Translational Nonequilibrium in a Rapid Expansion Flow from a High Temperature Source

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(February 1, 1983)

Summary: Terminal parallel temperature of a rapid expansion flow into a vacuum was examined numerically and experimentally. It was shown that for high temperature flow, inclusion of a more realistic intermolecular potential with the repulsive term gives a more accurate prediction of the translational relaxation during the expansion. In a temperature range, 7000–10000 K, the measured terminal parallel temperature in an expansion flow of shock heated argon agreed very well with these predicted values.

1. Introduction

During a rapid expansion into a vacuum, the gas molecules undergo a drastic change from a continuum behavior to a free molecular one with energy relaxations between the internal and external modes. For a monoatomic gas the translational nonequilibrium occurs, i.e. the velocity distributions of the molecules in the direction parallel to the flow direction and those in the direction perpendicular to it, or the temperatures of both directions characterizing those velocity distributions, become different downstream in the flow. The velocity distributions in the perpendicular direction may continue to become narrow due to the geometrical effect, while those in the parallel direction freeze to a certain value, since the relaxation of this direction occurs only by molecular collisions. This phenomenon has been studied by many authors [1] and the terminal (or freezing) parallel temperature observed in a flow from a room temperature source can be well predicted by a gas-kinetic treatment [2]. However, for a light gas like helium the observed terminal parallel temperature source is much lower than the predictions using the classical collision cross sections [3]. Toennies and Winkelmann [4] explained this as a quantum effect on the collision cross section, using the moment method of the Boltzmann equation. Therefore, the terminal parallel temperature is very sensitive to the collision cross section.

In this report we have examined the effect of the intermolecular potential on the terminal parallel temperature in an expansion flow of high temperature argon, in which the repulsive part of the potential may also be significant, while only the attractive term is dominant in a room temperature argon jet. Measurements of the parallel temperature of argon expanded from the reflected region of a shock tube were made and the results were compared with the numerical calculations including several potential parameters.

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2. Numerical Method

A numerical calculation was made after Toennies and Winkelmann's method assuming the flow as a source flow. We also assumed that the flow Mach number M is given by [5]

$$M=3.26\left(\frac{x}{d}-0.075\right)^{2/3}-0.613\left(\frac{x}{d}-0.075\right)^{-2/3}, \tag{1}$$

where x is the distance from the nozzle orifice of diameter d, and that the flow is isentropic near the orifice. From a certain value of x/d the flow is governed by the gas-kinetic equations, i.e., the gas molecules which have an ellipsoidal velocity distribution f.

$$f=n\left(\frac{m}{2\pi kT_{//}}\right)^{1/2}\left(\frac{m}{2\pi kT_{\perp}}\right)\exp\left[-\frac{m}{2kT_{//}}(v_{//}-u)^2-\frac{m}{2kT_{\perp}}v_{\perp}^2\right], \tag{2}$$

are governed by the Boltzmann equation in spherical coordinates:

$$\frac{df}{dr}(v_{//}f)-\frac{m}{kT_{//}}\frac{1}{r}(v_{//}-u)v_{\perp}^2f+\frac{m}{kT_{\perp}}\frac{v_{//}v_{\perp}^2}{r}f=\left(\frac{\partial f}{\partial t}\right)_{\text{coll}}, \tag{3}$$

where n is the number density of molecules, m the molecular mass, k the Boltzmann constant, u the flow velocity, $v_{//}$, $v_{\perp}$, $T_{//}$, $T_{\perp}$ are the velocities and the temperatures parallel and perpendicular to the flow direction r, respectively. Taking the moments of m, $mv_{//}$, $m(v_{//}^2+v_{\perp}^2)/2$ and $mv_{\perp}^2/2$ in connection with eq. (3) yields the governing equations of the flow. By using the reduced parameters,

$$r^*=\frac{r}{d},\quad n^*=\frac{n}{n_0},\quad T_{//}^*=\frac{T_{//}}{T_0},\quad T_{\perp}^*=\frac{T_{\perp}}{T_0},\quad u^*=\frac{u}{u_{\max}}, \tag{4}$$

where T_0 is the stagnation temperature, n_0 the stagnation number density and $u_{\max}$ the attainable maximum flow velocity ($=\sqrt{5kT_0/m}$), these equations can be written in nondimensional forms:

