Talk presented to the Second International
Symposium on High and Intermediate Energy
Molecular Beams, Cannes, France
July 1-4, 1969

UCRL-19034
Preprint

Cy. 2

CHARACTERIZATION OF MULTICHANNEL SOURCES AND THEIR UTILIZATION IN MOLECULAR BEAM SYSTEMS

RECEIVED
LAWRENCE
RADIATION LABORATORY

JUL 16 1969

LIBRARY AND
DOCUMENTS SECTION

D. R. Olander and R. H. Jones

June 1969

AEC Contract No. W-7405-eng-48

TWO-WEEK LOAN COPY

This is a Library Circulating Copy
which may be borrowed for two weeks.
For a personal retention copy, call
Tech. Info. Division, Ext. 5545

LAWRENCE RADIATION LABORATORY
UNIVERSITY of CALIFORNIA BERKELEY

UCRL-19034 Cy. 2

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

Second International Symposium
on High and Intermediate Energy Molecular Beams
Cannes, France, July, 1969

CHARACTERIZATION OF MULTICHANNEL SOURCES AND THEIR UTILIZATION IN MOLECULAR BEAM SYSTEMS

by D.R. Olander and R.H. Jones

Inorganic Materials Research Division of the
Lawrence Radiation Laboratory and the
Department of Nuclear Engineering,
University of California, Berkeley, California 94720

ABSTRACT

Multichannel arrays have become increasingly popular sources for generating molecular beams for a variety of beam surface and crossed beam experiments. The properties of a molecular beam from a channel source are calculated using a model which regards intermolecular collisions in the channel as a perturbation of the distribution function for free molecule flow. The angular distribution is predicted to be less peaked than that from the same source under free molecule flow conditions. The velocity distribution is harder than a Maxwellian. Experimental data supporting these predictions are reviewed.

Les réseaux multicanaux sont devenus des sources de plus en plus populaires pour obtenir des faisceaux moléculaires en vue d'expériences diverses sur l'interaction entre faisceau et surface et à faisceaux croisés. Les propriétés d'un faisceau moléculaire issu d'une source canal sont calculées en utilisant un modèle dans lequel les collisions intermoléculaires dans le canal sont considérées comme une perturbation de la fonction de distribution pour un écoulement de molecules libres. On prévoit que la distribution

angulaire doit présenter un pic moins prononcé que celle de la même source dans des conditions d'écoulement de libres molécules. La distribution des vitesses est durcie par rapport à une distribution Maxwellienne. Les données expérimentales supportant ces prédictions sont passées en revue.

INTRODUCTION

Within the past decade, beam-surface scattering studies and crossed beam chemical kinetic investigations have become important tools for exploring gas-gas and gas-solid interactions. These experiments require the primary molecular beam to interact with another entity (i.e., another beam or a surface) before detection. Because of the attenuation due to the introduction of another element between the beam source and the detector, the problem of signal level has become crucial.

Early experiments with molecular beams utilized thin-walled orifices operated at source pressures low enough to insure free molecule flow conditions. These "effusive orifices" provide a beam well characterized in terms of total flow and angular and velocity distributions, but of limited intensity. Increasing source pressure to improve beam strength results in departures from truly effusive flow with accompanying changes in the beam characteristics. With increasing source pressure, the nature of the flow in the beam-forming portion of the source gradually changes from free molecule flow to a poorly understood transition region and finally, at very high source pressures, to hydrodynamic flow of a nozzle or aerodynamic beam. The transition flow regime is analogous to the liquid state, in the sense that theoretical understanding of this region is far behind the understanding of the "rarefied" and "dense" states on either side.

The characterization of nozzle beams in greatly aided by the applicability of isentropic continuum flow theory, which permits prediction of the total flow rate with some confidence. In a fully expanded nozzle beam, all of the random thermal motion of the source molecules is converted to directed translational motion, and the beam is mono-energetic. The major drawback of present nozzle beam sources is the very large primary vacuum pumping system required to handle the substantial gas load from the source.

A practical compromise between the large, high intensity aerodynamic beam systems and the small but low intensity purely effusive systems is the multichannel array source. A number of laboratories have selected multichannel sources for experiments in crossed beam or beam-surface studies. A typical apparatus is shown in Fig. 1. Multichannel sources can provide adequate beam strength with modest pumping requirements. Their main disadvantage is the lack of experimental information and a theoretical foundation for

predicting the characteristics of the molecular beams formed by such sources.

