

Breakdown of Translational and Rotational Equilibrium in Gaseous Expansions

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The direct simulation Monte Carlo method has been used to study the breakdown of translational equilibrium in steady cylindrical and spherical expansions of hard sphere and Maxwell molecules. The study of spherical expansions was extended to the combined translational and rotational breakdown in a gas of rough sphere molecules. The breakdown of translational equilibrium in a complete one-dimensional rarefaction wave in a hard sphere gas was also investigated. In all cases, the breakdown of equilibrium was found to coincide with a constant value of the ratio of the logarithmic time derivative of density following the motion of the fluid to the collision frequency in the gas. This value is proposed as an empirical breakdown criterion for use in engineering studies of systems which involve low-density expansions from continuum to highly rarefied conditions. The onset of nonequilibrium was marked by the divergence of the separate kinetic temperatures based on the molecular velocity components parallel and normal to the flow direction. The parallel temperature in a steady expansion gradually froze to a constant value, in qualitative agreement with experiment and with analytical studies employing the BGK model. The rate of decay of the temperature based on the normal velocity components was greater than the isentropic rate for hard sphere molecules, but less than it was for Maxwell molecules.

Nomenclature

a	= speed of sound
K_n	= λ_0/r_* , throat Knudsen number
k	= power index for asymptotic dependence of P on M
L	= $\rho(dx/d\rho)$ density scale length
M	= Mach number
M'	= Mach number at which P is a minimum
n	= power index for the dependence of flow area on radius
P	= $(1/\rho v)(D\rho/Dt)$
r	= radius
R	= gas constant
Re_L	= $(\rho u L)/\mu$ Reynolds number based on L
T	= kinetic temperature
t	= time
u	= flow velocity
v	= molecular velocity component
v_s	= average molecular speed
x	= position coordinate
β	= reciprocal of most probable molecular speed
γ	= specific heat ratio of gas
λ	= mean free path
μ	= coefficient of viscosity
ν	= collision frequency
ρ	= density
ω	= power index for the dependence of μ on T

Superscript

' = peculiar velocity component

Introduction

THE breakdown of translational and rotational equilibrium in steady expansions and in unsteady rarefaction waves is a basic problem in fluid mechanics.

Experimental work¹⁻³ has concentrated on steady freejet expansions of monatomic gases. These are used in low-density testing and for the production of molecular beams, and in both cases, it is necessary to know the translational state of the gas. A feature of the early measurements¹ was the determination of the terminal or "freezing" Mach number of the flow. If the freezing process was "sudden" as had been assumed in some earlier discussions,⁴ determination of the terminal conditions would also provide information on the initial breakdown of translational equilibrium. However, the later studies^{2,3} clearly showed that the freezing of the parallel temperature occurs gradually over a large range of radius and Mach number. The experiments have been limited in both accuracy and range of parameters and do not provide a criterion for defining the point of initial breakdown. This is not important in many practical situations since the density and flow velocity are barely affected by the breakdown of equilibrium. However, there is an increasing requirement for the detailed analysis of very low-density nozzle flows and jet plumes. For these, it is necessary to have some criterion for the point at which a continuum approach must be replaced by a particle approach.

The most significant analytical studies have been those of Hamel and Willis⁵ and Edwards and Cheng,⁶ who independently obtained solutions for steady hypersonic source flow. These predicted the gradual freezing of the parallel kinetic temperature in a spherical expansion and provided a criterion for the onset of nonequilibrium. However, they employed the BGK model for the collisional terms in the Boltzmann equation, and the connection between the predicted flow behaviour and that of a real gas is uncertain.

