Studies of Translational Freezing in Free Expanding Jets Using Molecular Beam Techniques

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Experimental values for the perpendicular temperature in freely expanding jets are deduced from beam intensity measurements as a function of the skimmer diameter over a wide range of reduced distances ($40 < x/d < 300$) for high inverse source Knudsen numbers ($500 < Kn^{-1} < 6500$). The gases used are Ar, N_2, CO_2. The $T_\perp$ values obtained are well represented by the equation $T_\perp \sim (x/d)^{-m}$ where m depends on the gas and on the source Knudsen number and is intermediate between the values for isoentropic and molecular flow. Our measurements indicate that the flow is still in the transition region at rather high distances ($x/d \sim 300$) from the source. The assumptions made in deriving the ideal molecular beam equation (ideal skimming, negligible self-scattering upstream the skimmer, ellipsoidal velocity distribution) are discussed.

Introduction

IN the recent years the free jet expansion has been the object of several studies of rarefied gas dynamics. From the theoretical point of view the solution of the problem is equivalent to the solution of the Boltzmann equation with simple and well defined boundary conditions but, due to the different flow conditions along a single flow line, a satisfactory solution has been only recently reached[1-4] for monoatomic gases. The velocity distribution function is usually separated into two factors for the two degrees of freedom parallel and perpendicular to a flow line.

The problem of the temperature $T_{||}$ corresponding to the distribution of the velocities parallel to the flow, seems to have been satisfactorily solved. For monoatomic gases theory predicts a decrease of $T_{||}$ proportional to $(x/d)^{-4/3}$ where x is the distance from the nozzle and d the nozzle diameter. At high reduced distances $T_{||}$ becomes independent of distance reaching a value of $T_{||}^{\infty}$ that depends in both the source Knudsen number and the intermolecular interaction potential. The theoretical predictions are in agreement with experimental determinations of $T_{||}$ performed in the time of flight[5-7] and the electron fluorescence[8] techniques.

On the other hand a satisfactory solution for the problem of $T_\perp$ has not been found. According to theory $T_\perp$ should follow the isentropic expansion law $[T_\perp \sim (x/d)^{-4/3}]$ up to large values of x/d then, well after $T_{||}$ has reached its limiting value, the dependence should become $T_\perp \sim (x/d)^{-1}$.[1] It appears that the flow should not become free molecular $[T_\perp \sim (x/d)^{-2}]$ at any distance from the nozzle.

Recently other authors[2] have found a solution on the Boltzmann equation for the perpendicular velocity distribution function as a sum of two contributions. The first is due to molecules which have not taken part in a collision and the second is due to the molecules which have undergone a collision and have perpendicular temperature proportional to $(x/d)^{-1}$. The total $T_\perp$ is determined by this second class of molecules with great transverse velocity, i.d. by the tails of the distribution function in the transverse velocity. Similar results were obtained by other authors.[3-4]

The experimental results reported in the literature on this subject[7-10] are limited and very often obtained at relatively low inverse source Knudsen number and over a limited range of x/d values. It appears that these results are not in agreement with the theory. In particular, the asymptotic dependence $T_\perp \sim (x/d)^{-1}$ has never been found. We note that this dependence is determined by the tails of the distribution of the transverse velocity, while the experimental data are obtained near to the center of the distribution.

This work is an extension of a work previously published[11] which will be referred to as I. In I it has been shown how the problem of high intensity supersonic beam production can be solved by the use of small cryopumps, with a low liquid helium consumption, in the utilization chamber. Moreover, due to the high sticking coefficient of gases like CO_2 the use of directional cryopumps maintains the vacuum in the utilization chamber almost independent of the gas flow through the skimmer. It is, thus, possible to measure beam intensities with skimmers of different diameter in almost ideal conditions, i.e., with no background scattering, hence obtaining information on $T_\perp$.