BEAM CHARACTERISTICS

The properties of a molecular beam, which include total flow rate, angular distribution, and velocity spectrum, would be completely specified if the function $j(\theta,v)$, which is the beam intensity in molecules/sec-sr-unit speed at an angle θ from the beam axis and at a speed v, were known.

Fortunately, molecular beams are well collimated and a complete knowledge of $j(\theta,v)$ is unnecessary. We require only the total flow rate and centerline properties ($\theta=0$) which are obtained by integration of $j(\theta,v)$:

Total flow rate:

$$\ell = 2\pi \int_0^1 d(\cos\theta) \int_0^\infty dv\, j(\theta,v) \tag{1}$$

Centerline intensity:

$$J(0) = \int_0^\infty j(0,v)\,dv \tag{2}$$

Normalized number density speed spectrum along beam axis:

$$f(v) = \frac{j(0,v)/v}{\int_0^\infty j(0,v)\,dv/v} \tag{3}$$

The angular flux distribution, while not a centerline quantity, is directly measurable and gives important information for collimator design and system alignment:

$$J(\theta) = \int_0^\infty j(\theta,v)\,dv \tag{4}$$

For a truly Maxwellian beam from an ideal orifice of radius a: (denoted by *):

$$j^*(\theta,v) = \frac{1}{4}\, n_s a^2 v \cos\theta f_M(v) \tag{5}$$

where n_s is the number density in the source and $f_M(v)$ is the normalized Maxwellian speed distribution:

$$f_M(v) = \frac{4}{\sqrt{\pi}} \frac{v^2}{\alpha^3} e^{-(v/\alpha)^2} \tag{6}$$

where α is the most probable speed:

$$\alpha = \left(\frac{2kT_s}{m}\right)^{1/2} \tag{7}$$

where T_s is the source temperature and m the molecular mass. Eq(5) yields the well-known formula for effusive flow:

$$\ell^* = \frac{1}{4} n_s \bar{v}_s (\pi a^2) \tag{8}$$

where the mean speed is:

$$\bar{v}_s = \left(\frac{8kT_s}{\pi m}\right)^{1/2} \tag{9}$$

The other derived quantities for a beam from an effusive orifice are:

$$J^*(\Theta) = \frac{\ell^*}{\pi} \cos\Theta \tag{10}$$

$$J^*(0) = \ell^*/\pi \tag{11}$$

$$f^*(v) = f_M(v) \tag{12}$$

CHANNEL BEAMS NEAR THE MAXWELLIAN LIMIT

Most multichannel sources are operated in a region which is closer to the free molecule limit than the nozzle beam limit. In this flow regime, the flow rate through each channel may be expressed by the Knudsen formula:

$$\ell = K\ell^* \tag{13}$$

where K is the Clausing factor of a channel. The total flow from a source of N channels in N times the value given by Eq(13). The validity of this relation depends upon the ratio of the source driving pressure, p_s, to the pressure at which the mean free path in the source equals the channel diameter; the latter is:

$$p^* = \frac{kT_s}{\sqrt{2}\pi\sigma^2(2a)} \tag{14}$$

where a is the channel radius and σ is the molecular diameter. Eq(13) appears to be a valid description of the total flow rate up to reduced source pressures p_s/p^* (which is the inverse of the entrance Knudsen number) of about 10(1,2).

The angular and velocity distributions, however, begin to diverge from the free molecule limit at considerably lower source pressures. These features of the flow are much more strongly influenced by intermolecular collisions within the channel than is the total flow rate. While the applicability of Eq(13) is determined by a reduced source pressure based upon channel diameter, the angular and velocity distributions depend upon the reduced pressure based upon channel length, or p_s/p_L^*, where:

$$p_L^* = \frac{kT_s}{\sqrt{2}\pi\sigma^2 L} \tag{15}$$

It can be shown that the free molecule angular and velocity behavior is attained when $p_s/p_L^* \lesssim 0.1$. Since the length of most multichannel sources is considerably greater than the diameter of the tubes in the source, much lower source pressures are required to achieve free molecule angular and velocity behavior than to attain the effusion rate characteristic of Knudsen flow.

In our analysis of the characteristics of the flow in multichannel molecular beam sources, we assume that the source pressures are low enough for the Knudsen formula, Eq(13), to remain a valid description of the total flow rate. Each channel is considered to behave independently of the others, and free molecule flow is presumed to prevail downstream of the source exit. In developing perturbations to the free molecule flow formula for the centerline intensity, we have utilized the ideas of Giordmaine and Wang (3) and the concept of a velocity-dependent mean free path. Giordmaine and Wang offered an explanation of the angular distribution from long tubes at pressures slightly above the free molecule limit by considering two processes:

(1) Intermolecular collisions between non-directed gas in the channel and molecules moving along the beam axis.