$$n^*u^*r^{*2}=\text{const}, \tag{5}$$

$$u^{*2}=1-\frac{3}{5}T_{//}^*-\frac{2}{5}T_{\perp}^*, \tag{6}$$

$$\frac{dT_{\perp}^*}{dr^*}=-\frac{2T_{\perp}^*}{r^*}+F, \tag{7}$$

$$\frac{dT_{//}^*}{dr^*}=-\left(\frac{1}{2}-\frac{3}{10}\frac{T_{//}^*}{u^{*2}}\right)^{-1}\left\{F\left(1-\frac{1}{5}\frac{T_{//}^*}{u^{*2}}\right)+\frac{2}{5}\frac{T_{//}^*}{u^{*2}}\frac{T_{\perp}^*}{r^*}\right\}, \tag{8}$$

where F is the term resulted from the collision effect between the two translational modes and is written as

$$F=2\left(\frac{\pi}{5}\right)^{1/2}\frac{p_0d}{kT_0}\left(\frac{n^*}{u^*}\right)\left(\frac{T_{\perp}^{*2}}{T_{//}^{*1/2}}\right)\int_0^1\frac{3\xi^2-1}{(1-a\xi^2)^3}\frac{1}{\pi}\left(\frac{2\pi\mu}{kT_{\text{eff}}}\right)^{1/2}\Omega^{(2,1)}(T_{\text{eff}})d\xi, \tag{9}$$

where p_0 is the stagnation pressure and

$$T_{\text{eff}}=\frac{T_{\perp}}{1-a\xi^2}, \qquad a=\frac{T_{//}-T_{\perp}}{T_{//}}. \tag{10}$$

The collision integral $\Omega^{(2,1)}(T_{\text{eff}})$ can be written as

$$\Omega^{(2,1)}(T_{\text{eff}})=\left(\frac{kT_{\text{eff}}}{2\pi\mu}\right)^{1/2}\int_0^{\infty} Q^{(2)}(g)\gamma^5 e^{-\gamma^2}d\gamma \tag{11}$$

where g is the relative collision velocity, μ the reduced mass, γ^2 the reduced collision energy ($=\mu g^2/2kT_{\text{eff}}$) and $Q^{(2)}(g)$ is the viscosity cross section.

As decreasing the density in the flow the term F tends to zero, but the perpendicular temperature still decreases by the geometrical term $-T_{\perp}^*/r^*$. The parallel temperature, on the other hand, tends to freeze in a certain value as F tends to zero because $T_{//}^*/u^{*2}\ll 1$ at large r/d. This freezing value of the parallel temperature depends on the collision integral as well as the stagnation condition. Although the collision integral can be reduced in a simple form by assuming a hard sphere or a Maxwell molecule, we take more realistic interaction potential in order to evaluate the effect of the potential on the translational nonequilibrium in a flow from a high temperature source.

In Fig. 1 the reduced values of the viscosity cross sections by that of a rigid sphere are plotted as a function of the collision energy K reduced by the potential depth ε for several potentials. For (12-6) Lennard-Jones potential we used the formula [6],

$$Q^{(2)*}=\sum_{i=1} A_i K^{n_i}\exp(-a_i K). \tag{12}$$

For low collision energy as in the expansion from a room temperature source, the cross section can be well approximated only by the R^{-6} attractive term and $Q^{(2)}$ can be represented by [7]

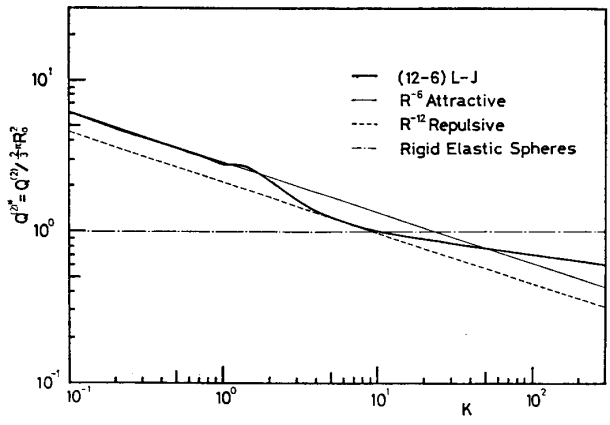


Fig. 1. Reduced viscosity cross sections for different potentials.