Nevertheless the measurements reported in I were taken over limited pressure and nozzle-skimmer distance ranges. In addition because of the poorer cryopumps performances with Argon, the $T_\perp$ values for this gas were reported for a too small x/d range which prevented any serious comparison with theory.

The aim of the present work is to obtain new experimental results for the dependence of $T_\perp$ on x/d for Ar and N_2 over a wide range of x/d values $[40 < x/d < 300]$ for high values of the inverse source Knudsen number $[500 < Kn^{-1} < 6500]$. The experimental results for CO_2 were reported previously.[12] We will also discuss assumptions made in the theory of beam formation and show how it is possible to measure the perpendicular temperature from molecular beam intensity measurements with different skimmer diameters. The study of the dependence of $T_\perp$ on x/d is essential for understanding the transition flow region in the free jet expansion of pure gases and mixtures.[13-14]

Beam Formation

A molecular beam is obtained by separating the central core of the free jet expansion of a gas with a collimator of the proper shape, called the skimmer.

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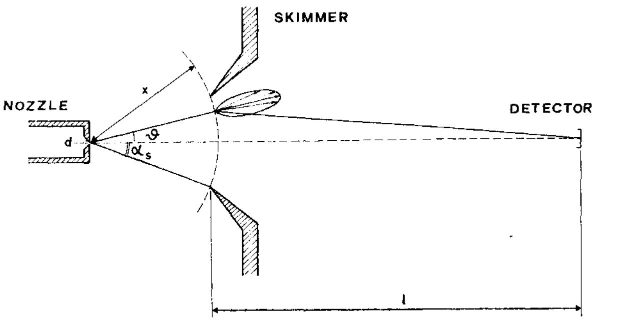

Fig. 1 Elementary contribution to the beam intensity for collisionless skimmer.

Almost every beam formation theory is founded on two hypotheses: 1) the skimmer does not interfere with the flow; and 2) the gas density after the skimmer is sufficiently low that the flow after the skimmer is collision free. The central problem of beam formation is then the determination of the expansion conditions at the skimmer opening. In an elementary theory one assumes that the flow is parallel to the skimmer axis. In the case of hypersonic flow, the intensity for unit solid angle is

$$I = (A_s n_s/\pi)u(3/2 + S^2) \tag{1}$$

where

A_s = skimmer area
n_s = number density at the skimmer
S = speed ratio at the skimmer defined by $S = u/(2KT/m)^{1/2}$
u = bulk velocity at the skimmer

and

T = temperature at the skimmer
K = Boltzmann constant
m = mass of a beam particle

Hagena and Morton,[15] among others, introduced the flow divergence, and found the following expression for the beam intensity

$$I = (1 - \cos^3\alpha_s \exp(-S^2 \sin^2\alpha_s))n_s u x^2 \tag{2}$$

where α_s is the semiangle which the skimmer opening subtends at the nozzle.

Equations (1) and (2) were obtained under the assumption that the speed ratio is isotropic at the skimmer. At high (x/d) values this assumption fails and the simplest method to allow for this is to suppose the existence of a last collision surface situated at a distance from the nozzle x_F which will depend on the stagnation conditions.[16–17]

A more general treatment has been made by Le Roy et al.[18] Following Hamel and Willis, they assume the velocity distribution function to be ellipsoidal with two different temperatures for the parallel and perpendicular degrees of freedom $(T_{||},T_\perp)$

$$f(v,\vartheta)d\Omega dv = \left(\frac{m}{2\pi KT_\perp}\right)\left(\frac{m}{2\pi KT_{||}}\right)^{1/2} e^{-(mv^2\sin^2\vartheta/2KT_\perp)} \times e^{-[(v\cos\vartheta - u)^2 m/2KT_{||}]}\, v^2 dv d\Omega \tag{3}$$

where $d\Omega$ is the element of solid angle measured from the center of the skimmer opening.