(2) Intermolecular collisions between gas molecules in the channel which produce molecules directed along the beam axis.

To this framework we have added the concept of a velocity dependent mean free path, which has been successfully used

by Manista (4) to calculate the hardening of the speed spectrum of a molecular beam traversing a background scattering gas. The velocity dependent mean free path is applied to the attenuation of the straight-ahead molecules within the beam-forming channel. The flux along the centerline of the beam from a single channel is given by (5):

$$j(0,z) = \frac{1}{4} n_s a^2 \alpha z f_M(z) P[\Theta(z),\gamma] \tag{16}$$

where z is the molecular speed relative to the most probable speed:

$$z = v/\alpha \tag{17}$$

and $f_M(z)$ is the analog of Eq(6) in terms of the reduced speed:

$$f_M(z) = \frac{4}{\sqrt{\pi}} z^2 e^{-z^2} \tag{18}$$

The perturbation function P results from intermolecular collisions within the channel which preferentially deplete the beam of its low speed molecules:

$$P[\Theta(z),\gamma] = \frac{1}{\Theta(z)} \left\{ [\Theta(z)-1]e^{-\gamma\Theta(z)} + \frac{\sqrt{\pi}}{2} \frac{\mathrm{erf}[\gamma\Theta(z)]^{1/2}}{[\gamma\Theta(z)]^{1/2}} \right\} \tag{19}$$

where

$$\gamma = \frac{1}{2} (p_s/p_L^*) \tag{20}$$

and

$$\Theta(z) = \frac{ze^{-z^2} + \frac{\sqrt{\pi}}{2}(1+2z^2)\mathrm{erf}(z)}{\sqrt{2\pi}z^2} \tag{21}$$

The normalized speed spectrum of the beam can be obtained by inserting Eq(16) into Eq(3):

$$f(z) = \frac{f_M(z)P[\Theta(z),\gamma]}{\int_0^\infty f_M(z)P[\Theta(z),\gamma]dz} \tag{22}$$

Spectra for several values of the parameter γ are plotted in Fig. 2. For source pressures such that $\gamma \gtrsim 0.05$, the distribution begins to deviate from a Maxwellian. Between $\gamma = 0.05$ and $\gamma = 5$, the spectrum progressively hardens as intermolecular collisions within the tube remove more slow speed molecules than fast ones. A "saturation" hardening is attained at $\gamma = \sim 5$. This equilibrium shape results from a balance between removal of molecules of a particular speed by collisions and replenishment by collisions which produce a molecule of the same speed. The phenomenon is similar to the photon spectrum equilibrium which occurs with deep penetration of gamma rays in a nuclear reactor shield.

The theory outlined above makes no attempt to predict the complete distribution function $j(\Theta,z)$.

THE PEAKING FACTOR

A convenient parameter for molecular beam design purposes is the "peaking factor" defined as the ratio of the centerline intensity from the channel source to the centerline intensity from an ideal thin-walled orifice source emitting at the same total flow rate. This ratio can be written as:

$$\chi \equiv \frac{J(0)/\ell}{J^*(0)/\ell^*} = \frac{\pi}{\ell} J(0) \tag{23}$$

The maximum value of the peaking factor is attained at very low source pressures when no intermolecular collisions occur:

$$\chi_{max} = 1/K \tag{24}$$

The degradation of the peaking factor which occurs as the source pressure is increased is due to intermolecular collisions within the tube. With the aid of Eqs(2), (23), (8), and (13), the peaking factor predicted by Eq(16) can be written as:

$$\frac{\chi}{\chi_{max}} = \frac{\sqrt{\pi}}{2} \int_0^\infty z f_M(z) P[\Theta(z),\gamma] dz \tag{25}$$

The calculation of Giordmaine and Wang (3) did not consider a velocity dependent mean free path. Their result may be obtained by setting $\Theta(z)$ equal to unity in Eq(19), which yields:

$$\left(\frac{\chi}{\chi_{max}}\right)_{G-W} = P(1,\gamma) = \frac{\sqrt{\pi}}{2}\,\frac{\mathrm{erf}\sqrt{\gamma}}{\sqrt{\gamma}} \tag{26}$$

The angular distribution of the flux of helium from a glass tube bundle source fabricated by fiber optics techniques is shown in Fig. 3. This source contained 4300 channels each 0.025 cm long and 11 microns in diameter contained in a circle of 1 mm diameter. For comparison, the distribution from a cosine emitter at the same total flow is also shown. The multichannel source produces a centerline beam intensity 12.6 times greater than that of the cosine source. At low source pressures, the peaking factor approaches the expected limit for a transparent tube, namely, the reciprocal of the Clausing factor. This significant improvement over a cosine emitter, however, is not sustained at high source pressures. Fig. 4 shows the variation in the peaking factor with source pressure, made dimensionless with respect to the pressure at which the mean free path is equal to the channel diameter. When the reduced source pressure approaches unity, the peaking factor has decreased to approximately 5.

