rarefaction. Since no macroscopic transport theory has yet been developed which can explain such phenomena as observed by Knudt and Warburg, Smoluchowski, and Knudsen, there appears a need to study this at the molecular level itself.

In this paper we seek to achieve this by simulating nonequilibrium heat and momentum transport using the molecular-dynamic methods.⁵ We are particularly interested in the density regime where the classical transport theory based on the first-order Chapman-Enskog theory may break down. The origin of the inherent limitation of the Chapman-Enskog method can be traced back to the fact that in this approach the distribution function is expressed in the power series of the Knudsen

number, which is defined as $Kn = \lambda/L$, where λ is the mean free path of the gas molecule and L is a characteristic length of the system. The first-order term gives rise to the Navier-Stokes and Fourier equations. Thus the continuum description is valid only as long as the Knudsen number is sufficiently small. Two interesting situations arise when the Knudsen number is large and the Chapman-Enskog solution is likely to break down. The first is when the geometric dimension of the system is very small. One such important area is that of heat generation in integrated chips in modern supercomputers where the appropriate cooling strategies require the heat analysis in a system having microscopic dimensions.⁶ Second, the Knudsen number could also be large when the mean free path of the particle is large. The mean free path being inversely proportional to density, the determination of the flow and heat transfer around a body (such as a space shuttle or an aeroassisted orbital transfer vehicle) moving in the upper atmosphere requires an extension of continuum hydrodynamic concepts.⁷

In the present Letter, we report a novel simulation of nonequilibrium heat and momentum transport in a two-dimensional channel in a density regime characterized by a Knudsen number ranging from 0.01 to 0.30. Using the method of molecular dynamics, we study the dynamics of 2500 gas particles (argon atoms), interacting via a soft-sphere Lennard-Jones-type potential⁸⁻¹⁰ and enclosed between two parallel stochastic thermal plates⁸⁻¹⁰ placed at $x = L/2$ and $-L/2$ and kept at 300 K. The flow in the y direction is induced by subjecting the particles to an external gravitational field of $g = 5 \times 10^{13}$ cm/sec², which produces a flow corresponding to a Reynolds number of the order of 20. We show that as the density is decreased progressively, the flow and temperature profiles exhibit a sharp jump at the wall and the shear viscosity decreases monotonically. Simulation results are also used to calculate the slip coefficient, ap-

TABLE I. Parameters used in the simulation of the two-dimensional channel flows (total number of the particles in the system is 2500).

Run No.	Length of box (Å)	Kn	λ/σ
1	469.7	0.012	1.7
2	1183.6	0.031	10.9
3	2024.0	0.053	31.9
4	3212.9	0.085	80.3
5	4799.5	0.127	179.3
6	5494.1	0.145	235.0
7	6992.1	0.185	380.6
8	10304.1	0.272	826.6

pearing in the Maxwell model of slip velocity at the wall, and this is compared with the theoretical results obtained by several authors.¹¹⁻¹⁵ The agreement with the theoretical results is very good. The details of the molecular-dynamics simulation followed closely those of Hannon, Lie, and Clementi,⁸⁻¹⁰ except that the time step used here was 2×10^{-14} sec. Other parameters used in the present simulations are summarized in Table I. Statistics were collected by dividing the $L \times L$ box into cells with $x:y = 100:1$ and averaging over 10000 steps.

In a two-dimensional system we define the Knudsen number Kn, which characterizes the degree of rarefactions by

$$\text{Kn} = \lambda/L = (\sqrt{2} \pi n \sigma L)^{-1}, \quad (1)$$

where n is the number density per unit area and σ is the Lennard-Jones parameter signifying the particle's diameter. Another index of the rarefaction is the ratio λ/σ . In Table I, we have given the values of Kn and λ/σ for the parameters shown in our simulations. For $\text{Kn} = 0.012$, we find $\lambda/\sigma = 1.71$, and thus it would reflect the properties of a dense fluid, while the others (Kn varying from 0.03 to 0.27) represent the dilute gas at various degrees of rarefaction.