This paper presents results for both steady and unsteady expansions obtained from the direct simulation Monte Carlo

Subscripts

*	= sonic conditions
0	= stagnation conditions
1	= upstream boundary of simulated flow
2	= downstream boundary of simulated flow
a	= axial component
i	= leading edge of rarefaction wave
x	= longitudinal, radial, or parallel component
n	= lateral, circumferential, or normal component

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method.^{7,8} This is a technique for the computer modelling of a real gas flow by several thousand simulated molecules. The velocity components and position coordinates of the simulated molecules are stored in the computer and are modified with time as the molecules are concurrently followed through representative collisions and boundary interactions in simulated physical space. The paths of the molecules are therefore followed exactly in phase space and the left hand side of the Boltzmann equation is obviously satisfied. The procedures for the selection of collision pairs are consistent with the formulation of the loss term of the collision integral, which forms the right hand side of the Boltzmann equation. The simulated molecules form a closed set, so the correct choice of collision pairs automatically assures that the gain term in the collision integral is correctly represented. The arbitrary assumptions regarding this term that are made in the BGK and similar model equations are therefore avoided, and the Monte Carlo method can be shown⁹ to provide a numerical solution of the full Boltzmann equation.

Outline of Method

The general procedures in the direct simulation Monte Carlo method are common to all applications and have been described in recent papers.⁷⁻⁹ The special features of the programs used in this study are as follows.

Steady Radial Expansion

Since the flow has either cylindrical or spherical symmetry, the only position coordinate that need be stored for each molecule is the radius. For spherical flows of monatomic gases, only the radial and circumferential velocity components need be stored, and colliding molecules may be assigned random azimuth angles about the radial direction. In the case of cylindrical flows, the velocity distribution is not axially symmetric in velocity space, and the axial velocity component must also be stored for each molecule. For rough sphere molecules, the three angular velocity components must be stored in addition to the three translational velocity components. The collision mechanics of rough sphere molecules have been presented by Chapman and Cowling.¹⁰ The mass of the molecule is here assumed to be uniformly distributed over its volume, and the resultant rotational relaxation time is only two mean collision times.

It should be noted that although advantage is taken of the over-all spherical or cylindrical symmetry, the molecule moves in three dimensions and the axes for the velocity components must be constantly realigned with the radial and circumferential directions. A stream or directed velocity exists only in the radial direction and is given by

$$u = \langle v_x \rangle$$

The over-all temperature is defined for monatomic gases by

$$\begin{aligned} 3RT &= \langle v_x'^2 + v_n'^2 \rangle \\ &= \langle v_x^2 \rangle - u^2 + \langle v_n^2 \rangle \end{aligned}$$

Separate parallel and normal temperatures are defined for spherical flows by

$$RT_x = \langle v_x^2 \rangle - u^2$$

and

$$2RT_n = \langle v_n^2 \rangle$$

For cylindrical flows, the normal temperature component may be subdivided into the axial and circumferential components. These equations define the translational temperature for the rough sphere molecules and this must be combined with the rotational temperature based on the rotational velocity components.

The flow was simulated between two specified radii. Approximately 3000 simulated molecules were set up initially (at zero time) with positions and velocity components appropriate to a steady isentropic expansion between the two radii. This region was divided into approximately 300 equal width cells. The molecules were then set in motion and typical collisions were computed as the time parameter was advanced. Fresh molecules were generated at the upstream boundary with velocity components appropriate to the downstream moving molecules in an equilibrium flow at the upstream Mach number. Molecules which moved back upstream across this boundary were removed from the calculation. Similarly, molecules which moved downstream across the downstream boundary were removed. Since the flow Mach number at the downstream boundary was always supersonic, it was not necessary to generate any upstream moving molecules. The throat and stagnation conditions are those appropriate to an ideal isentropic flow to the upstream boundary.

If the throat Knudsen number was such that the flow remained in equilibrium throughout the simulated region, the macroscopic flow properties would remain constant with time. In practice, this Knudsen number was chosen such that nonequilibrium began at a point between the two radii. An unsteady process then occurred during which the initially isentropic flow downstream of the breakdown point decayed to a steady nonequilibrium flow. It was found that the steady flow was established in the time required for a fluid element to traverse the nonequilibrium region. Sampling of the flow properties commenced when the steady flow had been established. The basic cells were combined into groups of five or six for the sampling, so the flow properties were printed out at approximately 50 locations in the flow. The sampling process was repeated at time intervals sufficiently large for the successive samples to be independent. This continued until the sample size at each location was of the order of three thousand. The computations were carried out on the IBM 360-75 at the California Institute of Technology. A typical case involved the computation of about 100,000 typical collisions and required 5 to 10 min computing time.

