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PROJECT SQUID

TECHNICAL REPORT PR-111-P

STUDIES OF LOW DENSITY SUPERSONIC JETS

By

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CHARLOTTESVILLE, VIRGINIA

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STUDIES OF LOW DENSITY SUPERSONIC JETS*

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Abstract

Measurements of axial and radial velocity distributions in axisymmetric free jets of pure gases and binary gas mixtures are reported. Results are compared with extensions of the kinetic theory approach developed by Hamel and Willis and by Edwards and Cheng. The use of binary gas mixtures to produce molecular beams in the 1-10 ev range is described. Measurements of diffusive separation of mixtures in free jets are reported and compared with the theoretical predictions of Zigan and Sherman for nearly continuum flow.

STUDIES OF LOW DENSITY SUPERSONIC JETS

Introduction

The process of free expansion from a sonic nozzle or orifice into a low pressure region is of substantial practical and theoretical interest for a number of reasons. Such free jets are an effective means of obtaining flows at high Mach number and very low density. Because of this, they play an important role in the production of supersonic molecular beams of the type first proposed by Kantrowitz and Grey and first developed by Becker and Bier (1,2). Free jets are also attractive for certain kinds of wind tunnel tests (3). Since the jet passes rapidly from a collision-dominated region near the orifice to a practically collision-free region further downstream, it serves as an ideal flow field for studying very fast collisional processes. Indeed, information has been obtained on vibrational, rotational and translational relaxation by probing such flows (4,5,6). Various other rapid processes may be "frozen" and studied in this manner. In particular, probing free jet flows appears to be a powerful technique for studying homogeneous nucleation (7,8,9). Aside from these practical aspects, the free jet is intriguing as a theoretical problem in rarefied gas dynamics. Recently Hamel and Willis and Edwards and Cheng, using the moment equations of kinetic theory, independently proposed a hypersonic approximation for the spherical source expansion of a monatomic gas; their approach appears to warrant extension (10,11). Zigan and Sherman have also presented a continuum treatment of the extent of diffusive separation of gas mixtures in free jets (12,13).

Our own interest in free jets originated with our work in developing a molecular beam system of the Kantrowitz-Grey type and in using "seeded beam" techniques to produce high intensity neutral beams in the 1-10 ev range (14). Basic to such a program is the ability to extract a relatively unperturbed sample of the jet to form the molecular beam. When unperturbed samples are obtained conditions in the free jet may be inferred from the properties of the molecular beam formed. Earlier results of such free molecular sampling were presented at the Toronto Symposium (15,16). The purpose of the present paper is to summarize some of our recent findings in the hope that they will not only be of interest in the design of nozzle beam systems but also contribute to elucidating the behavior of free jets.

Discussion

Results for Pure Gases

An isentropic free jet attains the characteristics of a source flow with a mean speed very close to its adiabatic limit within a distance of about two nozzle-diameters downstream of the nozzle exit (17). As a result, it is difficult to study the jet by density or total momentum measurements. But because the density in the jet decreases rapidly with increasing distance, it is sometimes possible to pass an undisturbed sample through a skimmer into a second vacuum chamber and obtain the detailed velocity distribution of the resulting molecular beam. In this section we will report some recent measurements we have obtained in this manner with argon, helium and neon jets.

In the absence of perturbations due to interaction of the jet with the skimmer or due to further collisions among the beam molecules themselves, the radial spread of molecules downstream of the skimmer is directly related to their initial velocity distribution perpendicular to the streamline sampled. This radial distribution of molecular flux was measured by moving an enclosed ionization gauge along a line perpendicular to the beam axis as described by Anderson and Fenn (6). The detector had a 0.15-cm diameter opening and was located 20.0 cm downstream of the skimmer entrance. It was equipped with a movable flag which was used to block the beam so that the base signal due to background molecules could be measured.

Figure 1(a) shows an intensity distribution obtained for a beam extracted from an argon jet formed by a 0.033-cm diameter orifice with a stagnation pressure of 46 torr and temperature of 300°K. The 0.015-cm diameter skimmer entrance was located on the jet axis 39 nozzle diameters from the nozzle exit. The solid curve shown is the calculated intensity distribution for a Maxwell-Boltzmann radial velocity distribution (at temperature $T_{\perp}$) which gives the best fit to the data. The tails of the experimental curve are slightly broader than those of the calculated curve. This is typical of the results. While there is some uncertainty in the base signal in these measurements, the slight deviation from ideal behavior appears to be real.

Velocity distributions parallel to the beam axis were measured with a time-of-flight technique which has been described previously (6). The beam is chopped by a rotating shutter and the density-time pattern of the beam pulses is measured downstream of the chopper with a nude ionization gauge. A typical density-time curve is shown in Fig. 1(b). This distribution is for a beam extracted from a helium jet formed by a 0.033-cm diameter orifice at a stagnation pressure of 400 torr and temperature of 300°K. The skimmer entrance was 58 nozzle diameters from the nozzle exit. The solid curve is, as before, that calculated for the Maxwell-Boltzmann distribution at the skimmer entrance (with axial temperature $T_{\parallel}$ and mean velocity u) which gives the best fit to the experimental curve. Several methods of fitting were employed, including a least squares analysis and a simple measurement of the relative width of the curve at half height. If there is no disturbance of the flow by the skimmer these methods yield essentially the same values for u and $T_{\parallel}$.

As mentioned earlier, the key question in relating these measurements to conditions in the jet is that of disturbance of the state of the gas sample by interaction with the skimmer. The best proof of ideal sampling would be to probe the isentropic, continuum region near the nozzle exit and verify the results obtained with Pitot probes. Unfortunately, this has not been possible. Fenn and Deckers observed, however, that as Kn/M (the ratio of Knudsen number based on free stream mean free path and skimmer diameter to the free stream Mach number given by method-of-characteristics calculations) became larger than unity the centerline intensity of their beam approached theoretical predictions (18). Such a relation (at least for Knudsen number dependence) should account for collisions of beam molecules with molecules reflected from the skimmer walls and for collisions within the beam itself. Consequently, if one samples a given flow with geometrically similar skimmers of different diameters and obtains identical velocity distributions, the sampling process must be ideal. Such a result is shown in Fig. 2 in which esssentially the same axial Mach number $M_{\parallel}$ is indicated for two slit skimmers, one 0.008 cm wide - the other 0.014 cm wide.

Figure 2 also shows the effect of increasing nozzle pressure with a given skimmer and nozzle-skimmer distance. Since the skimmer is located in the transition region of the jet, the observed axial Mach number increases with increasing pressure until skimmer interference becomes important. With further pressure increases the observed Mach number actually decreases although the Mach number in the jet continues to increase. Such experiments offer a simple way of estimating the critical Kn/M for a given geometry but are not as convincing as the use of a series of skimmers of different sizes.

Most of the velocity distribution experiments to date have been directed to characterizing the asymptotic axial Mach number of a jet in terms of nozzle conditions (6, 15, 19). This is partly due to the importance of terminal Mach number in the design of beam experiments but is also due to the difficulty in avoiding skimmer interaction at small nozzle-skimmer distances. Slit skimmers, however, with widths of 0.010 cm and less can be easily fabricated and can be used to probe the transition region of the jet (5). Figure 3 indicates the results of such an experiment. The radial temperature $T_{\perp}$ along the axis of an argon jet was determined in the range of 8 to 100 nozzle-diameters for three values of $P_o d_n$ (product of stagnation pressure and nozzle exit diameter). In each case $T_{\perp}$ appears to decrease initially as $x^{-4/3}$ and shift to an x^{-2} dependence at larger values of x.

This behavior of the radial temperature is in direct disagreement with the theoretical treatments of Hamel and Willis and of Edwards and Cheng which predict an asymptotic behavior of the form x^{-1} (10,11). These treatments are for source flow; with the additional assumptions that a) the moment equations can be truncated to yield an "ellipsoidal" distribution function (not necessarily of Boltzmann form) characterized by a density, a mean velocity, an axial temperature and a radial temperature and b) a simple relaxation form can be used for the collision terms arising in the theory (corresponding essentially to the assumption of Maxwell molecules). We have undertaken an extension of the theory by evaluating the collision terms using a Lennard-Jones (6-12) potential and assuming an ellipsoidal Boltzmann form for the distribution function. The details of this work will be published in full at a later date; however, some of the results are shown in Fig. 4.

The experimental data shown are for an argon jet with $p_o d_n$= 1.54

torr cm. A systematic study of $T_{\parallel}$ was not performed. The $T_{\parallel}$ points in the transition region where taken to determine the threshold of skimmer interaction for various skimmers and are apt to be high. As a result the experimental curve for $T_{\parallel}/T_{o}$ is mainly of heuristic value. The values of $T_{\perp}$ measured in the transition region also appear to be slightly high, nevertheless the shape of the curve is believed to be free of systematic errors.