The flow of particles toward the detector with velocity between v and $v + dv$ leaving the portion of spherical surface dA delimitated by ϑ and $\vartheta + d\vartheta$ is, per unit solid angle

$$dI_{dA} = n_s(\vartheta)v \cos\vartheta f(v,\vartheta)dvdA \tag{4}$$

where ϑ is defined in Fig. 1.

Integrating over the velocities, in the range of validity of the hypersonic approximation ($S_{||} \geqslant 3$) and taking $tg\vartheta \simeq \vartheta$ we obtain

$$I_{dA} = 2x^2 n_s e^{-S_{||}^2[1 - 1/(1+t^2)]} u S_\perp^2[1 + 3/2S_{||}^2(1 + t^2)] \times (1 + t^2)^{-7/2}\vartheta d\vartheta \tag{5}$$

with $t = (S_\perp/S_{||})\vartheta$. For $t^2 \ll 1$ Eq. (5) can be integrated straightforwardly to give:

$$I = I_0(1 - e^{-S_\perp^2\alpha_s^2}) \tag{6}$$

where $I_0 = n_s u x^2(1 + 3/2S_{||}^2)$. Equation (6) is consistent with Eq. (2) if one considers the speed ratio isotropic at the skimmer. Therefore, we note that it is implicit in the analysis of Hagena and Morton that $S_\perp$ is the essential quantity for the beam intensity.

The previous formulas were obtained for a detector of small aperture situated at large distances from the skimmer ($l \gg x$). In the case of finite distance l the Eq. (6) must be changed as follows[18]†

$$I = I_0/(1 + x/l)^2(1 - e^{-S_\perp\alpha_s^2(1+x/l)^2}) \tag{7}$$

In Ref. 18 the intensity calculated by Eq. (5) with fixed values of $S_\perp$ and $S_{||}$ are compared with the intensity calculated from Eq. (2) with the same $S_\perp$ value. The agreement between the intensities calculated with the two equations is good, differences of few percent were obtained only for small skimmer diameters. As a consequence we see that beam intensity measurements as a function of α_s will give information on the perpendicular temperature in the expansion.

Experimental

The apparatus is shown schematically in Fig. 2. Relevant dimensions are given in Table 1. More details are given in I. The nozzle diameter is quite small in order to have small Knudsen numbers and yet reasonable stagnation pressures in the expansion chamber. This chamber is evacuated by an unbaffled booster-diffusion pump of 10.000 l/sec^{-1} nominal pumping speed. The central core of the expansion goes through the skimmer into the utilization chamber where it condenses on a liquid helium cooled directional cryopump.[19] A small 350 l/sec diffusion pump evacuates the incondensable gases. A typical background pressure in the utilization

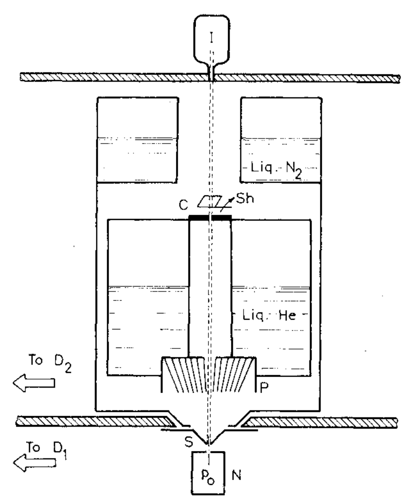

Fig. 2 Schematical view of the beam production and detection apparatus.

I Ionization gauge detector.
Sh Shutter.
C Collimator.
P Cryopump.
S Skimmer.
N Nozzle.
D_1 10'000 l sec^{-1} booster diffusion pump.
D_2 350 l sec^{-1} diffusion pump.

† We thank R. L. Le Roy for bringing this point to our attention.