Complete Rarefaction Wave

One position coordinate and three velocity components were stored for each simulated molecule. The zero time configuration was a stationary uniform gas in thermal equilibrium between the origin and a point at a specified number of mean free paths from the origin. The molecules could be regarded as being confined between two specularly reflecting walls and, at zero time, the wall at the origin was removed and the gas expanded into a vacuum.

A feature of this program was that a Lagrangian system of cells moving with the fluid was adopted instead of the fixed Eulerian cells employed in the program for the steady expansion. The first cell consisted of the first (say) ten molecules in physical space, the next cell the following ten and so on. The sorting of molecules required by this procedure caused a significant increase in computing time as compared with the simple cross-referencing system used with the conventional Eulerian approach. This additional time is proportional to the square of the total number of simulated molecules. The final runs were therefore made with the total number of molecules restricted to 1600 in 160 cells. The initial extent of the flow was 30 mean free paths and the calculation was terminated when the leading edge of the expansion wave arrived at the fixed wall. The flow properties were sampled at set time intervals during the flow. The cells were combined into groups of four, and in order to further build up a sufficiently large sample size at each location and time, an ensemble average was taken of the results from 40 separate runs. A single run required approximately 40 sec on the IBM 360-75 and involved about 12,000 collisions.

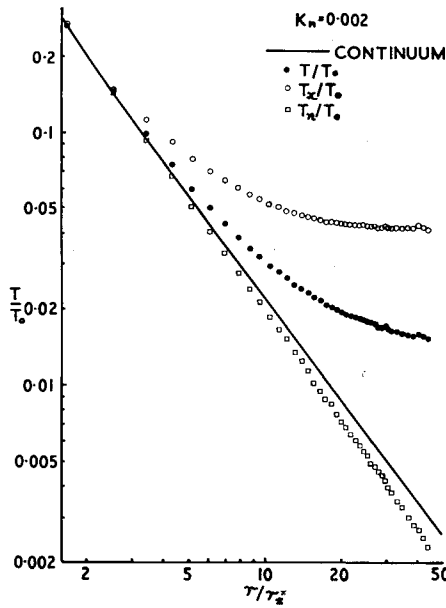


Fig. 1 Breakdown of translational equilibrium in the spherical expansion of a hard sphere gas.

It is sometimes claimed that Monte Carlo methods make extravagant demands on both programming effort and computing funds. The steady flow and rarefaction wave programs comprised approximately 350 and 200 FORTRAN statements, respectively, and were assembled from what have become standard routines. Moreover, the total cost of the computing time including both development and production runs was approximately \$1600.

Results for Steady Expansion

Although the major objective was to obtain a criterion for the initial breakdown of equilibrium, runs were made over a wide range of Mach number for both hard sphere and Maxwell molecules in order to obtain an over-all picture of the process. The result for hard sphere molecules is shown in Fig. 1. This gives qualitative support to one of the major predictions of the BGK solutions—that the freezing of the radial or parallel temperature T_x occurs gradually over a wide range of radius

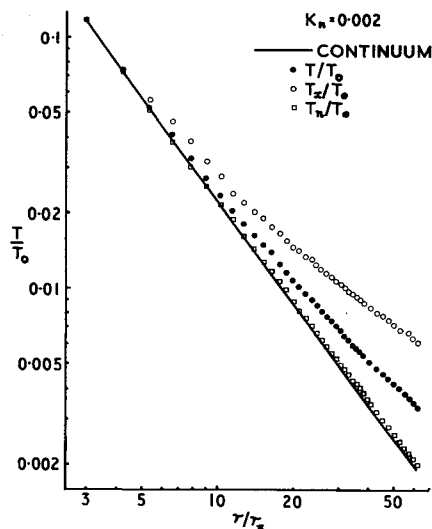


Fig. 2 Breakdown of translational equilibrium in the spherical expansion of a Maxwell gas.

and Mach number. The Hamel and Willis theory⁵ predicts a value of 10.8 for “pseudo-hard sphere molecules, $c = 2$ ” at this throat Knudsen number, while the Monte Carlo result is 8.4. Figure 2 shows a similar calculation for a gas of Maxwell molecules. The freezing of the parallel temperature is much more gradual and it is not possible in a calculation of reasonable magnitude to reach the frozen situation.