The dashed lines correspond to a numerical integration using the Lennard-Jones model. The calculation was started one nozzle-diameter downstream of the nozzle exit assuming an isentropic Mach number of 2.5. This yields an isentropic temperature profile for the source flow that corresponds at large distrances to the method-of-characteristics solution for an axisymmetric jet. The axial temperature computed with the model is seen to relax to a point below the level experimentally observed. (For comparison we have included the asymptotic results obtained for hard sphere and Maxwell molecules having the same viscosity as argon at 300^{o}K.) The computed radial temperature lags slightly below the isentropic temperature that would be observed in a continuum expansion and then crosses over to decrease at a slower rate (in disagreement with the experimental data). Thus even the use of a realistic molecular model does not as yet yield an accurate description of the velocity distribution.

It appears that either a) the assumption of a Maxwell-Boltzmann distribution for radial velocities is at fault or perhaps b) the finite nature of the jet propagates to the axis and negates the source flow assumption. If $T_\perp$ were taken as the radial energy moment of an arbitrarily shaped velocity distribution rather than the temperature giving the best fit for a Boltzmann distribution, the discrepancy might be eliminated. It should be noted that the tails of the experimental radial distributions can contain a relatively large amount of energy in radial motion while appearing as only small deviations from the Maxwell-Boltzmann distribution. Thus the definition of $T_\perp$ is a delicate matter. Regarding b) above, it should be noted that the break in the $T_\perp$ curve is quite sharp and the break in the $T_\parallel$ curve is somewhat sharper than predicted. If it were assumed that the flow follows the model up to a certain axial distance and then departs into free molecular flow, the revised predictions would agree surprisingly well with experiment.

The results of some recent velocity distribution experiments to determine the asymptotic axial Mach numbers of free jets are shown in Fig. 5. The data shown are for pure helium, neon and argon at room temperature and for pure helium and argon at elevated temperatures (to 2100°K). To achieve the high stagnation temperatures, a tantalum tube positioned perpendicular to the beam axis was heated electrically. An orifice cut into the wall of the tube served as the nozzle. To achieve high values of $p_o d_n$ within the limits of pumping capacity several types of small nozzles were used: converging nozzles formed from glass capillary tubes by heating the ends of the tubes to form tapered internal closures and grinding the ends away; mounted watchmakers' jewels of several shapes; and holes cut in thin metal foil.

In determining the terminal Mach number of these jets it is necessary to avoid disturbance of the velocity distribution by skimmer interaction and by scattering by background gas in the nozzle exhaust chamber. The absence of background scattering effects was demonstrated by varying the background pressure and observing no effect on the velocity distributions. For large nozzle-skimmer distrances background scattering effects were found to be important; these data have been discarded.

In the correlation of experimental results in Fig. 5 the Knudsen number Kn^* used is the ratio of mean free path (hard sphere, based on viscosity at stagnation conditions) to nozzle exit diameter. Mach terminal M_T is that defined by Anderson and Fenn (6). The results for neon and argon are in agreement with previous work in that $M_T = 1.17\,Kn^{*-0.4}$ (6). For helium there is a difference. The theoretical points indicated for argon were calculated for a stagnation temperature of 300°K using the Lennard-Jones potential as described previously. Each theoretical calculation was terminated at the point where $T_{\perp}/T_0$ begins to follow an x^{-2} dependence. For helium it is clear that use of $M_T = 1.17\,Kn^{*-0.4}$ is not adequate. Calculations taking into account the quantum mechanical effects on collision cross-section indicate a significant difference between the variation of helium cross-section with temperature and that of argon. Calculations of theoretical values of M_T for helium, using quantum mechanical calculations for collision cross-section at low temperatures, are now under way.

Results for Gas Mixtures

Aerodynamic acceleration of a heavy gas by a light gas in the expansion from a nozzle (the "seeded beam" technique) makes possible the production of a high intensity of neutral molecules in the 1-10 ev range. The essential principles have been described previously and the effectiveness of the technique has been demonstrated in our laboratory (14,20). The experiments reported here were directed to examining the details of the expansion process for the mixtures: argon-helium, xenon-helium and xenon-hydrogen in the stagnation temperature range 300-2100°K.

The experimental procedures for axial velocity distribution analysis were the same as those for pure gases. The nude ionization gauge used as a beam detector was about ten times more sensitive to argon than helium. This, together with the increased concentration of argon on the beam axis, assured measurements strongly biased to the argon species. Similarly, with xenon-helium and xenon-hydrogen the measurements are biased to xenon expect in cases of widely differing species velocities for which individual species velocity distributions could be observed.

For nozzle Reynolds number Re_o (based on stagnation conditions and nozzle exit diameter) greater than 1000 the measured mean velocities of argon-helium mixtures were in agreement with predictions based on continuum, isentropic expansion to high Mach number. Figure 6 indicates the variation of velocity with composition for expansion at high Reynolds number at 298°K. Departures from the theoretical mean velocities are less than two per cent.

Similar results were obtained for a 1% argon - 99% helium mixture ($300^{o}K < T_o < 2100^{o}K$); see Fig. 7. For 1% xenon - 99% hydrogen mixtures, the incomplete rotational relaxation of hydrogen prevents a simple interpretation of the results. However, for $T_o = 298^{o}K$ and $Re_o = 3000$ the observed velocity was only 15 percent lower than that predicted for continuum expansion with complete rotational relaxation of the hydrogen.

These experiments and others previously reported demonstrate that aerodynamic acceleration of a heavy gas by a light gas to produce high intensity molecular beams of 1 to 10 ev range is simple and effective. The technique has been used in gas-surface interaction studies by Marsden and Abuaf (21).

At the low Reynolds number limit (free molecular flow) there is, of course, no coupling between the velocities of the light and heavy species in the flow field. With increasing Reynolds number one expects the velocites to become more nearly equal. At the high Reynolds number limit they become the same. We have, in fact, observed the phenomenon of velocity "slip" in the transition regime with argon-helium, xenon-helium and xenon-hydrogen mixtures.

The experimental results are most dramatic for xenon-hydrogen mixtures since separate peaks can be seen in the time-of-flight oscillograms. Figure 8 shows the time-of-flight oscillograms for a 1% xenon - 99% hydrogen mixture expanding from a 0.051-cm orifice at about 1980°K. Nozzle pressure increases from about 10 torr to 100 torr in the oscillograms shown. Time increases right to left. The first peak - a time zero marker, the second - hydrogen, the third - xenon. As pressure increases the hydrogen velocity decreases and the xenon velocity increases. At still higher pressures the two peaks merge.

In order to correlate the experimental information obtained for a number of gas mixtures and to enable a priori prediction of the velocity slip, we have developed a mathematical model for expanding mixtures similar to that used for pure gases. The assumptions required are a) the free jet can be approximated along the axis as a spherical source and b) the velocity distribution function of each component can be approximated by a local Maxwell-Boltzmann distribution having a single temperature. A Lennard-Jones (6-12) potential was used in evaluating the collision terms. While this model cannot account for detailed velocity distributions for the two species far from the nozzle exit, the predictions of slip obtained by numerical integration of the equations are in excellent agreement with the experiments. A "sudden freeze" approximation applied to the model yields the simple correlation shown in Fig. 9. The correlation relates the fraction of the ideal velocity obtained by the heavy component in the gas mixture to an effective inverse Knudsen number based on stagnation conditions:

$$Kn_{slip}^{-1} = \sqrt{2}\,\pi n_0 \sigma_{hl}^2\, \Omega^{(1,1)*}\left(kT_0/\epsilon_{hl}\right) \frac{m_{avg}}{m_h - m_l}$$

Here σ_{hl} and ϵ_{hl} are the parameters characterizing the assumed Lennard-Jones potential between the two molecules and $\Omega^{(1,1)*}$ is the reduced collision integral important in calculating the coefficient of binary diffusion (22). The mass dependence is the average molecular mass divided by the difference in molecular masses. Results are presented in Fig. 9 for several different gases in mixtures with helium. The measured mean velocities are accurate within about two per cent. Table I presents data for the 1% xenon - 99% helium mixtures.

Asymptotic Mach numbers computed from axial velocity distributions observed for argon-helium mixtures from room temperature nozzles are shown in Fig. 10. For initial argon concentrations of ten per cent or greater the Mach numbers are essentially those for argon in the final beam. For lower initial argon concentrations the helium must be taken into consideration in interpreting the results. Included in Table I are the terminal Mach numbers observed for 1% xenon - 99% helium mixtures; these Mach numbers may be assumed to apply to the xenon species since the helium peaks were observed to be negligible in comparison to the xenon peaks.