Table 1 Relevant dimensions of the beam production apparatus

Nozzle diameter	0.085 mm
Nozzle geometry	Simple converging conical
Skimmer diameter	a) 0.27 mm
	b) 0.40 mm
	c) 0.49 mm
	d) 0.62 mm
	e) 0.75 mm
	f) 1.21 mm
	g) 1.49 mm
	h) 1.80 mm
Skimmer geometry	40° internal semiangle
	45° external semiangle
Nozzle-skimmer distance	2–28 mm
Skimmer-collimator distance	185 mm
Collimator diameter	1.03 mm

chamber is 2 10^{-7} torr, this is quite independent of the flow through the skimmer. Beam intensities are measured by a storage detector which uses a Bayard-Alpert ionization manometer and whose entrance channel conductance has been calculated taking in account the Clausing correction.[20]

In Figs. 3 and 4 we show a few typical measurements of the intensity as a function of the nozzle-skimmer distance with different stagnation pressures. The difference between the shape of these plots for CO_2 and for Ar has been previously discussed in I. In the present work the Ar plots show less pronounced minima as a consequence of a better cryopump design.

The experimental results have to be corrected, taking into account the scattering from the background in the expansion chamber. Effective scattering cross sections have been determined experimentally for every skimmer by measuring beam intensity as a function of the background pressure P_1, and the results are in agreement with previous determinations.[21–22] An example of such a determination is given in Fig. 5. Corrected intensity measurements as a function of skimmer diameter for CO_2 and Ar are reported in Figs. 6 and 7, respectively.

Discussion of the Fitting Procedure

The shape of the plots shown in Figs. 6 and 7 is in contrast with the predictions of Eq. (7). No saturation of the intensity is reached at the limit of large skimmer diameters. Therefore, we have fitted our results to the equation

$$I = A(1 - e^{-S_{\perp}^2\alpha_s^2(1+x/l)^2}) + B\pi r_s^2 \qquad (8)$$

obtained by adding a term proportional to the skimmer area by Eq. (7). That is equivalent to postulating the existence of a second contribution to the intensity, from molecules which have a velocity distribution with a very low $S_{\perp}$ values.

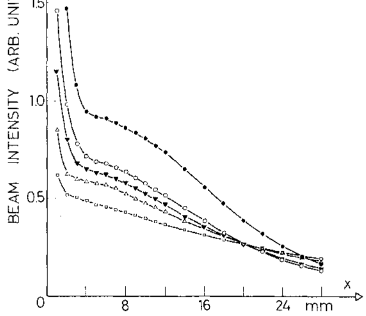

Fig. 3 Beam intensity vs nozzle-skimmer separation. Gas CO_2, skimmer diameter 1.21 mm; □ = 340 mm Hg, △ = 600 mm Hg, ▼ = 1000 mm Hg, ○ = 1400 mm Hg, ● = 2300 mm Hg.

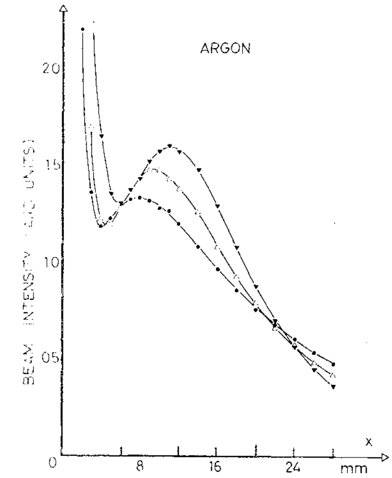

Fig. 4 Beam intensity vs nozzle-skimmer separation. Gas Ar, skimmer diameter 1.21 mm; ● = 800 mm Hg, △ = 1300 mm Hg, ▼ = 2500 mm Hg.

The following equation has also been used to fit the results

$$I = A'(1 - e^{-S_{\perp 1}^2\alpha_s^2(1+x/l)^2}) + B'(1 - e^{-S_{\perp 2}^2\alpha_s^2(1+x/l)^2} \qquad (9)$$

The best fit obtained in this case produced very low values of $S_{\perp 2}$ and completely justified the use of Eq. (8).