In the case of the normal or circumferential temperature T_n , there is a qualitative difference between the results for the two molecular models, and the comparison with theory is more complex. The BGK analysis predicts T_n to follow the collision-dominated isentropic curve with $T_n \propto r^{-4/3}$ and then change quite suddenly to an r^{-1} decay. On the other hand, the “sudden freeze” model would give a transition to an r^{-2} decay law caused entirely by the geometric effect. The Monte Carlo result for the hard sphere gas is that the T_n curve gradually drops below the $r^{-4/3}$ curve and does not appear to have reached an asymptotic rate in this example. The Maxwell gas result in Fig. 2 is significantly different in that the T_n curve remains above the $r^{-4/3}$ curve. The rate of decay is greater than the r^{-1} rate predicted by the BGK analysis. However, a 9° deflection angle cut-off was applied in the computation, and although this is satisfactory in most applications, the neglect of the many weak interactions might have produced a significant error in this situation. Hamel and Willis⁵ pointed out that $T_x \gg T_n$ at large radii and a very small amount of collisional transfer can produce large effects on T_n . Edwards and Cheng¹¹ elaborate on this point and show that the far-field collisional transfer is largely contained in the tail of the distribution function. This raises the point as to whether the comparatively small sample of molecules in the Monte Carlo simulation accurately models a phenomenon depending on the tail of a distribution. The time averaged sample at each of the points in Figs. 1 and 2 is over 5000 and this normally provides an adequate representation of the tail of a distribution. Also, the computational details such as cell size and sample number were identical in the two cases, and since there were sufficient collisions in the Maxwell case to keep the T_n curve above the $r^{-4/3}$ rate even with the cut-off, the falling of the hard sphere curve below this rate is almost certainly significant. The rate of decay of T_n in a real monatomic gas would be between the Maxwell and hard sphere rates and might therefore be expected to be very close to the isentropic rate. Muntz⁹ measured T_n in free expansions and obtained values which did not depart significantly from the isentropic curve. Values of T_n lying well below the isentropic curve are quoted in Ref. 2, but T_n was there defined by the Boltzmann distribution with the best fit to the observed distribution, and the low values of T_n may have been due to the failure to properly account for the energy in the tails of the distribution.

The more detailed runs concentrating only on the region of initial breakdown are listed in Table 1. These runs conform to the specification of sample, cell, and molecule numbers given in the previous section. The run shown in Fig. 1 was necessarily made with wider cells and a smaller number of cells than is normally considered desirable. The left-hand set of curves in Fig. 3 corresponds to the initial part of Fig. 1, and the two are in excellent agreement. This and other tests indi-

Table 1 List of cases

Case number	Flow geometry	Molecular type	$Kn = \lambda_0/r_0$	M_1	M_2 (continuum)	r_1/r_0	r_2/r_0
1	Spherical	Hard sphere	0.002	2	7	1.24	4.91
2	Spherical	Hard sphere	0.001	4	10	2.38	8.14
3	Spherical	Hard sphere	0.0005	4	12	2.38	10.6
4	Spherical	Maxwell	0.002	4	10	2.38	8.14
5	Spherical	Maxwell	0.0005	8	20	5.93	22.5
6	Spherical	Rough sphere	0.002	2	5	1.32	5.05
7	Spherical	Rough sphere	0.001	2.5	6	1.75	9.40
8	Cylindrical	Hard sphere	0.005	2	10	1.53	66.3