Diffusive Separation of Mixtures

Measurements of diffusive separation of argon-helium mixtures in free jet expansions were made with a mass spectrometer flux detection system. The apparatus consisted of a nozzle (0.002 cm I.D. converging or 0.020 I.D. sharp-edged orifice) and a magnetic mass spectrometer located in a high vacuum chamber with high pumping capacity. The detector entrance was a 2-mm orifice at the tip of a cone located 24.5 cm from the nozzle exit. The flux at the detector was sufficiently low that free molecular flow was assured. In operation the chamber background pressure was normally 10^{-6} torr; thus, no effects of background gas were encountered. The nozzle was installed on a device which permitted rotation of the nozzle about the center of its exit plane for flux measurements up to $\pm 90^{\circ}$ from the axis. The mass spectrometer was calibrated as a flux detector with pure gases flowing from an effusive source.

A typical species distribution is shown in Fig. 11. The curve for each species is normalized to unity on the axis. Conditions are: converging nozzle, 0.002 cm I.D., 477 torr stagnation pressure, $298^{\circ}K$ stagnation temperature, initial composition 5.0 mole per cent argon. The flux on the axis is 14.4 mole per cent argon. At high Reynolds number ($Re_o > 1000$) the two curves merge to form a single curve with flux approximately proportional to $\cos^4 \theta$ (θ is the angle of inclination to the axis). At low Reynolds number ($Re_o < 1$) a single curve is obtained with flux proportional to $\cos \theta$ with the sharp-edged orifice.

Figure 12 shows the increase in mole fraction argon in the flux on the axis for initially 5.0 mole per cent argon plotted vs. the inverse nozzle Reynolds number (based on stagnation density, speed of sound and viscosity and nozzle exit diameter). Also indicated is the result of a calculation based on Sherman's theory for diffusive separation under conditions of nearly inviscid flow (13). The required Mach number distribution in the transonic region of the jet was obtained for a sharp-edged orifice in separate experiments with Pitot-static probes. As can be seen there is reasonable agreement between experiment and theory in the nearly inviscid regime. Rothe's results with electron beam measurements have previously confirmed the agreement in this region (23). For $Re_o < 50$ large deviations occur and separation decreases with decreasing Re_o. In the free molecule limit no separation occurs in the jet itself. However, the composition of the flow through the nozzle is richer in helium than the composition of the gas in the stagnation chamber as expected for effusive flow.

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Table I

Xenon - Helium Mixtures*

Nozzle Pressure torr	Mean Xenon Velocity cm/sec	Terminal $M_{\parallel}$ for Xenon
25	10.6×10^4	8.8
43	11.7×10^4	10.2
77	13.0×10^4	12.1
130	13.8×10^4	13.5
205	14.4×10^4	14.4
309	14.8×10^4	15.5
405	15.0×10^4	16.3
700	15.2×10^4	17.0

* $d_N = 0.0328$ cm; $T_o = 298^{o}K$.

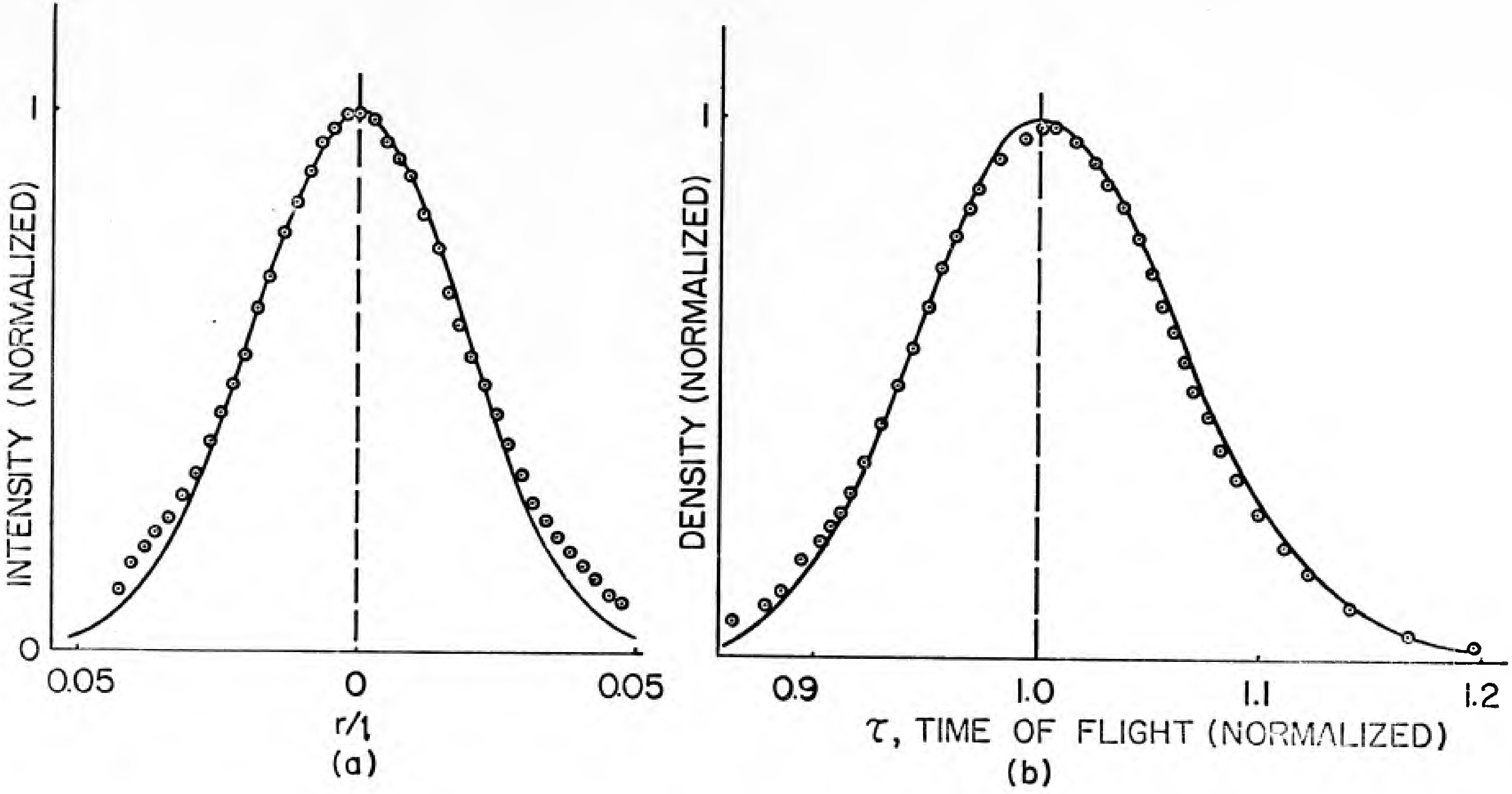

Fig. I. Typical radial intensity distribution (a) and axial velocity distribution as density-time trace (b) for molecular beams extracted from free jets.