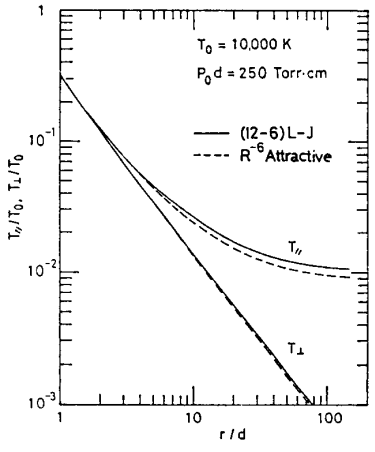


Fig. 2. Temperature change during the expansion flow of high temperature argon along stream line.

$$Q^{(2)} \cong 0.331 \cdot 2\pi \left(\frac{6C_6}{\frac{1}{2}\mu g^2} \right)^{1/3}, \tag{13}$$

where $C_6 = 4R_0^6\varepsilon$ and R_0 is the nuclear distance at zero potential energy. However, in a flow from a high temperature source, contributions of the cross section to the collision integral in an energy range, K=1–10, where the cross section is dominated by the repulsive term, will be significant.

The differential equations (7) and (8) were integrated by RKG method using FACOM M-382 computer at Data Processing Center, Kyoto University. Typical calculated results of the temperature change along the stream line are shown in Fig. 2, for two viscosity cross sections, eqs. (12) and (13). The effect of the cross section on the perpendicular temperature is small, but yields an appreciable difference in the parallel temperature.

3. Experimental Method

Continuous heating of a gas up to 3000 K is possible, but above this to 10000 K, shock heating is a convenient and well established technique, although its duration time is only order of 1 msec. We have already shown by the molecular beam time-of-flight (TOF) method that a measured effective stagnation temperature of the shock heated gas in the reflected region coincides with one calculated from the Rankine-Hugoniot relation up to 10000 K, where the ionization relaxation time is still longer than the duration time.

In the previous experiments, the expansion was made through a long nozzle and the beam flight chamber was a part of the expansion chamber separated by a partition plate, therefore the pressure rise by the flow of the gas from the shock tube

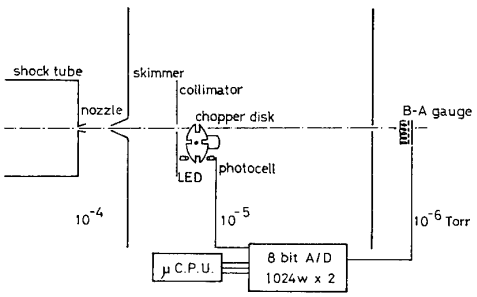


Fig. 3. Schematic diagram of apparatus.

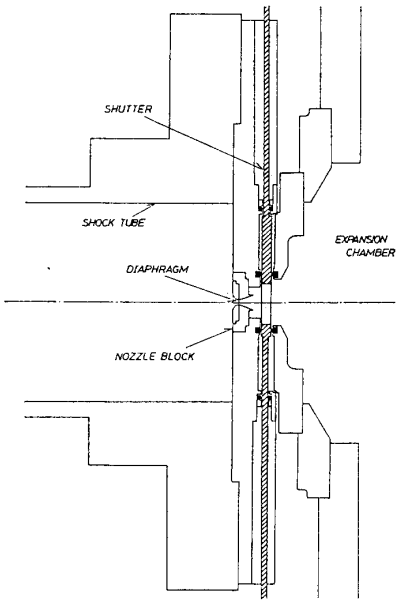


Fig. 4. Details of nozzle and shutter mechanism.