The fits were made using the least squares method for the experimental points obtained with seven different skimmers of diameters 0.27; 0.40; 0.49; 0.62; 0.75; 1.21 and 1.49 mm.

A fit performed with also the data for the 1.80 mm diameter skimmer produced no significant deviations at large x values and deviations of few per cent at small x values where the condition $\alpha_s^2 S_{\perp}^2/S_{\parallel}^2 \ll 1$ fails. In Figs. 8 and 9 we show the results obtained for Ar and N_2, respectively. The general behavior of $T_{\perp}$ is rather similar for the gases measured up to now. This is consistent with the fact that after few nozzle diameter in the expansion the rotational temperature freezes out.[23–24]

The dependence of $T_{\perp}$ on x/d is well described by the equation

$$T_{\perp} \sim (x/d)^{-m} \qquad (10)$$

where m is a decreasing function of the inverse Knudsen number and depends also on the nature of the gas.

The results for CO_2 were reported previously.[12] In this case beam intensity measurements were performed with a collimator of large diameter (2.50 mm). The finite detector aperture and skimmer-dector distance introduced an error.

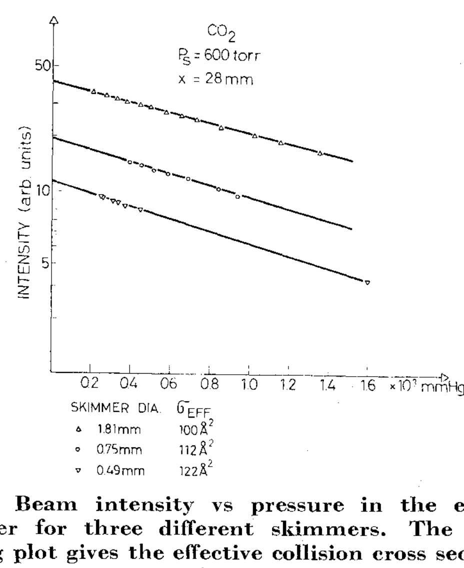

Fig. 5 Beam intensity vs pressure in the expansion chamber for three different skimmers. The slope in semilog plot gives the effective collision cross section σ_{eff}. Gas CO_2, source pressure 600 mm Hg, x 28 mm in all cases.

Table 2 Experimental values for the exponent m measured with different gases and different stagnation conditions

Carbon dioxide			Argon			Nitrogen		
Curve	Source press. (torr)	m	Curve	Source press. (torr)	m	Curve	Source press. (torr)	m
a	340	1.74	a	400	1.62	a	760	1.69
b	480	1.68	b	600	1.54	b	1520	1.52
c	600	1.66	c	800	1.47	c	2280	1.45
d	800	1.56	d	1000	1.45	d	3040	1.36
e	1000	1.38	e	1300	1.37	e	3800	1.33
f	1200	1.34	f	1600	1.35			
g	1400	1.33	g	2000	1.35			
			h	2500	1.33			

The error of the exponent m was estimated to be less than 4% for all experimental conditions.

In Table 2 all measured m values are collected. The results are qualitatively in agreement with the results obtained with different techniques. In Fig. 10 we compare our results for Ar with the data obtained by the Princeton group[7] in similar conditions. Our absolute $T_\perp$ values are about 25% larger, but the m value is the same. The recent results of Fisher and Knuth[9] are also consistent with our measurements.‡

It is worthwhile to note that the intensity measurements performed with the Snorkel gage technique[7] at great angle from the centerline are qualitatively consistent with the contribution of molecules with very low values of $S_\perp$ [second term of Eq. (8) in our analysis]. Similar features were found by other authors (Refs. 9, 18, and 25).

General Discussion

We have seen that our results for $T_\perp$ are qualitatively in agreement with the results obtained in other laboratories, but in fair agreement with the previsions of the theory of free expanding jets. In order to justify these results we discuss how the hypotheses given in the derivation of the ideal beam intensity equation are corrected and in particular how it is correct to speak about $T_\perp$.