Agreement between the data and Eq(26) is generally satisfactory. The discrepancy at low pressures is believed due to (1) inability to precisely align the beam detector on the tip of the very sharp distribution of Fig. 3 and, (2) finite angular resolution of the detector.

The accord between theory and experiment suggests that peaking factor calculations from the Giordmaine-Wang theory can be used to estimate centerline intensity of the molecular beam provided that the reduced source pressure (based on tube diameter) is not much greater than unity. At high source pressures, the peaking factor approaches a limiting value for a fully expanded nozzle beam (6), which has been indicated on Fig. 4.

The limiting feature of most molecular beam systems is the total flow rate from the source. Arbitrarily large flow rates cannot be handled by the pump evacuating the chamber containing the source. High pressures in the source chamber will either contaminate the molecular beam by effusion of background gas through the collimating orifice or reduce its intensity by intermolecular collisions upstream of the collimator. Once the maximum allowable flow rate from the source is determined, the system design and operating pressure of the source can be adjusted to maximize the centerline intensity, thereby yielding the strongest molecular beam for experimental purposes. In view of the results discussed in the previous paragraph, it would seem desirable to operate at a very low source pressure to max-

imize the peaking factor. However, the gross diameter of the source needed to produce the required total flow rate would be excessive, and the source could not be considered as a point.

The effects of the gross diameter of a molecular beam source and its alignment in the system upon source efficiency have been investigated analytically. (7) If the diameter of the source approaches that of the collimating orifice used to form the molecular beam, the peaking factor may be reduced by 50% or more. Reasonable displacement and tilt misalignments can be tolerated before source efficiency is substantially reduced. The effects of size and misalignment are more critical for multichannel sources with highly peaked angular distributions than for cosine sources.

EQUIVALENT BEAM PROPERTIES

One is often interested in comparing the results of molecular beam experiments with results obtained from conventional experiments utilizing a non-directed gas. Since the thermodynamic state of a random gas is completely characterized by its pressure and temperature, interpretation of molecular beam experiments requires knowledge of the equivalent pressure and equivalent temperature of the beam. We consider here the calculation of these equivalent properties only for the case of beam-surface interaction; beam-beam interactions require only minor modification.

EQUIVALENT BEAM PRESSURE

The equivalent pressure of a molecular beam striking a solid target is defined as that pressure of a non-directed gas at the temperature of the source which would yield the same impingement rate as the molecular beam. The rate at which the beam impinges on a unit area of target at an axial distance x from the source is $J(0)/x^2$. The rate at which molecules from a random gas impinge upon a unit surface area is

$$\frac{1}{4}\,(p_{eq}\bar{v}_s/kT_s).$$

Equating these rates yields:

$$p_{eq} = \frac{4kT_s}{x^2\bar{v}_s}\,J(0) \tag{27}$$

The equivalent pressure may be conveniently expressed in terms of the peaking factor by:

$$p_{eq} = \chi K N \left(\frac{a}{x}\right)^2 p_s \tag{28}$$

The ratio of the equivalent pressure of the molecular beam to the source pressure is seen to be the product of three factors:*

(1) The peaking factor χ which represents the increased directivity of the channel source compared to a cosine emitter.

(2) The Clausing factor K, which represents the reduction in flux due to the geometry of a channel.

(3) The geometric factor $N(a/x)^2$, which reflects the dimension of the channels to the length of the beam path in the vacuum system multiplied by the number of channels in the array.

Factors (2) and (3) depend only on the geometry of the beam source and the experimental vacuum chambers and are easily determined. The peaking factor may be estimated by the considerations discussed previously.