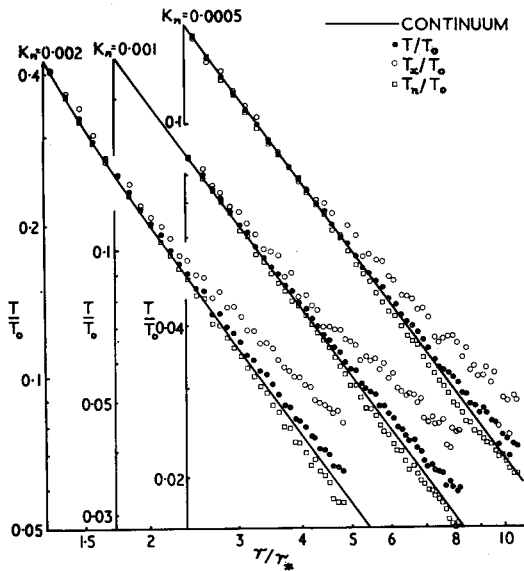


Fig. 3 Initial breakdown in spherical expansions of a hard sphere gas.

cated that the specification of the numerical aspects of the computation was conservative.

Cases 1, 2 and 3 are for the spherical expansion of a hard sphere gas, and the kinetic temperatures are shown as a function of radius in Fig. 2. As expected, the breakdown of translational equilibrium occurs at progressively larger radii as the throat Knudsen number decreases. The effect of the change to Maxwell molecules in cases 4 and 5 is shown in Fig. 4. The behavior is generally similar, although for a given Knudsen number the breakdown occurs at a larger radius and Mach number. Also, the rate of divergence of T_x and T_n is much smaller.

Cases 6 and 7 involve rough sphere molecules that have six degrees of freedom and form a gas with $\gamma = \frac{4}{3}$. The rate of decrease of temperature with radius is therefore much slower than in the monatomic gas simulations in cases 1–5. The results for the rough sphere gas are shown in Fig. 5 with the over-all kinetic temperature divided into the translational and rotational temperatures, with the former further divided into the parallel and normal components. The rotational and translational temperatures depart from equilibrium at the same time as the parallel and normal components of the translational temperature do so. The rotational temperature departs from equilibrium at a slightly greater rate than the parallel component of the translational temperature. The

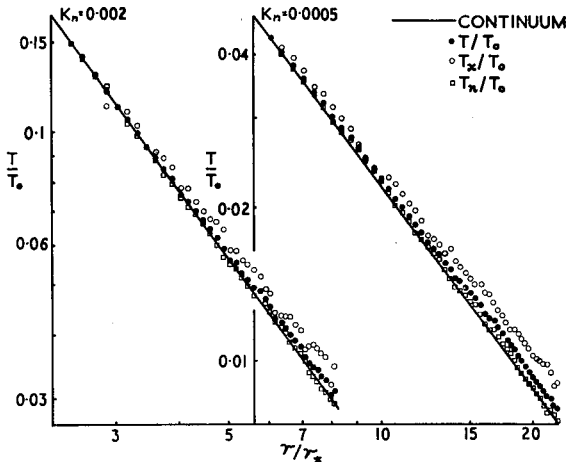


Fig. 4 Initial breakdown in spherical expansions of a Maxwell gas.

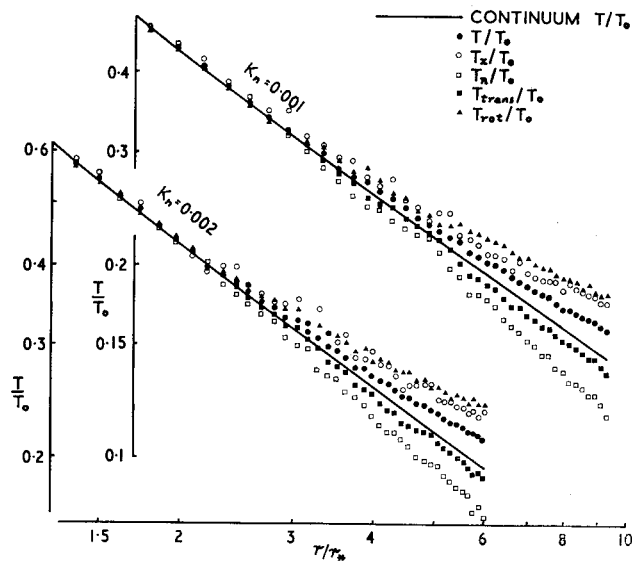


Fig. 5 Combined breakdown of translational and rotational equilibrium in spherical expansions of a rough sphere gas.

normal component falls at a higher rate due to the geometrical effect. The net result is that the rate of fall of the over-all temperature is slightly less than the continuum rate while that of the translational temperature is slightly greater.