The average velocity u calculated in the x direction is zero, as expected. The velocity profiles in the y direction are shown in Fig. 1 for five of the eight runs reported in Table I. The velocities have been scaled with a reference velocity $v_r = 0.14 \times 10^6$ cm/sec which represents the velocity at the center of the channel for $\text{Kn} = 0.27$. For a dense fluid at $\text{Kn} = 0.012$, the velocity at the wall is almost zero, which has also been observed in several other simulations of the channel flow.⁹ However, as the density decreases there is a pronounced slip at the wall. In the scaled unit the slip velocity increases from 0.05 for run 2 to 0.26 for run 8. Furthermore, as the density decreases, the temperature jump (the difference between the wall temperature and the average temperature of gas particles next to the wall) increases rapidly. The velocity and the temperature slips found in our simulation are analogous to the ones observed by Smoluchowski and

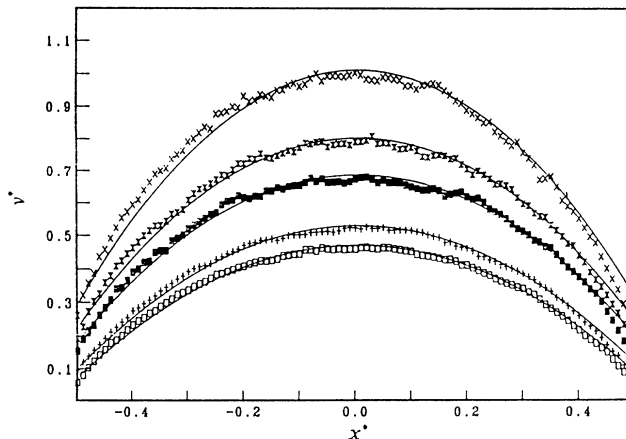


FIG. 1. Velocity profiles for various values of the Knudsen number. From bottom to top: $\text{Kn} = 0.031, 0.053, 0.12, 0.185,$ and 0.27 . The solid lines are the fitted curves of Eq. (2) to the simulation results.

Knudsen in their pioneering experiments,^{3,4} and are a direct consequence of the nonequilibrium arising from infrequent collisions among the gas particles at very low densities.

If the variation in density and transport coefficients across the channel can be neglected, then the Navier-Stokes and the Fourier equations can be solved exactly giving a quadratic velocity and a quartic temperature profile across the channel.^{9,10} Thus if the simulations obey the continuum mechanics, the velocity and the temperature profiles should exhibit, respectively, a quadratic and a quartic dependence on x^* ($\equiv x/L$). Our simulations, as shown in Fig. 1 for the velocity, show that this is indeed the case: The standard deviation in the fitting for the velocity profile with a parabolic form is less than 4%, while the error in fitting the temperature profile with a quartic form is around 10%. This may sound surprising at first for very dilute gases, but as has been demonstrated in many different situations,^{8-10,16} the validity of the Navier-Stokes equation seems to extend far beyond their "theoretical" limits.

The shear viscosity can be estimated either from the maximum velocity at the center of the channel or from the curvature of the velocity profile.⁹ At $\text{Kn} = 0.031$, when the gas is *dilute* and the temperature at the center of the channel is 480 K, the viscosity obtained through the simulation, $\eta = 18.3 \times 10^{-12}$ g/sec, is in good agreement with the theoretical result, 18.5×10^{-12} g/sec, obtained using the analysis of Gass for dilute gases.¹⁷

In Fig. 2, we display the variation of the shear viscosity with the density. Consistent with the prediction of Knudt and Warburg,² we find that the shear viscosity of a slightly rarefied gas decreases with decreasing density. This also indicates that the first-order Chapman-Enskog solution of the Boltzmann equation showing viscosity independent of density is no longer true in the range of the

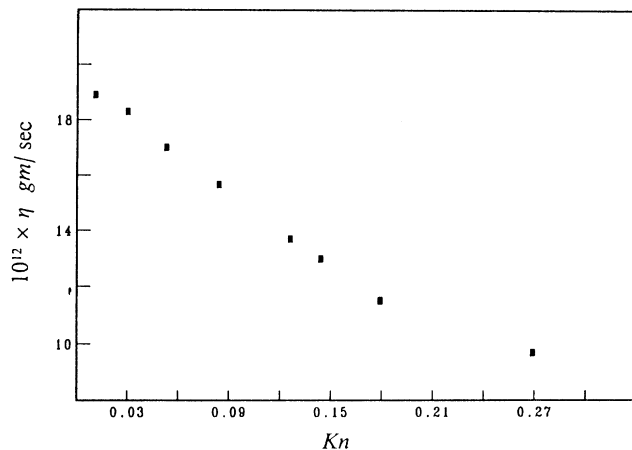


FIG. 2. Variation of the shear viscosity with the Knudsen number obtained from the simulations.