EQUIVALENT BEAM TEMPERATURE

The average translational energy of the beam molecules is of particular interest in molecular beam experiments. We would like to be able to relate the average energy to the source temperature in a manner similar to that for a truly effusive beam. The average translational energy of a beam molecule striking the target is:

$$\bar{\varepsilon} = \frac{1}{2} m\alpha^2 \frac{\int_0^\infty z^3 f(z)\,dz}{\int_0^\infty z f(z)\,dz} \tag{29}$$

If the speed distribution in the beam, f(z), is Maxwellian, application of Eq(18) to Eq(29) yields:

$$\bar{\varepsilon}_M = m\alpha^2 \tag{30}$$

Since the average energy of the molecules of a random gas is proportional to the gas temperature, an equivalent beam temperature is defined by:

---

* The equivalent pressure of the beam in terms of number density rather than impingement rate is 1/4 of the value given by Eq(28).

$$\frac{T_{eq}}{T_s} = \frac{\bar{\varepsilon}}{\bar{\varepsilon}_M} = \frac{\int_0^\infty z^3 f_M(z) P[\Theta(z),\gamma]dz}{2\int_0^\infty z f_M(z) P[\Theta(z),\gamma]dz} \tag{31}$$

where f(z) has been expressed by Eq(22).

Speed distributions in a molecular beam are generally determined by velocity selection (8) or time-of-flight techniques (9). Our experiments (5) were of the time-of-flight variety but utilized symmetric modulation and lock-in detection. The attenuation of the amplitude and the shift of the phase angle of the signal were measured as the speed of a mechanical chopper interrupting the beam was varied. For the purpose of inverting the data from frequency space to speed space, a two-parameter distribution function (a drifting Maxwellian) was utilized. The equivalent beam temperatures were computed from the best fit parameters to this empirical distribution function. Measurements were made with a single channel source, 0.48 cm long and 0.038 cm in diameter for both hydrogen and nitrogen gas. The results are compared to the predictions of Eq(31) on Fig. 5. Despite the scatter of the experimental points, the expected hardening of ∿ 15% is observed.

CONCLUSIONS

The properties of near-Maxwellian molecular beams from channel sources have been analyzed using a collision model based upon a combination of the calculations of Giordmaine and Wang (3) and Manista (4). The model predicts a decrease in the peaking factor with increasing source pressure and a concommitant hardening of the centerline speed spectrum. The available data supports these implications.

The advantages of a multichannel array source over a cosine emitter are embodied in the peaking factor. It appears that practical molecular beam sources fabricated from multi-channel arrays cannot achieve their maximum single channel peaking factors. When operated at source pressures as low as possible commensurate with source size, they may still be able to provide a 5-6 fold improvement in average beam intensity, but probably never will attain the order of magnitude increase which was originally anticipated.

This work was performed under the auspices of the United States Atomic Energy Commission.