Skimmer Interference

The problem of skimmer interference has been the subject of long discussions[5,26,27] and mainly to clarify the disagreement between experiment and theoretical predictions of absolute beam intensities. Under our experimental conditions (high x/d values) skimmer interference, if present, is probably of the kind suggested in Ref. 26, that is, molecules reflected by the outer surface of the skimmer collide with the expanding molecules and alter their spatial distribution.

However, such a process cannot explain our experimental results. Indeed if we plot the percentage of the intensity α originating from the low $S_\perp$ molecules for a given skimmer (say 1.49 mm) we find (see Fig. 11) that at low x/d α rapidly decreases§ and at high x/d is almost constant, while the influence of skimmer interference is expected to decrease rapidly with the distance.

Moreover, α is quite high (20 ÷ 30% for the 1.49 mm skimmer, 40 ÷ 50% for the 1.80 mm one) requiring high density of molecules reflected by the skimmer, while electron beam probe density measurements[28] have shown only a slight increase of density in front of the skimmer even in flow conditions which would lead to a severe interference.

Self-Scattering Downstream from the Skimmer

It is quite justified to suppose that the flow is free molecular downstream of the skimmer for small skimmers and high x/d values. Nevertheless our analysis assumes a negligible self-scattering downstream from bigger skimmer at shorter x/d values.

The theory of self scattering, i.e., of the transition flow, is not yet fully developed. A useful approach, valid when self scattering is not dominant, is due to Valleau and Deckers.[29-30]

Using their model the rate equation for the density can be integrated (disregarding the flow divergence) to obtain the following expression for the density at distance l from the skimmer opening (n_l)

$$n_l = n_s(S_{\perp s}d_s^2/4l^2)(1 + d_sS_{\perp s}/2\lambda_s)^{-1} \qquad (11)$$

$d_s \ll l$ has been assumed; where d_s is the skimmer diameter

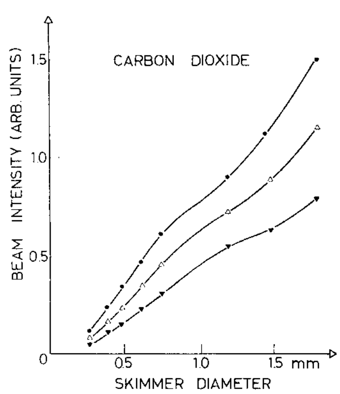

Fig. 6 Beam intensity corrected for background attenuation as a function of skimmer diameter. Gas CO_2, source pressure 1200 mm Hg; ● = x/d 95, △ = x/d 165, ▼ = x/d 235.

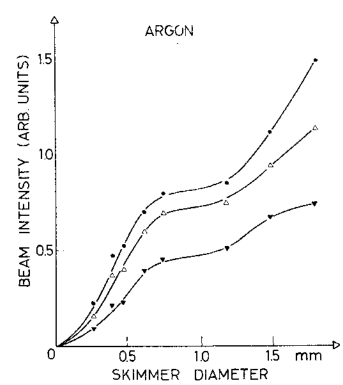

Fig. 7 Beam intensity corrected for background attenuation as a function of skimmer diameter. Gas Argon, source pressure 1300 mm Hg; ● x/d 95, △ x/d 165, ▼ x/d 235.

‡ Fisher and Knuth give three different values for the perpendicular Mach number using three different fitting criteria. Our results are compared with their values of $M_{\perp 1}$ (using their notation).

§ In this region probably α is determined mainly by the collisions *inside* the skimmer.