Knudsen number considered here. Note that Fig. 2 also contains a temperature dependence because as the Knudsen number increases, the temperature of the gas inside the channel also increases. However, as the viscosity usually increases with increasing temperature, the density dependence would be even more pronounced if we take the temperature into account. Since the $T^{1/2}$ dependence of the viscosity may no longer hold for the rarefied gases studied here, it apparently needs more detailed investigation, which is currently underway in our laboratory.

We now turn our attention to the calculation of the slip coefficient. Maxwell, on the basis of his analysis of the kinetics of a dilute gas near an idealized solid wall, predicted that the slip velocity at the wall in the first-order approximation is given by¹⁸

$$v_w = a\lambda (dv/dx)_w, \quad (2)$$

where a is the *slip coefficient*, which in turn depends on the momentum accommodation coefficient of particles representing how well the gas molecules stick at the wall. Several authors have calculated the slip coefficient, making various approximations in the Boltzmann equation and presuming that the Knudsen number is in the slip-flow regime ($\text{Kn} \approx 0.01-0.1$).¹¹⁻¹⁵ We can rewrite Eq. (2) as

$$l_s = v_w / (dv/dx)_w = a \text{Kn}, \quad (3)$$

where l_s is the dimensionless slip length. This equation shows that the slip length should be proportional to Knudsen number. In Fig. 3, we study the variation of the slip length with the Knudsen number. For a small Knudsen number ($\text{Kn} \lesssim 0.05$), the slip coefficient 1.16 (measured by the slope of the curve) is indeed constant and is in good agreement with the various theoretical predictions ranging from 1.134 to 1.230, as shown in Table II. For larger Knudsen numbers ($\text{Kn} > 0.05$) the

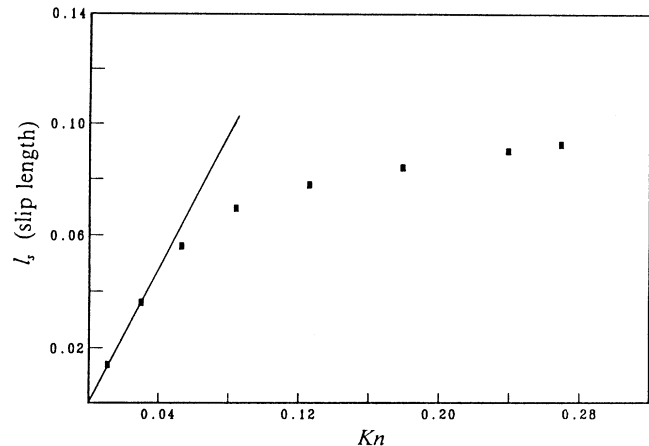


FIG. 3. Variation of the slip length with the Knudsen number obtained from the simulations. The straight line is obtained by a least-squares fit of the first two simulation data and the origin (which implies that we are assuming there would be no slip as the Knudsen number goes to zero). The slope of the line gives the slip coefficient discussed in the text.

slope clearly varies with the density. This has an important implication. It appears that the Maxwell model with an experimentally determined value of the slip coefficient can be used to calculate the slip velocity at the wall only if the Knudsen number is sufficiently small. The apparent breakdown of the Maxwell model at a large Knudsen number may be due to the importance of the higher-order velocity derivatives or to the coupling with the temperature gradient which has been neglected in the derivation of Eq. (2).

Since the velocity and the temperature slips at the wall could also depend, apart from the density, on the absorbed-gas-film characteristic, it would be necessary to

TABLE II. Comparison of the slip coefficient obtained from the present molecular-dynamics simulations with various theoretical calculations based on the Boltzmann equation.

Authors	Slip coefficient	Ref. No.
Welander ^a	1.230 ^b	11
Wang Chang and Uhlenbeck ^a	1.134	12
Willis ^c	1.142	13
Ziering ^a	1.16	14
Albertoni, Cercignani, and Cususo ^c	1.146	15
Bhattacharya and Lie	1.16	This work

^aBased on approximate solutions of the Boltzmann equation for Maxwell molecules.

^bThere are some doubts as to the accuracy of this work (see Refs. 16 and 18).

^cBased on numerical calculations of an exact analytical expression of the solution for an approximate model of the Boltzmann equation for Maxwell molecules (Ref. 20).

incorporate a more detailed analysis of the gas-surface interaction. We are now in the process of refining the simulation reported here. The improvement we envisage consists in attributing molecular properties to the wall.¹⁹ In the new simulation, the system is divided into two distinct regions, one consisting of wall molecules and the other consisting of gas molecules. The specification of the various molecular interactions and the molecular dynamics of the composite system then allows us to study how the chemical and the physical state of the bounding surface influences the nonequilibrium flow properties of a very dilute gas.

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