The simulation of cylindrical expansions introduces practical difficulties because of the low rate of fall of density with radius compared with spherical expansions. This means that the simulation must extend over a large distance, and only one case (number 8 in Table 1) was computed with the result shown in Fig. 6. The velocity distribution is no longer symmetrical about the flow direction in velocity space, and the three components of the kinetic temperature are shown. The geometric effect now applies to only one component and the temperature based on the velocity components parallel to the axis of the cylinder departs from the continuum curve at much the same rate as the parallel temperature. The circumferential temperature now contains one third rather than two thirds of the energy and the increased energy transfer prevents it from falling significantly below the continuum curve.

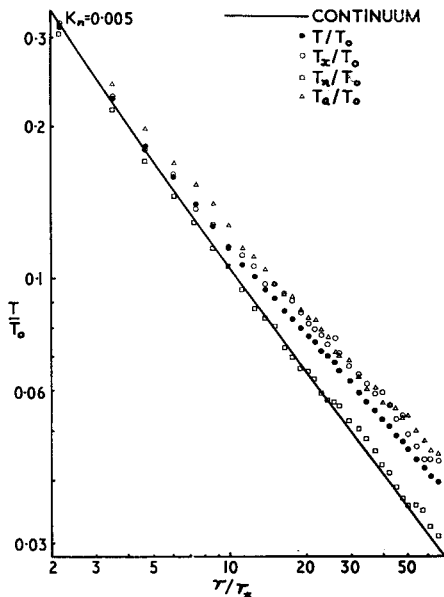


Fig. 6 Initial breakdown in a cylindrical expansion of a hard sphere gas.

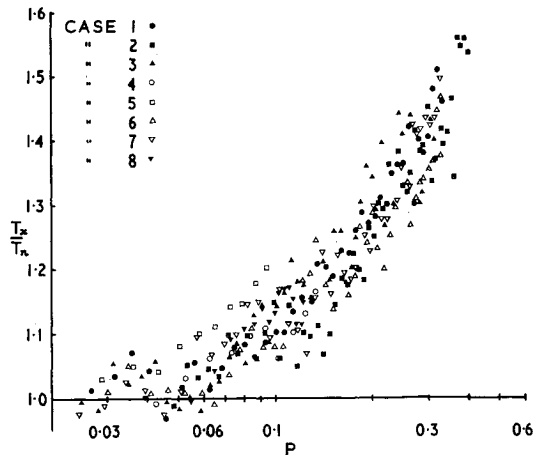


Fig. 7 Correlation of results by the parameter P .

Analysis of Steady Expansion

The establishment of a criterion for the onset of non-equilibrium requires the determination of a parameter which correlates all the results. The simplest parameter is probably the ratio of the magnitude of the logarithmic time derivative of density following the motion of an element of the fluid to the collision frequency in the element. That is,

$$P = 1/\nu |D(\ln \rho)/Dt| \quad (1)$$

For a one-dimensional steady flow

$$P = (u/\rho\nu) |d\rho/dx| \quad (2)$$

But

$$\nu = v_s/\lambda$$

so that

$$P = (\lambda u/v_s \rho) |d\rho/dx| \quad (3)$$

This form of P may be interpreted as the Lagrangian mean free path divided by the density scale length and is therefore consistent with the parameter proposed by Scott and Phipps.¹²

A mean free path may be defined for all gases by assuming the universal application of the hard sphere relation

$$\lambda = (16/5)(\mu/\rho) 1/(2\pi RT)^{1/2}$$

The effect of molecular model may be introduced by assuming that the coefficient of viscosity is proportional to temperature