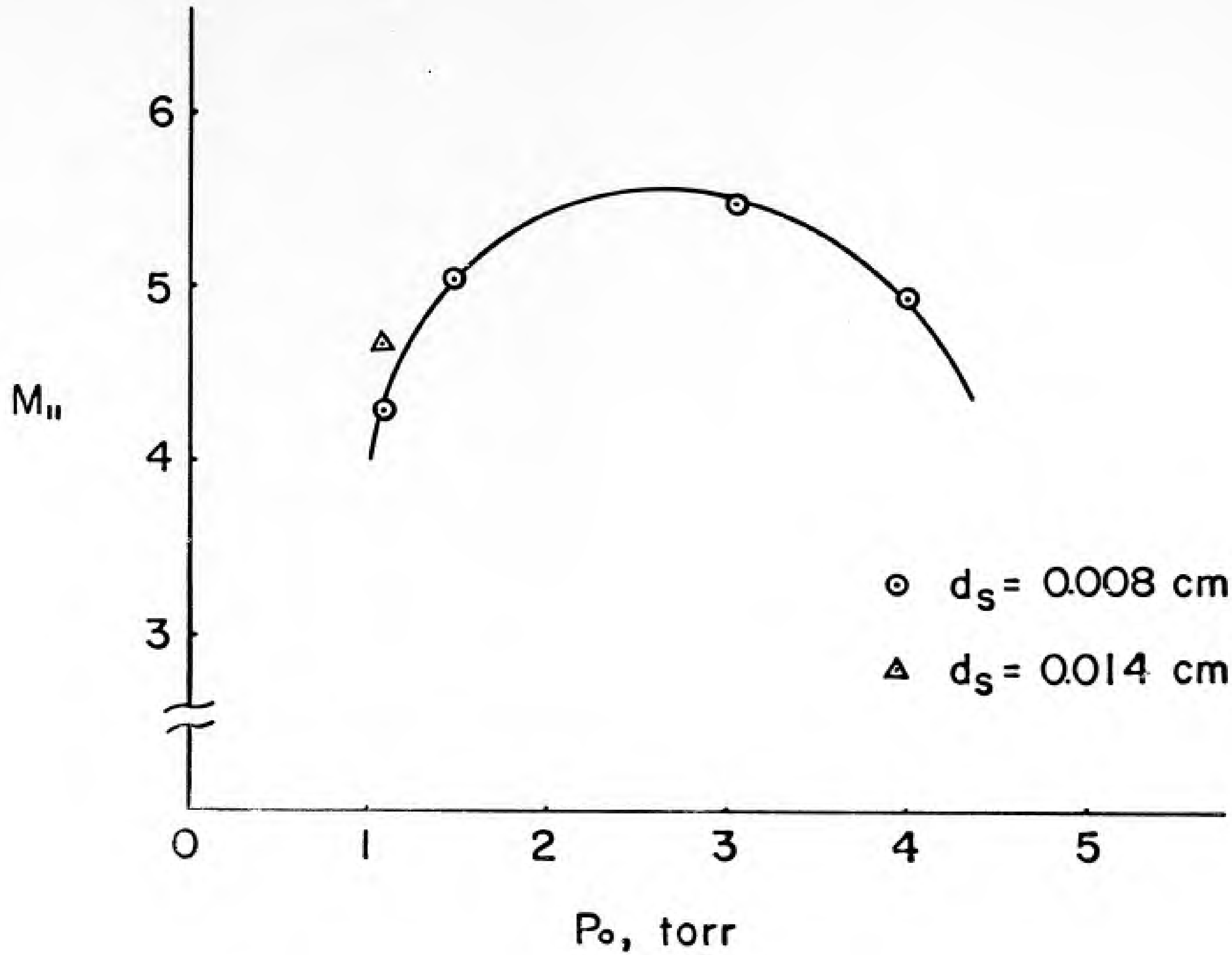

Fig. 2. Variation of measured axial Mach number with nozzle pressure and skimmer opening (argon; $x/d_n = 4$; $d_n = 0.484$ cm; $T_o = 300^oK$).

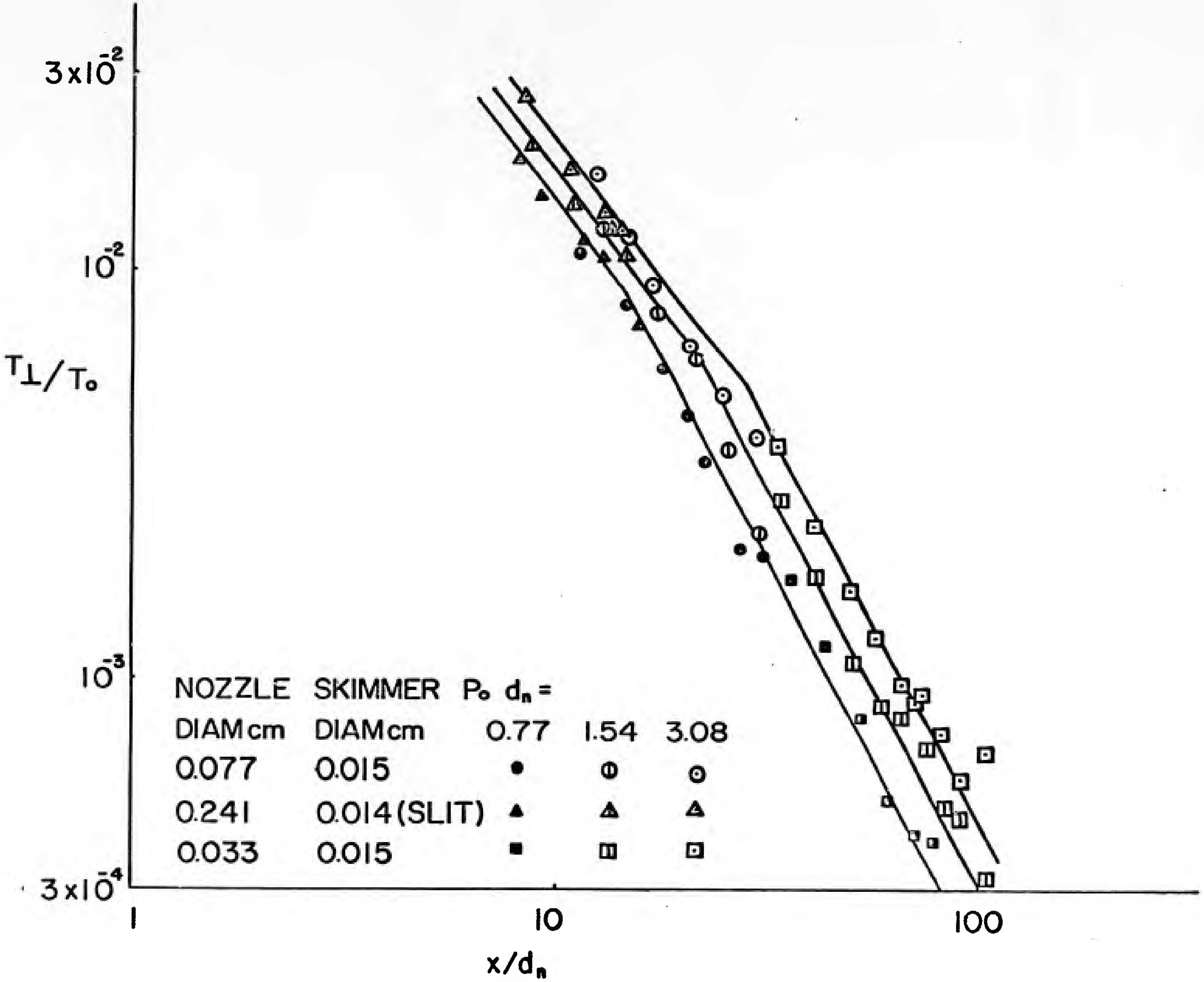

Fig. 3. Experimental measurements of radial temperature in a free jet as a function of distance and nozzle conditions. The solid line represents continuum expansion.