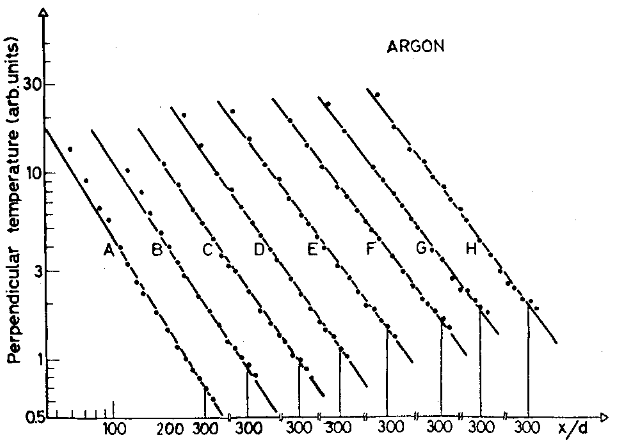

Fig. 8 Perpendicular temperature for Ar vs reduced distance from the nozzle (the source pressures and the values of the exponent *m* are given in Table 2).

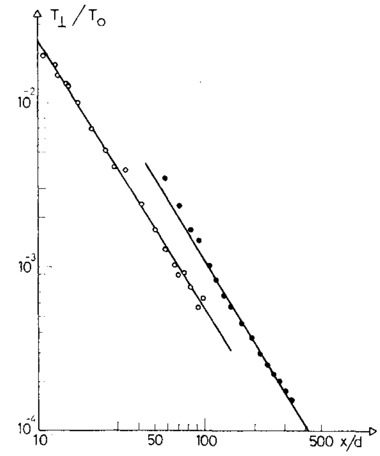

Fig. 10 Perpendicular temperature for Ar vs reduced distance; ○ from Ref. 7, $P_o \cdot d$ = 3.08 torr cm, and ●, our results (curve A, Fig. 8) $P_o \cdot d$ = 3.4 torr cm.

and

$$\lambda_s^{-1} = [(2)^{1/2}\pi\sigma^2 n_s]/u[(2/\pi)KT_{||s}/m]^{1/2} \quad (12)$$

The subscript *s* in the two preceding equations indicates that the values of the parameters are taken at the skimmer entrance; σ is the collision radius. Neglecting the second term in the denominator of Eq. (11) (hereafter called γ) we obtain the usual expression for beam intensity for skimmers of small diameter. The self-scattering term

$$\gamma = (2\pi)^{1/2}\sigma^2 n_s(S_{\perp s}/S_{||s})(d_s/2) \quad (13)$$

decreases rapidly with increasing x/d.

To estimate its order of magnitude we can proceed as follows. We can obtain: a) the density at the skimmer from the absolute value of the intensity; b) $S_{\perp s}$ from the analysis reported in the present work; c) $S_{||s}$ from the experimental determination reported in Ref. 7; and d) σ (the collision radius at the temperature $T_{||}$) from the experimental work recently reported by Knuth and Fisher.[31]

In this way we obtain a value of γ less than 0.2 in the worst conditions, i.e., with the biggest skimmer (1.49 mm) and at the shortest distance (4 mm).

Ellipsoidal Velocity Distribution at the Skimmer

In the derivation of the beam formation equation we assumed an ellipsoidal velocity distribution as did Hamel and Willis.[1] Such a function, $f(v)$, being the simplest solution of the moment equation, has lost many details of the real distribution function.

A similar derivation, using the hypersonic approximation is due to Edwards and Cheng.[2] In particular they find that the lateral velocity distribution can be thought of as describing two different classes of molecules. The first one is the class of molecules that, in agreement with Hamel and Willis, have a temperature $T_{1\perp}$ which changes as $(x/d)^{-1}$, the second class dominates at small angles with the centerline and has a temperature $T_{2\perp}$ which changes as $(x/d)^{-2}$.