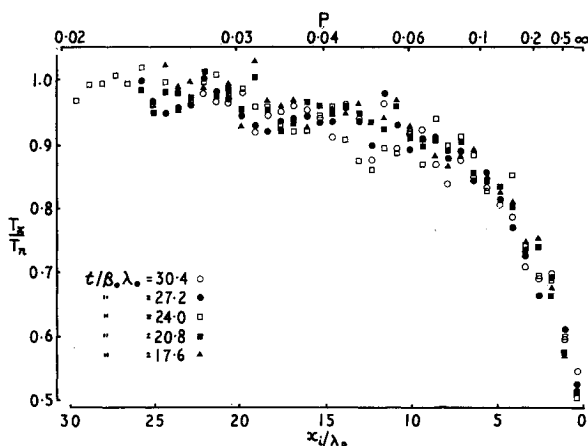


Fig. 8 Breakdown of translational equilibrium in a complete rarefaction wave in a hard sphere gas.

raised to the power ω . Hard sphere molecules correspond to $\omega = \frac{1}{2}$ and Maxwell molecules to $\omega = 1$.

Considering a steady isentropic expansion with flow area proportional to distance raised to the power n , it may be shown that,

$$P = \frac{\lambda_0}{r_*} \frac{1}{2} \left(\frac{\pi\gamma}{2} \right)^{1/2} \frac{nM^{3+1/n}}{M^2 - 1} \left(\frac{\gamma + 1}{2} \right)^{[(\gamma+1)/2n(\gamma-1)]} \times \left(1 + \frac{\gamma-1}{2} M^2 \right)^{[(\gamma+1)/2(\gamma-1)][1-(1/n)]-\omega} \quad (4)$$

The ratio of parallel to normal temperature for all cases listed in Table 1 is plotted against P in Fig. 7. The result provides very strong support for P as the correlating parameter. Considerable scatter has to be accepted in the ratio of two temperatures, but a close examination shows that, with the exception of case 5, all the results are consistent with a single curve. The points for case 5 all lie near the top of the band and this may have been caused by the upstream boundary being set at a radius where the flow was already slightly out of equilibrium. The only justification for the choice of density as the macroscopic property in Eq. (1) is that the density is the zero-order moment of the velocity distribution and leads to the simplest parameter. The use of temperature rather than density would introduce an additional factor of $1/(\gamma-1)$ which would significantly reduce the correlation between the gas with $\gamma = \frac{5}{3}$ in cases 1-5 and the gas with $\gamma = \frac{4}{3}$ in cases 6 and 7. A factor of $\gamma/(\gamma-1)$ would be introduced by the use of pressure and the reduction in the correlation would be almost as serious. The use of velocity or Mach number would introduce factors of M^2 and $M^2/[1 + M^2(\gamma-1)/2]$, respectively, and the correlation would be drastically affected.

The general behavior of P with Mach number is that it decreases from $M = 1$ to M' , where M' is the positive real solution of

$$M^4 \left[\frac{\gamma+1}{2} \left(1 - \frac{1}{n} \right) - (\gamma-1)\omega + \frac{\gamma-1}{2} \left(1 + \frac{1}{n} \right) \right] - M^2 \left[\frac{\gamma+1}{2} \left(1 - \frac{1}{n} \right) - (\gamma-1)\omega + 2 - \frac{3-\gamma}{2} \left(3 + \frac{1}{n} \right) \right] - 3 - \frac{1}{n} = 0 \quad (5)$$

P then increases with M and, for very high Mach numbers,

$$P \propto M^k \quad (6)$$

where

$$k = 1 + 1/n + [(\gamma+1)/(\gamma-1)](1 - 1/n) - 2\omega$$

M' and k are listed for a number of cases of interest in Table 2.