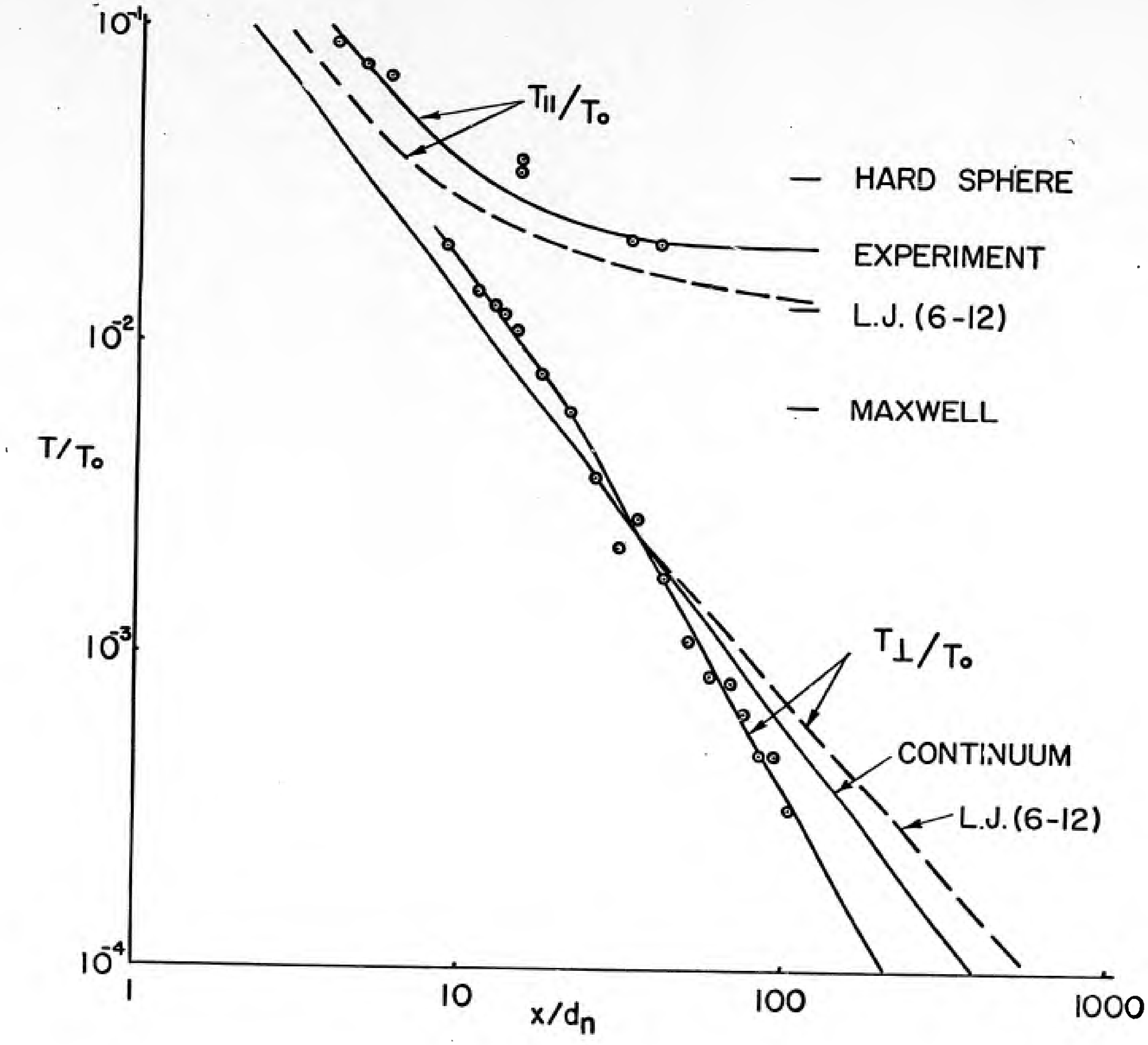

Fig. 4. Theoretical and experimental values of $T_{\perp}$ and $T_{\parallel}$ for an argon jet.

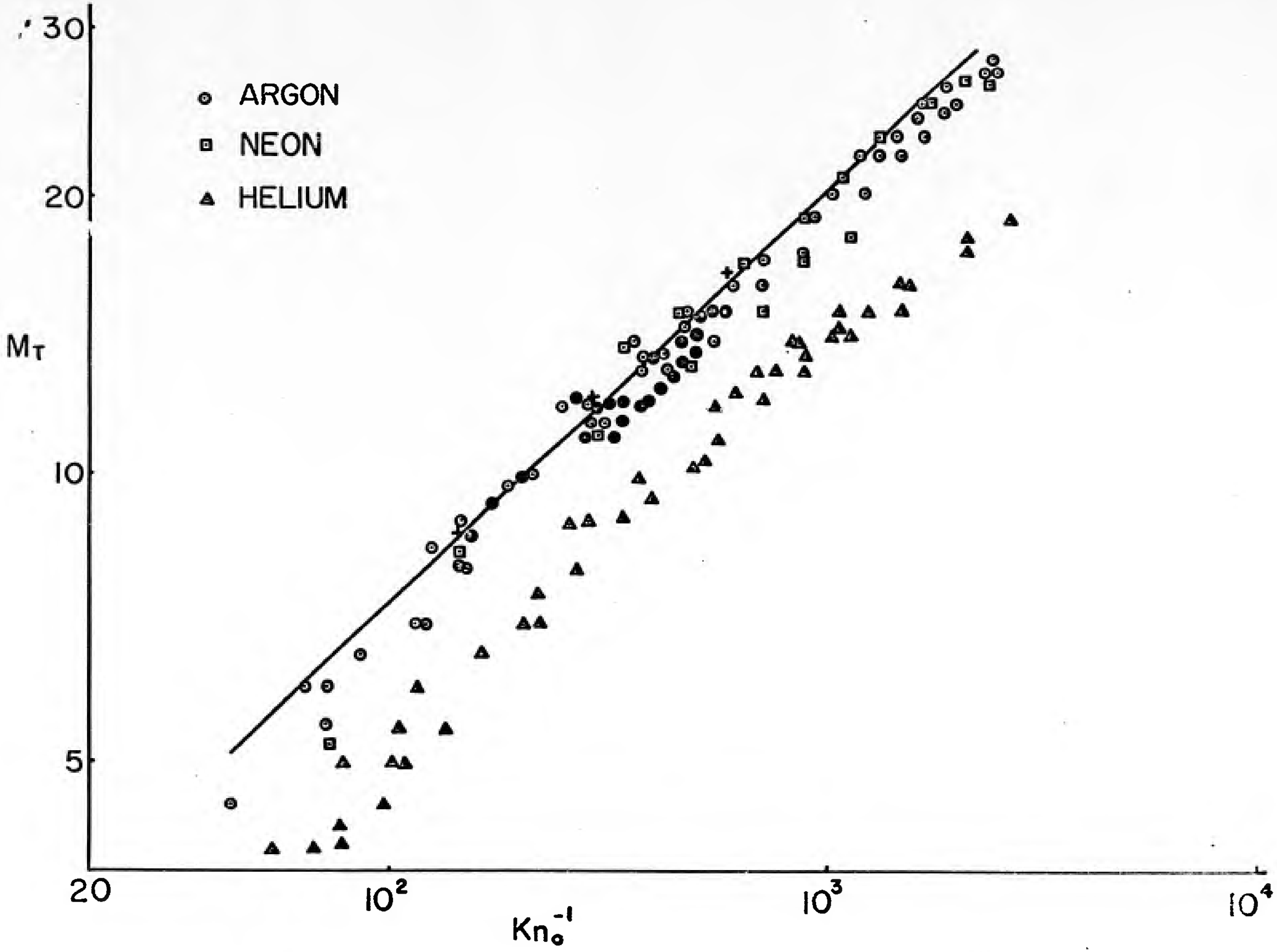

Fig. 5. Correlation of asymptotic axial Mach number for argon, neon and helium jets. Open points indicate $T_o = 300^\circ K$; filled points indicate elevated nozzle temperatures. The crosses are the results of a computation using the Lennard-Jones potential.

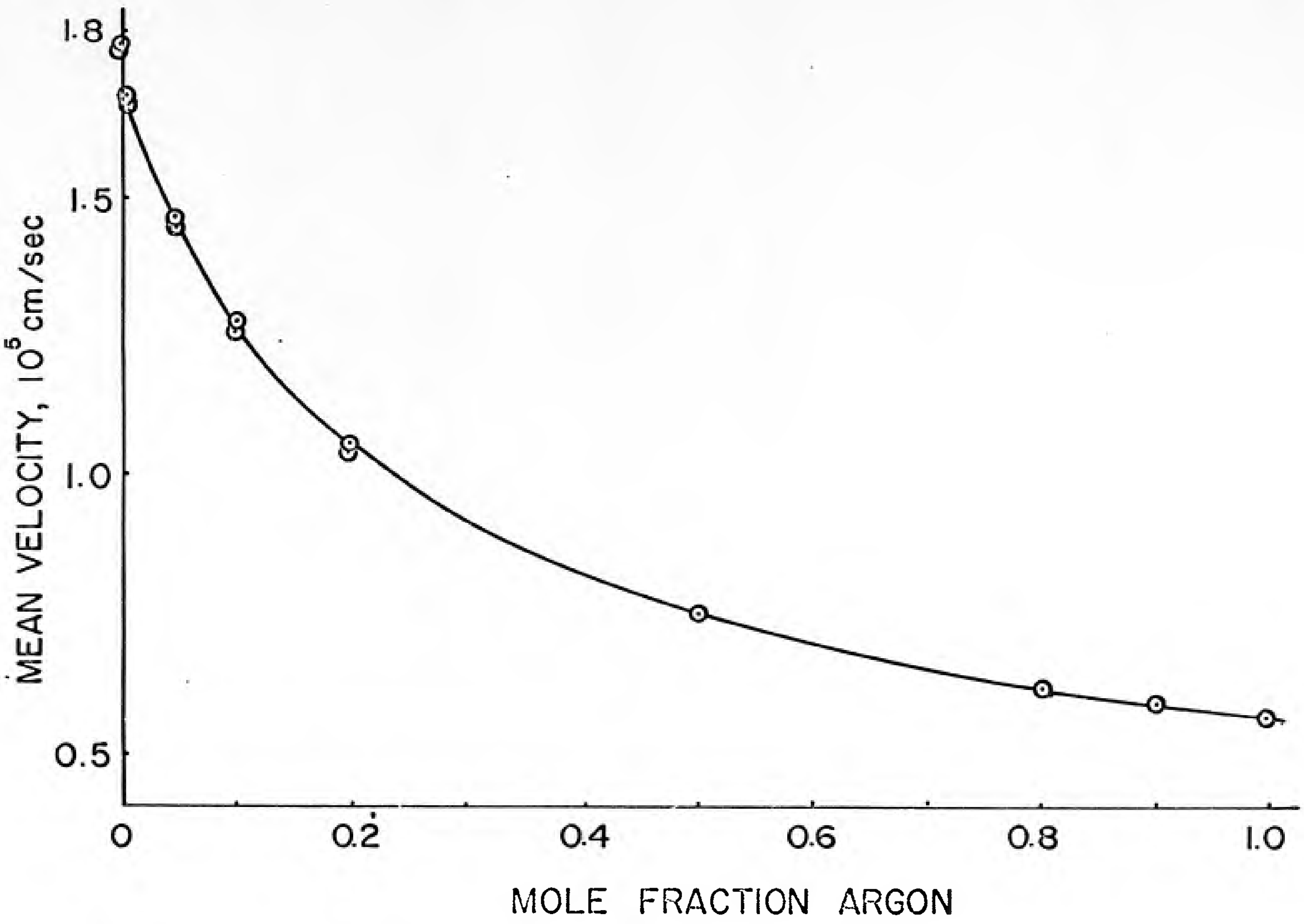

Fig. 6. Variation of mean axial velocity with stagnation composition for beams formed with argon-helium mixtures at high Re_o. The solid line represents theoretical values.