Freeman and coworkers[3,32] have recently shown that it is possible to obtain similar results using a different approximation to obtain the moment equations. Computing the higher order moments it is possible to obtain more information on the velocity distribution to check the validity of separating the distribution function into the product of two functions (one for each of the two degrees of freedom: parallel and transverse). The authors suggest that the two degrees of freedom are not independent but, the coupling being weak, it is still reasonable to speak about $T_{||}$ and $T_\perp$. In particular they find higher tails for the lateral velocity distribution function than for the Maxwellian distribution function. The last result is consistent with the behavior of the term proportional to the skimmer area obtained from the fit of our experimental results (see Fig. 11).

We are led to the conclusion that the agreement between experimental results and theoretical predictions for $T_\perp$ is rather poor. A possible reason for the disagreement is that molecular beam sampling is limited to small angles with respect to the centerline while the transverse temperature predicted by the theory is essentially determined by the tails of the distribution function in the transverse velocity.

On the other hand we can obtain qualitative agreement with a simple model due to Le Roy et al.[33,34] In this model $f(v)$ is computed as in molecular beam sampling at small

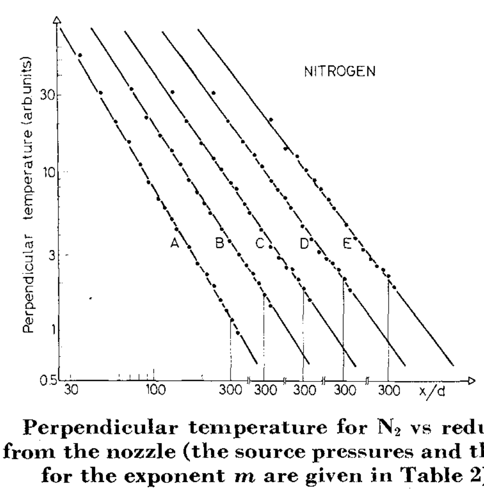

Fig. 9 Perpendicular temperature for N_2 vs reduced distance from the nozzle (the source pressures and the values for the exponent *m* are given in Table 2).

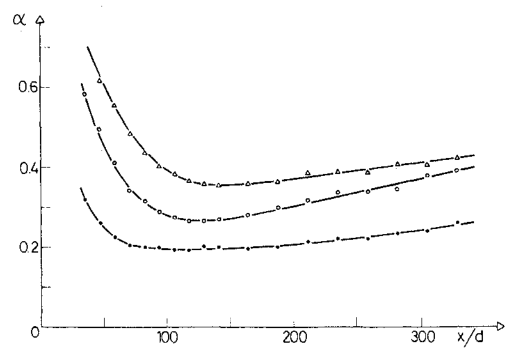

Fig. 11 Percentage beam intensity contribution proportional to the skimmer area (α) vs nozzle skimmer separation. α is defined as follows (see Eq. 8) $\alpha = Br_s^2\pi/A(1 - e^{-S_\perp^2\alpha_s^2(1+x^2/l)}) + B\pi r_s^2$ Gas Argon d_s 1.49 mm; ● 400 mm Hg, ○ 1000 mm Hg, △ 2500 mm Hg.

angles, on the surface of spheres of increasing radius, starting from a region near the nozzle where the flow is still continuous. The distribution function on each following sphere is computed as the sum of two terms; the first due to the particles that have not undergone a collision between the two spheres, the second due to the particles which have had a collision.

The collision model adopted by these authors is very simple. They assume equipartition of energy among the different degrees of freedom and use collision cross section for hard spheres.

The main advantage of the model is that the different parameters have a clear physical meaning. For $T_\perp$ the authors find that the distance dependence changes continuously from $(x/d)^{-4/3}$ (isentropic flow) near the source to $(x/d)^{-2}$ (free molecular flow) at greater distances. If the transition from continuous to free molecular flow is rather slow ($B > 0.5$ in the notation of Le Roy et al.) we can represent the distance dependence of $T_\perp$ with an equation of the kind $T_\perp \sim x^{-m}$ over a wide range of x/d values.

Our measurements then indicate that, for the Knudsen numbers used, the flow is still in the transition region at rather high distances from the source.

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