The fact that P tends to infinity at $M = 1$ for any finite value of n is of no significance. The usual choking condition for an isentropic expansion requires that $n = 0$ at $M = 1$. Any real expansion will have effective values of n which vary both along and normal to the streamlines. However, the high Mach number regions of axisymmetric and plane freejets are well represented by spherical ($n = 2$) and cylindrical ($n = 1$) flows, respectively. If a flow is in equilibrium at any Mach

Table 2 Behavior of P

Flow geometry	Molecular type	M'	k
Spherical	Hard sphere	1.431	2.5
Spherical	Maxwellian	1.525	1.5
Spherical	Diatomic ($\omega = 0.75$)	1.425	3
Spherical	Rough sphere	1.396	4
Cylindrical	Hard sphere	1.508	1
Cylindrical	Maxwellian	1.731	0

number below M' , departure from equilibrium will not occur until the Mach number has increased beyond M' . Conversely, if the throat Knudsen number is such that the flow is out of equilibrium at $M = M'$ it will be out of equilibrium at all Mach numbers below this, as long as n remains constant.

For breakdown occurring in a hypersonic flow at a constant value of P , the breakdown Mach number will be equal to a constant multiplied by the throat Knudsen number to the power $-1/k$. This power is equal to -0.4 for the spherical expansion of a hard sphere gas and -0.6667 for a Maxwell gas. This agrees with the behavior of the freezing and breakdown Mach numbers in the BGK analysis of Hamel and Willis.⁵ Anderson and Fenn¹ obtained the index -0.4 for the freeze Mach number in an argon freejet. However, as pointed out by Scott and Phipps,¹² a parameter based on the near-continuum properties of the freejet may not be valid in the extreme nonequilibrium region and the agreement with the hard sphere index is probably fortuitous.

Figure 7 indicates that the value of P for the breakdown of equilibrium is approximately 0.04. The most useful relationship for engineering purposes is obtained if the hard sphere formula

$$\nu = (5/4)\rho RT/\mu$$

is substituted into Eq. (2) to give

$$P = (4\gamma/5)M^2/Re_L \quad (7)$$

Therefore, the empirical criterion for breakdown becomes

$$\gamma M^2/Re_L = 0.05 \quad (8)$$

with the Reynolds number based on the density scale length.

Rarefaction Wave

The parameter P may be applied to unsteady as well as steady expansions. Consider a complete rarefaction wave with the gas moving in the negative x direction. At any point x, t in the wave

$$x/t = u + a \quad (9)$$

and

$$[2/(\gamma - 1)]a - u = [2/(\gamma - 1)]a_0 \quad (10)$$

Since the flow is isentropic,

$$\rho \propto a^{2/(\gamma-1)} \quad (11)$$

Equations (9-11) may be substituted into Eq. (1) to give

$$P = [2/(\gamma + 1)]1/\nu t = [2/(\gamma + 1)]\lambda/v_s t \quad (12)$$

This may be evaluated at the leading edge of the expansion to give the initial value of P as

$$P_i = [1/(\gamma + 1)](\pi\gamma/2)^{1/2}(\lambda_0/x_i) \quad (13)$$

for the element of fluid entering the expansion at $x = x_i$. Equations (9-12) show that the rate of change of P along the particle path is

$$DP/Dt = (P/t)[(\gamma - 1)/(\gamma + 1)](1 - 2\omega) \quad (14)$$

Therefore, for a hard sphere gas, the parameter P remains constant along a particle path. This means that if a fluid

element remains in equilibrium when it first enters the expansion, it should remain in equilibrium at all subsequent times. Alternatively, if the element goes out of equilibrium in the very early part of the expansion, it would be expected to remain in much the same state as it progresses along a particle path.

The key results from the computation of a complete rarefaction in a hard sphere gas are shown in Fig. 8. The ratio of the kinetic temperatures based on the parallel and normal components of molecular velocity is plotted against the initial position of the fluid element for a number of values of time. There is no significant difference between the curves for the various times. This is exactly as expected from the previous analysis and discussion. The value of the parameter P is also shown in Fig. 8 and its value at the initial breakdown of equilibrium is consistent with the value obtained for the steady expansion. This gives further support for P as the correlating parameter.

For "softer" molecules with $\omega > \frac{1}{2}$, Eq. (14) indicates that P decreases along a particle path so that a "progressive freeze" will never occur. On the contrary, there may be a tendency for fluid elements that go out of equilibrium in the early part of the expansion to move back towards equilibrium.

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