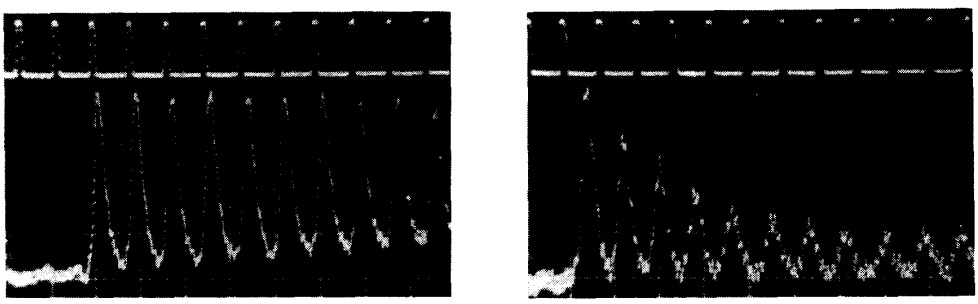
limited the test time and caused some contamination of the beam detector system. In the present apparatus an additional expansion chamber was connected to the shock tube, so that the main flight chamber can be evacuated separately from the former. A schematic diagram of the present apparatus is shown in Fig. 3. The shock tube has a 128 mm inner diameter and a 4.7 m length. At the end of the tube a small nozzle, which has a 3.2 mm throat diameter and 8 mm length with a 60° divergent section, was attached as shown in Fig. 4. Due to the geometrical reason we could not attach a free-jet-type orifice.

The expansion chamber was evacuated by a 4 inch oil diffusion pump to a pressure of 10^{-4} Torr. A skimmer with 3 mm orifice diameter was placed at 275 mm downstream of the nozzle throat. The flight chamber and the detector chamber were evacuated by 10 and 2 inch oil diffusion pumps to pressures of 10^{-5} and 10^{-6} Torr, respectively. Before firing the shock tube a thin polyester diaphragm slightly upstream of the throat separated the expansion chamber from the shock tube in order to prevent from flowing the gas into it. After passing the shock heated gas a mechanical shutter just downstream of the nozzle was closed. It took about 30 msec after firing the shock tube and the pressure rises in the expansion, the flight and the detector chambers were 0.1, 10^{-4} and 10^{-5} Torr, respectively, in the present experimental conditions.

A time-of-flight system had a slit function of 28 μsec half width, a flight path of 135 cm and a 15 mm detector width. The molecules were detected by a Bayard-Alpart ion gauge. The measurements were made for the expansion flow of the shock heated argon with 530–2750 Torr stagnation pressures and 3640–10800 K stagnation temperatures. The measured TOF signals together with photocell signals were recorded by a storage-type oscilloscope (NF-WM852) and stored into a microcomputer (Sord M223 mark V), with which a simulated TOF signal fitted to the measured one was generated in order to obtain the mean flow velocity and the parallel temperature.

4. Results and Discussions

Typical records of TOF signals are shown in Fig. 5 as the lower traces. They correspond to series of the photocell signals in the upper traces. For a high tem-



(a) (b)

Fig. 5. Typical records of TOF signals.
(a) P_0=1040 Torr, T_0=9150 K, $T_{//}$=130 K, u=3.00×10^5 cm/sec.
(b) P_0=1780 Torr, T_0=4000 K, $T_{//}$=38.0 K, u=2.02×10^5 cm/sec.

perature stagnation condition the beam signal was strong and almost constant during the test time as shown in Fig. 5 (a), but for a low stagnation temperature or a high stagnation density condition the beam intensity decreased strongly during the test time as shown in Fig. 5 (b) by the reason described below.

Assuming an energy balance during the expansion, we can determine an effective stagnation temperature T_{eff} from the measured values of the flow velocity and the parallel temperature [8]. This measured effective stagnation temperature is compared with the Rankine-Hugoniot value calculated from the measured shock velocity, in Fig. 6. As in the previous experiment, a very good agreement between them was obtained for a wide range of the stagnation condition. Reduction in the effective stagnation temperature due to the thermal boundary layer effect with small diameter orifices [9] are also shown in the figure and no appreciable effect of this is not seen with the present nozzle. Within the measured time (1–2 msec) the ionization relaxation is not completed even for the highest stagnation temperature case and then no appreciable effect of the ionization is seen.