REFERENCES

1. M. Fryer, Trans. Faraday. Soc., 293A, 329 (1966).

2. R.H. Jones, V.R. Kruger, and D.R. Olander, USAEC report UCRL-17859 (1968).

3. J.A. Giordmaine and T.C. Wang, J. Appl. Phys., 31, 463 (1960).

4. E.J. Manista, NASA TN D-2617 (1965).

5. R.H. Jones, D.R. Olander, and W. Siekhaus, to be published.

6. R.H. Jones, D.R. Olander, and V. Kruger, J. Appl. Phys., to be published.

7. D.R. Olander, J. Appl. Phys., to be published.

8. K.C. Wang and P.G. Wahlbeck, J. Chem. Phys., 49, 1617 (1968).

9. J.B. Anderson, R.P. Andres, and J.B. Fenn, Adv. in Atomic and Molecular Phys., 1, (Academic Press, 1965), p. 345.

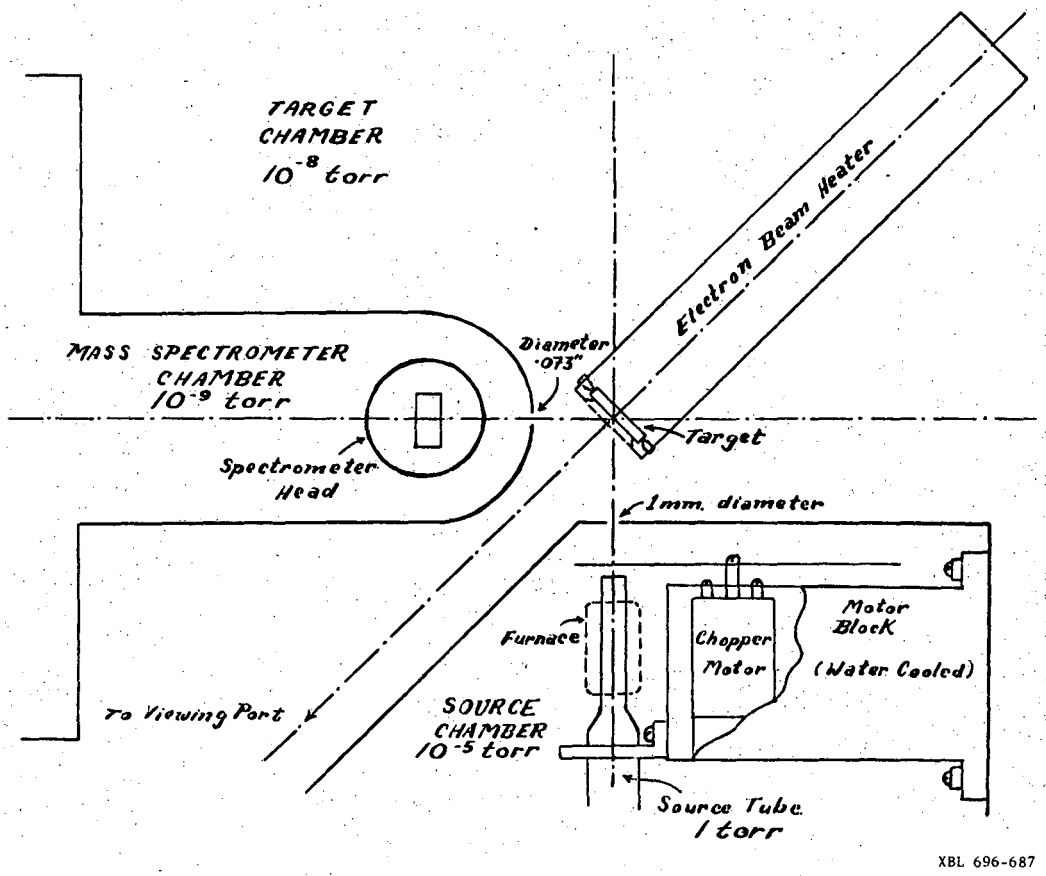
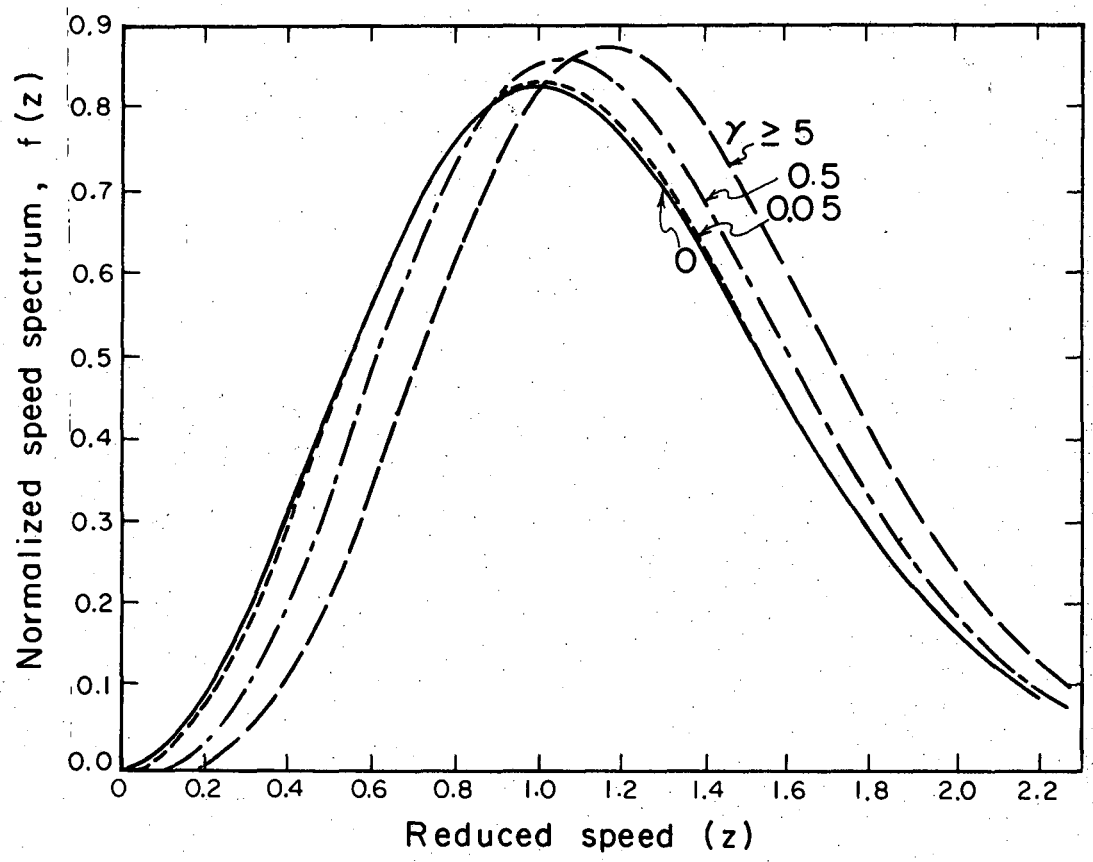
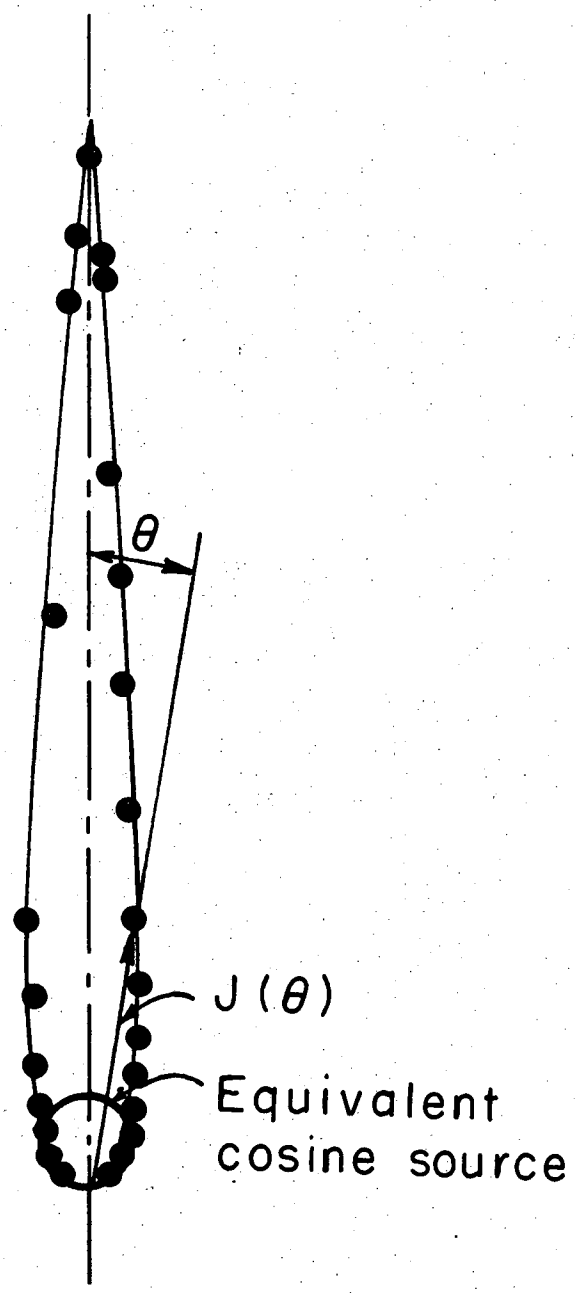