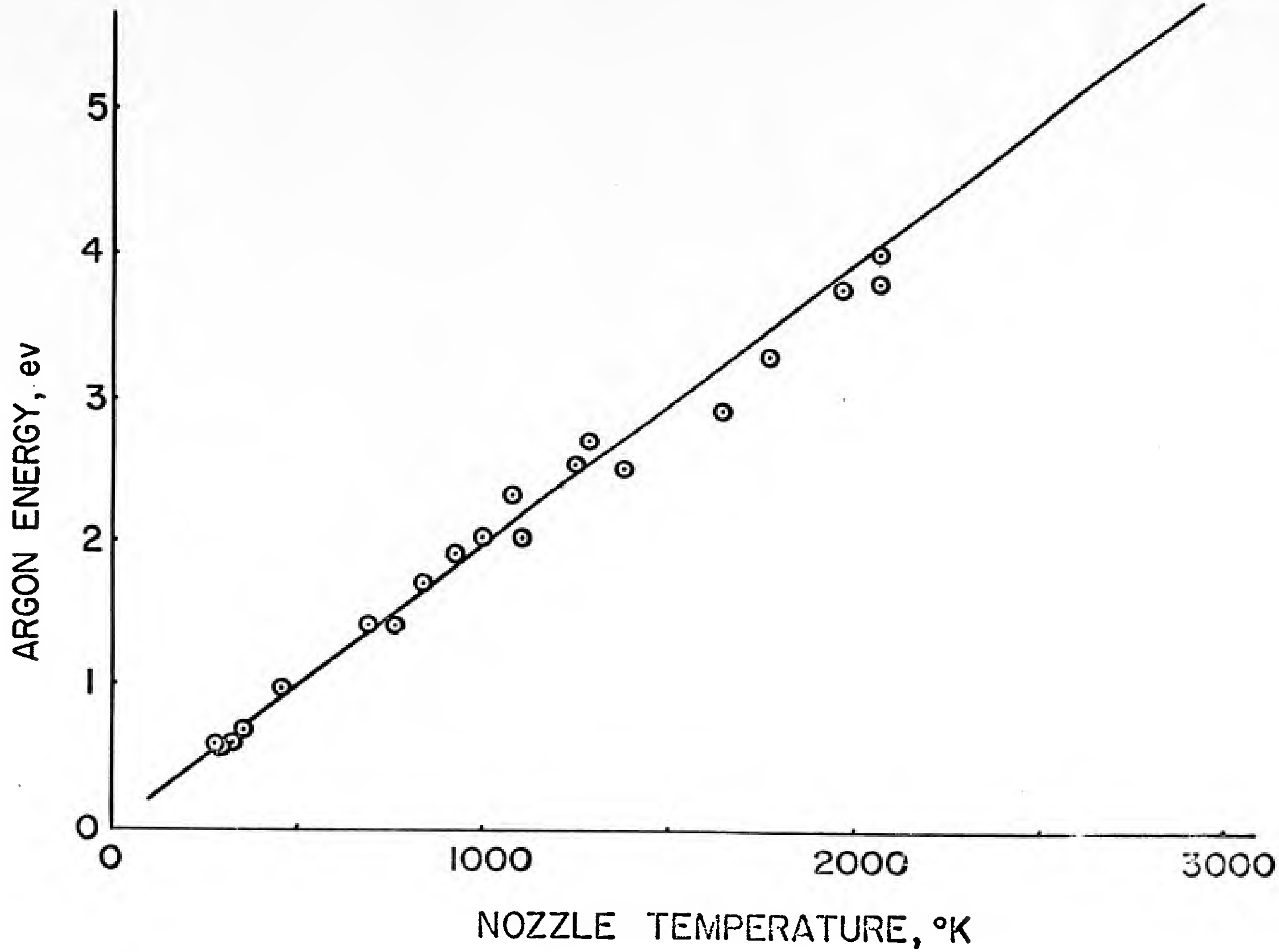

Fig. 7. Measured argon energy for beams formed with 1% argon - 99% helium mixtures at elevated temperatures with $Re_o > 1000$. The solid line represents the theoretical argon energy.

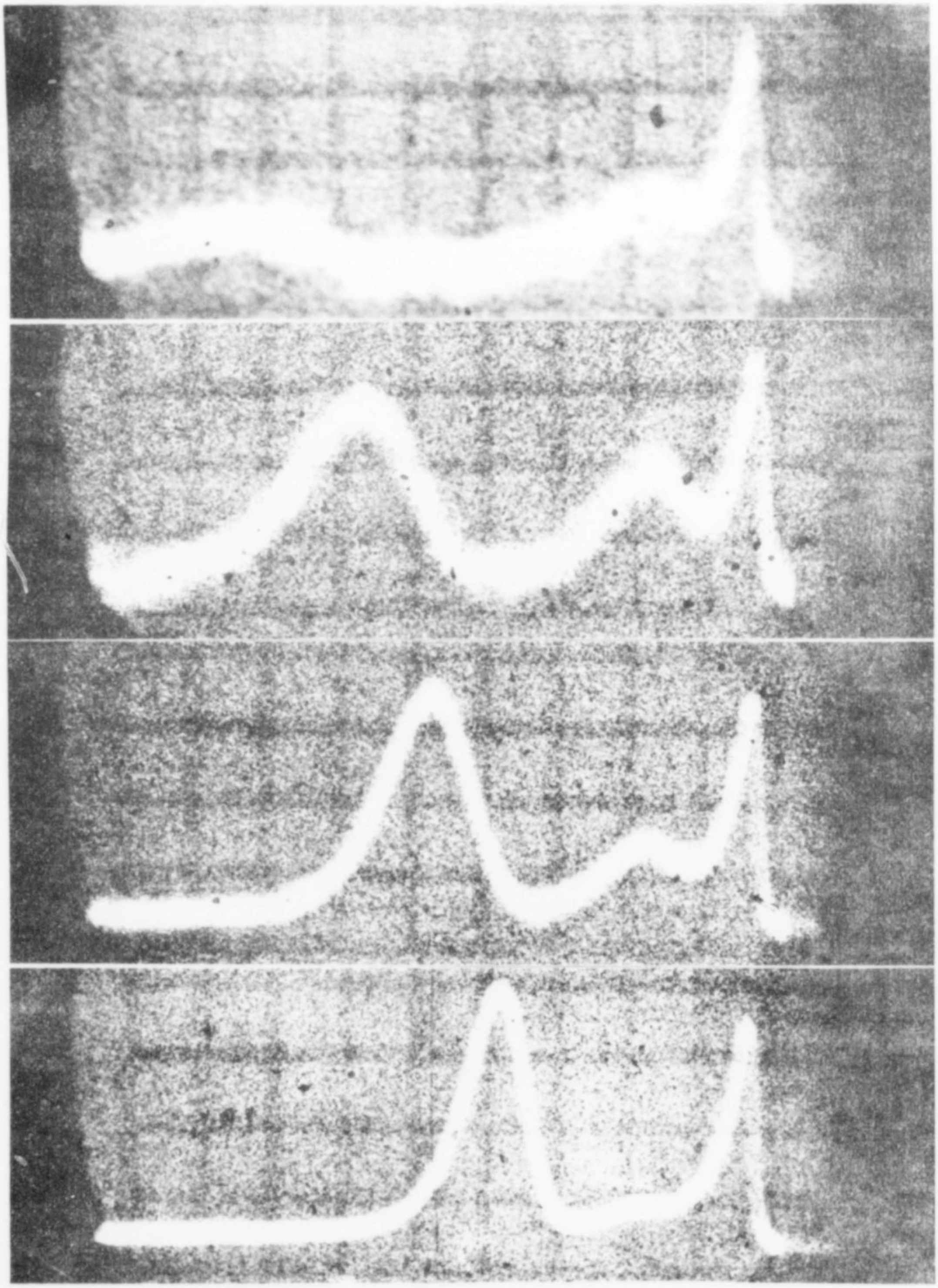

Fig. 8. Time-of-flight oscillograms showing separate peaks for hydrogen and xenon obtained with a 1% xenon - 99% hydrogen mixture at about 1980°K. Nozzle pressure increases from top to bottom oscillograms.