In Fig. 7 the measured parallel temperatures are plotted as a function of the scaling parameter λ,

$$\lambda = p_0 d \varepsilon^{1/3} R_m^2 T_0^{-4/3} (\text{Torr} \cdot \text{cm}\ meV^{1/3} A^2 K^{-4/3}), \tag{14}$$

which is a measure of the collision effect on the translational relaxation reduced using the attractive cross section eq. (13), where R_m is the nuclear distance at the potential minimum. The calculated frozen parallel temperatures for (12–6) Lennard-Jones and R^{-6} attractive potentials are shown in the figure. The parallel temperature decreases with increasing λ but it becomes almost unchanged for $\lambda > 0.05$. This value of λ corresponds to the stagnation condition where the beam intensity decrease during the test time become strong as shown in Fig. 5 (b). For a small value of

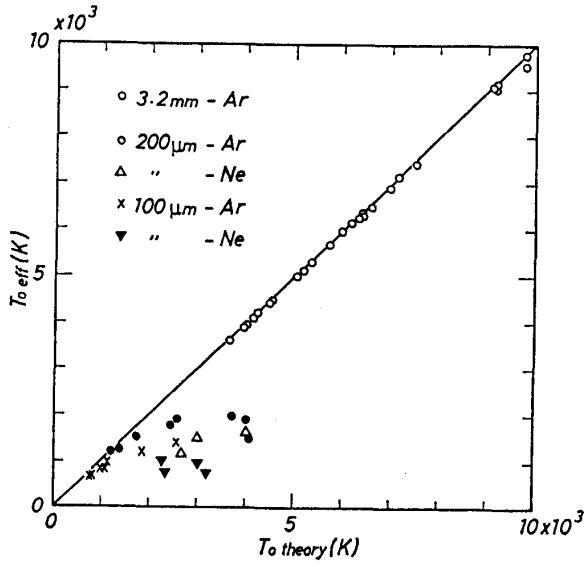


Fig. 6. Effective stagnation temperature compared with Rankine-Hugoniot value.

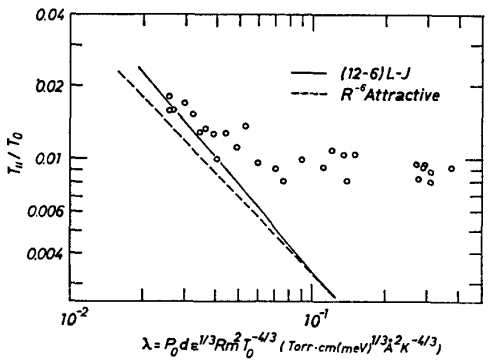


Fig. 7. Measured terminal parallel temperature plotted as a function of scaling parameter λ in comparison with numerical results.

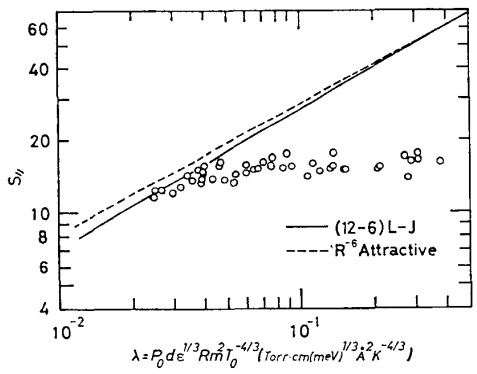


Fig. 8. Measured terminal speed ratio plotted as a function of scaling parameter λ in comparison with numerical results.

λ which corresponds to a stagnation pressure less than 640 Torr and a stagnation temperature higher than 7000 K, the flow density at the skimmer is low enough to neglect the skimmer interaction, which reduces the beam intensity. For a stagnation condition corresponds to a larger value of λ than 0.05 the skimmer interaction may become significant and broadens the velocity distributions of the flow as well as reduces the beam intensity. Therefore, in the present arrangement the terminal parallel temperature of the flow could not be measured for $\lambda > 0.05$. But for room temperature argon of this range of λ the measured parallel temperatures agree very well with the theoretical prediction. In Fig. 8 the speed ratio $S_{//}$ calculated from the measured velocity and the terminal parallel temperature is plotted against λ together with the theoretical predictions. It can be seen that for $\lambda < 0.05$ it agrees well with the predictions and that for smaller values of λ or higher stagnation temperatures the effect of the repulsive term on the terminal temperature (or speed ratio) becomes significant.

Although the experiments are still in progress, it can be concluded that the terminal parallel temperature of a rapid expansion flow from a high temperature source can be well predicted by a simple kinetic treatment with a real interaction potential.

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