FIG. 1: Molecular beam apparatus for beam-solid reaction studies.



XBL696-2979

FIG. 2: Centerline speed spectrum from channel source.



XBL6811-7185

FIG. 3: Angular distribution of flux from multichannel source.

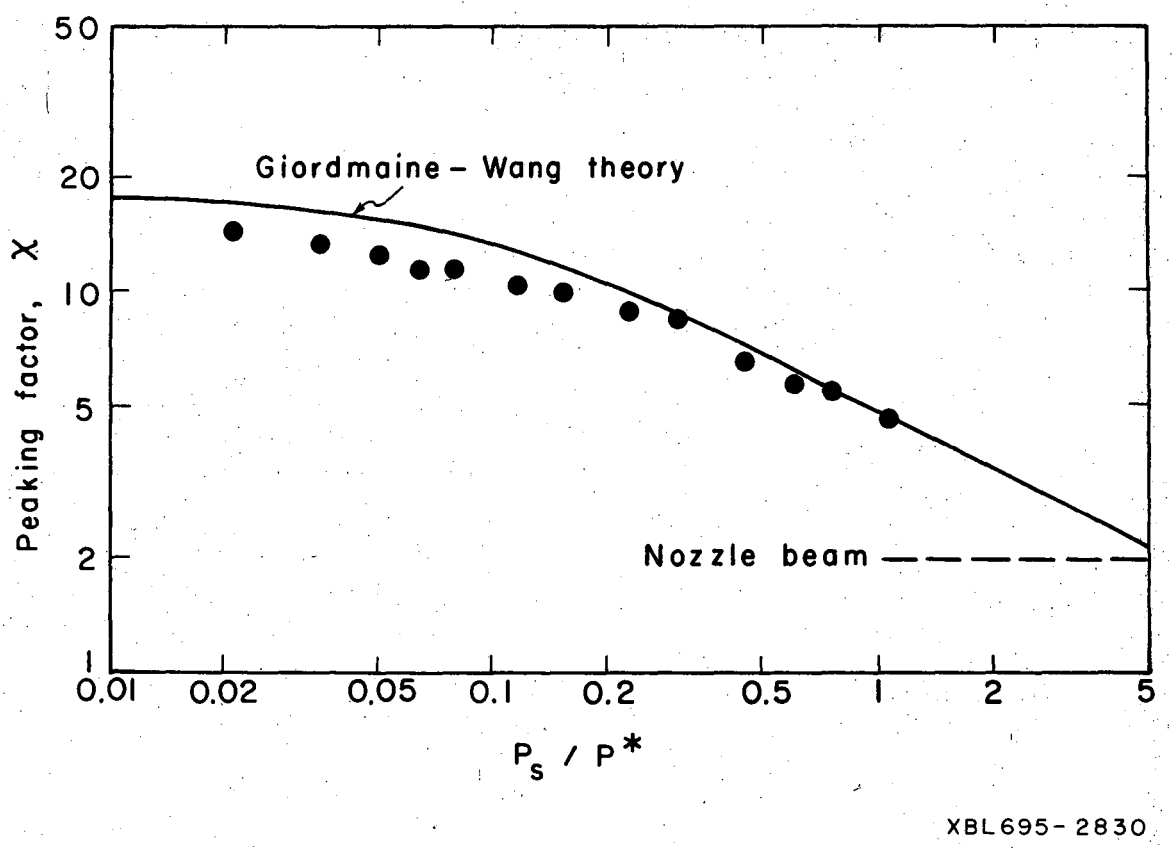
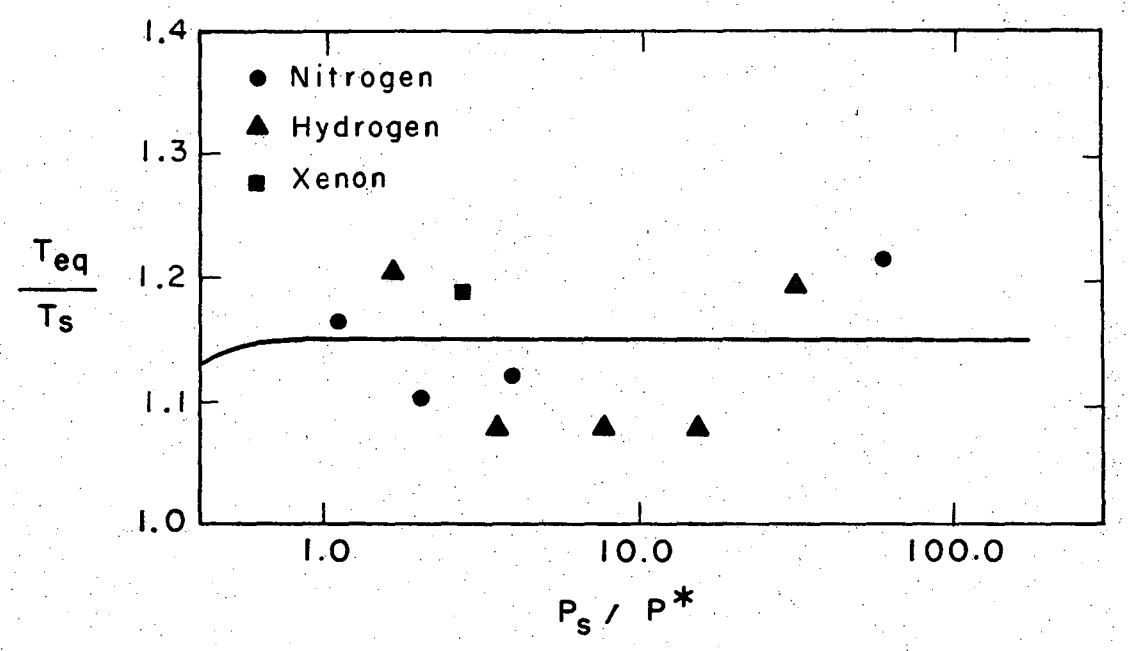


FIG. 4: Variation of peaking factor with source pressure.



XBL696-2978

FIG. 5: Variation of equivalent beam temperature with source pressure.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

TECHNICAL INFORMATION DIVISION
LAWRENCE RADIATION LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720