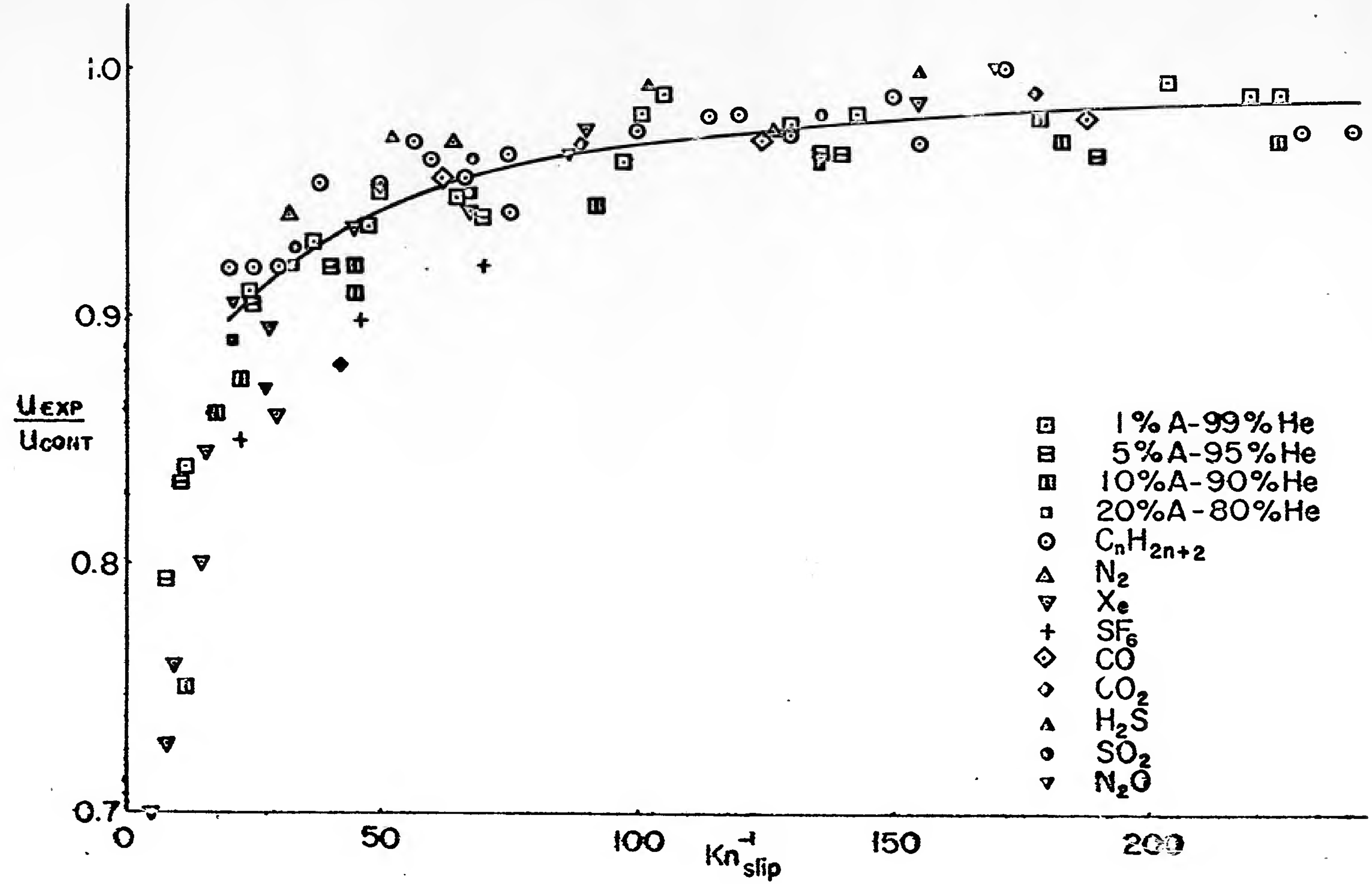

Fig. 9. Correlation of measured terminal velocity with initial conditions for several gases in mixtures with helium

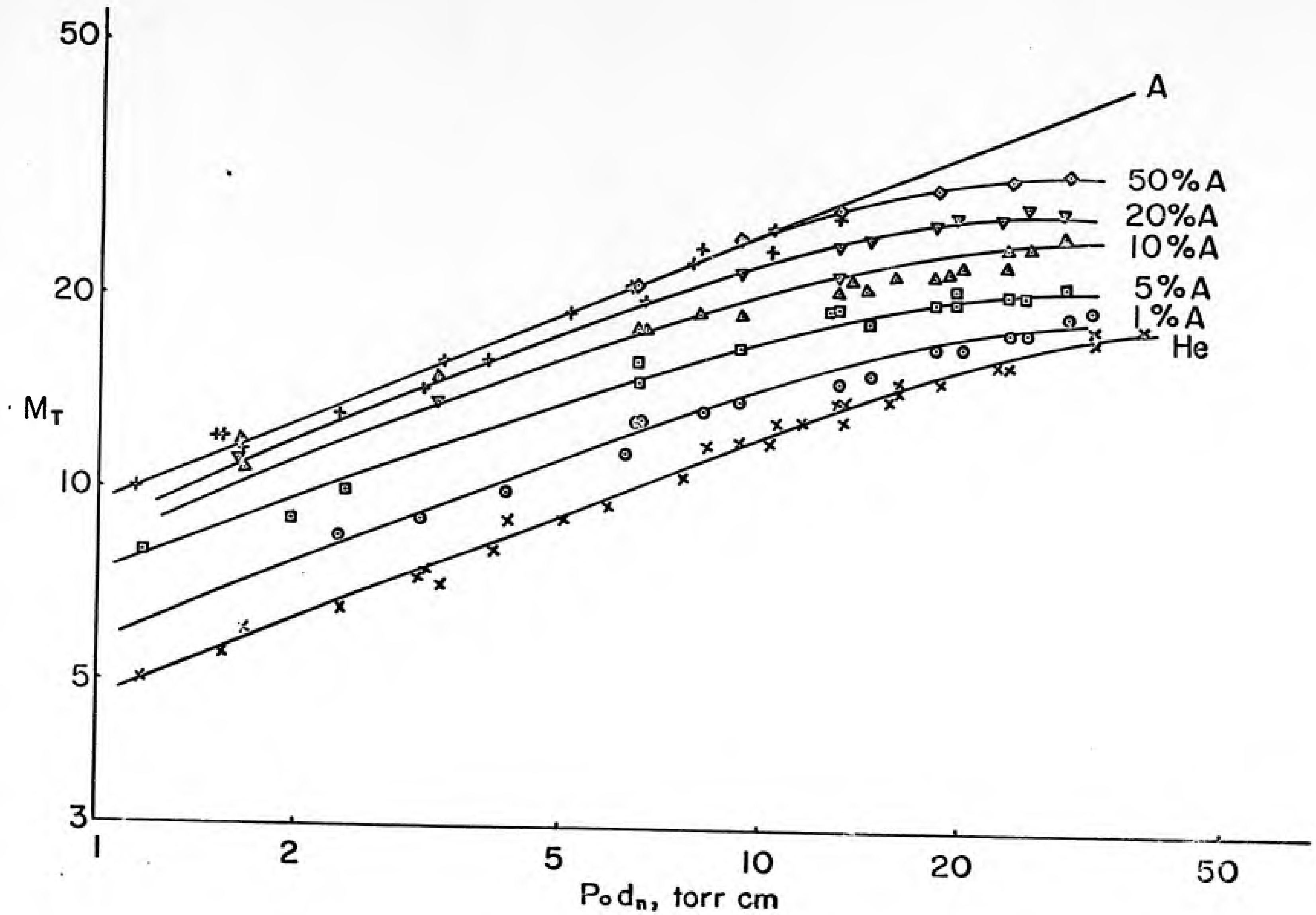

Fig. 10. Asymptotic axial Mach numbers for argon-helium mixtures expanding from room temperature nozzles.

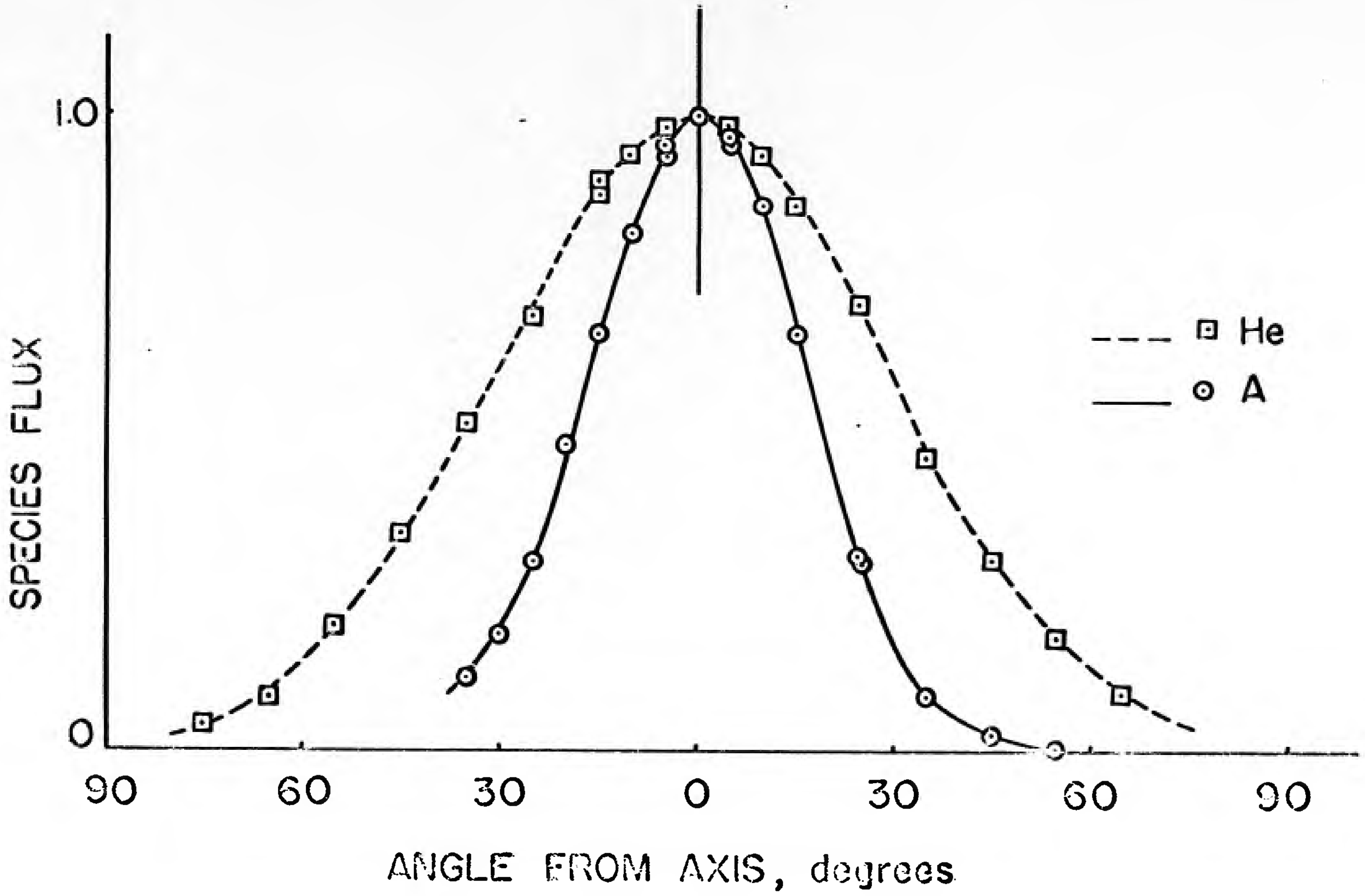

Fig. 11. Typical spatial distribution of species resulting from diffusive separation of argon-helium mixtures in a free jet.

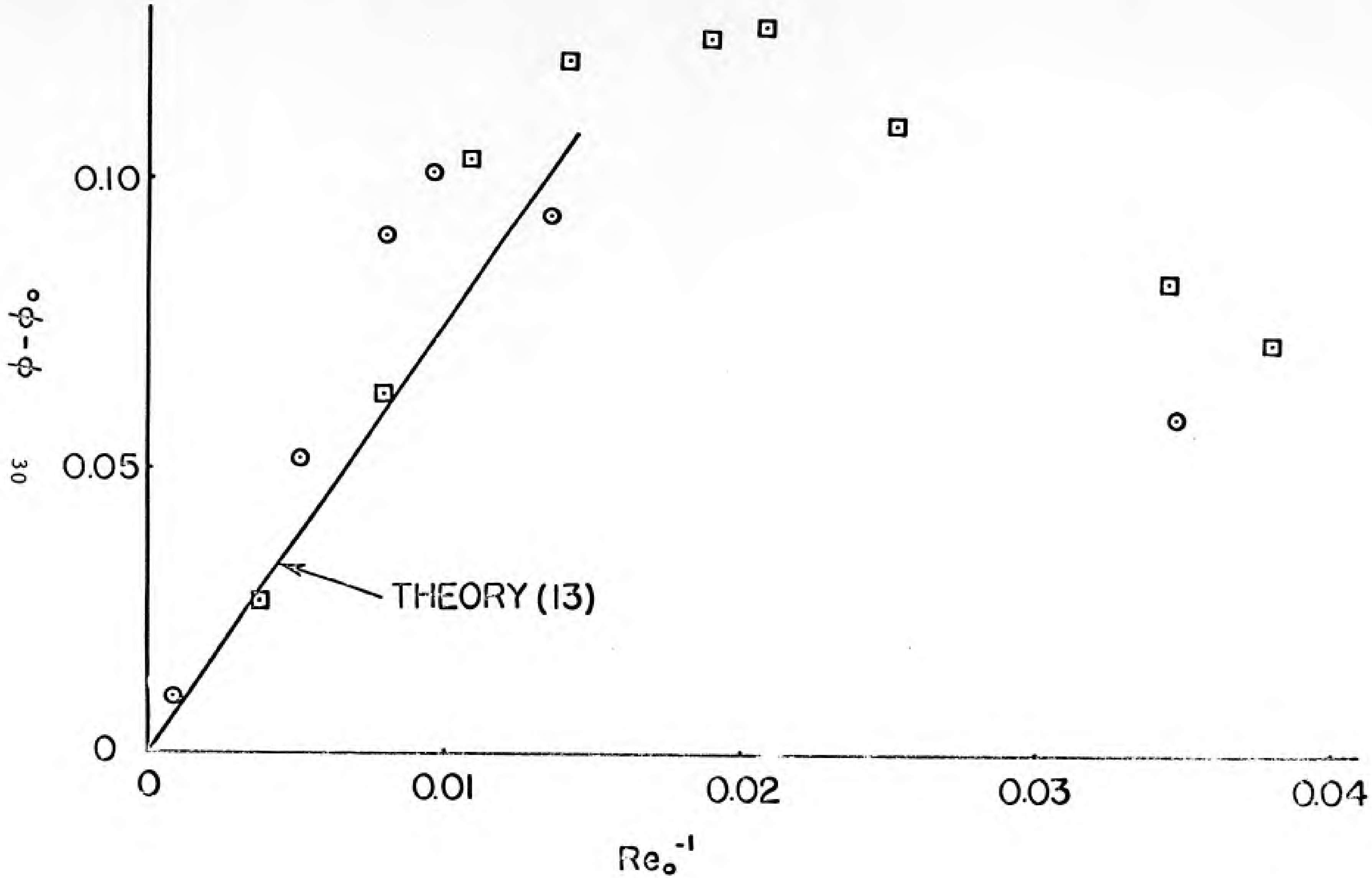

Fig. 12. Comparison of experimental measurements of diffusive separation of argon-helium mixtures with theoretical predictions. Circles indicate results for a converging nozzle ; squares indicate results for a sharp-edged orifice.

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13. ABSTRACT

Measurements of axial and radial velocity distributions in axisymmetric free jets of pure gases and binary gas mixtures are reported. Results are compared with extensions of the kinetic theory approach developed by Hamel and Willis and by Edwards and Cheng. The use of binary gas mixtures to produce molecular beams in the 1-10 ev range is described. Measurements of diffusive separation of mixtures in free jets are reported and compared with the theoretical predictions of Zigan and Sherman for nearly continuum flow.

14. KEY WORDS	LINK A		LINK B		LINK C	
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freely expanding jets						
molecular beams						
kinetic theory						
nonequilibrium flow						
rarefied gas dynamics						
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