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Molecular Beam Sources Fabricated from Multichannel Arrays: III The Exit Density Problem

by

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ABSTRACT

Although the theory of Giordmaine and Wang adequately predicts the centerline intensity of a molecular beam from a channel source, it is less successful in describing the angular distribution. This deficiency has been ascribed to a non-zero number density at the tube exit. The end conditions chosen by previous workers lead to angular distributions which fail to satisfy total flow and average cosine restrictions. A method of choosing the parameters of the linear density profile which satisfied these integral constraints is described. The computed angular distributions according to the theories utilizing different end conditions differ very little from each other, but represent a clear improvement over Giordmaine and Wang's angular distribution.

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As evidenced by the results reported in the first paper of this series (1) the centerline intensity of the molecular beam is quite adequately predicted by the theory originally proposed by Giordmaine and Wang (2). This theory assumes that the number density in the tube decreases linearly from the density in the reservoir to zero at the tube exit. While this simplification is satisfactory for centerline properties, it is less successful in predicting the off-axis beam intensity. As noted by Giordmaine and Wang, the deficiency in the theory lies in the assumption of zero density at the tube exit. There have been a number of attempts to modify the theory to rectify this fault (3-5), and it is the purpose of this paper to show that: (1) although an end density other than zero is required to adequately match experiment, the angular distribution is relatively insensitive to the particular value utilized, (2) previous end density calculations suffered from internal inconsistencies (e.g., the integral of the angular distribution did not equal the total flow rate) and (3) an internally consistent linear density profile can be determined.

To place the question of entrance and exit densities in its proper perspective, we first review collision-free flow in a cylindrical tube, since this limiting case forms the basis of the attempts to rectify the Giordmaine and Wang theory.

Collision-Free Flow in Cylindrical Tubes

In the absence of collisions between molecules, the properties of the flow of gases through cylindrical tubes are determined solely by geometrical considerations. The transmission probability (or Clausing factor) of the tube is defined by:

$$\dot{N} = K\, \nu_s (\pi a^2) \tag{1}$$

where $\dot{N}$ is the total flow rate through the tube, K is the transmission probability, a the tube radius and ν_s the ~~wall collision~~ rate at which ~~density~~ molecules in the gas reservoir ~~driving the flow~~ enter a unit area of the tube. Since the gas in the reservoir is Maxwellian, ν_s is related to the number density in the source, n_s, by:

$$\nu_s = \frac{1}{4} n_s \bar{v} \tag{2}$$

where $\bar{v}$ is the mean speed of the molecules.

In analagous fashion, the angular distribution of the molecules leaving the tube exit into the vacuum defines a distribution function $j(\theta)$ given by:

$$J(\theta) = \frac{\nu_s}{\pi} (\pi a^2)\, j(\theta) \tag{3}$$

where $J(\theta)$ is the intensity in molecules/sec-sr at a polar angle θ from the tube axis. For an ideal thin-walled orifice, $j(\theta) = \cos\theta$. ~~Since~~ for all tubes in collision-free flow, $j(0)=1$, $j(\theta)=J(\theta)/J(0)$.

Since the integral of $J(\theta)$ over the forward hemisphere must equal the total flow rate, K and $j(\theta)$ are related by:

$$K = 2 \int_0^1 j(\theta)\, d\,(\cos\theta) \tag{4}$$

Both K and $j(\theta)$ are dependent only upon the length-to-diameter ratio of the tube,

$$\gamma = \frac{2a}{L} \tag{5}$$

where L is the length of the tube.

Both K and $j(\theta)$ are determined once the collision ~~density~~ rate on the tube walls is known. If x is the distance from the entrance measured in units of tube length, $\nu(x)$ denotes the rate at which molecules strike a unit area of wall at location x. Assuming the law of diffuse reflection to apply to the molecule-wall interaction, $\nu(x)$ is the solution to the integral equation (7, 8, 4):

$$\frac{\nu(x)}{\nu_s} = \int_0^1 \frac{\nu(x')}{\nu_s} \left\{ 1 - \frac{|x'-x|}{\left[\gamma^2 + (x'-x)^2\right]^{1/2}} - \frac{\gamma^2 |x'-x|}{2\left[\gamma^2 + (x'-x)^2\right]^{3/2}} \right\} \frac{dx'}{\gamma} + \frac{1}{\gamma} \left\{ \frac{(\gamma^2/2) + x^2}{(\gamma^2 + x^2)^{1/2}} - x \right\} \tag{6}$$

Clausing (7) and Demarcus (8) have obtained numerical solutions to Eq. (6). The transmission probability was determined directly, and not by first determining $j(\theta)$ and then K from Eq. (4).

The function $\nu(x)$ determined by Eq. (6) is nearly linear over the tube length, except for regions close to the entrance and exit. All angular distribution functions, $j(\theta)$, have been computed by the use of the approximate formula:

$$\frac{\nu(x)}{\nu_s} = \zeta_1 - (\zeta_1 - \zeta_o)x \tag{7}$$

where the constants ζ_1 and ζ_o denote the dimensionless tube wall collision rates at the entrance and exit of the tube respectively.

The distribution function $j(\theta)$ includes contributions from the source reservoir and from the walls. The computational method described by Clausing (9) yields:

$$j(\theta) = \zeta_o \cos\theta + \frac{2}{\pi}\cos\theta\left[(1-\zeta_o)\, R(p) + \frac{2}{3}(\zeta_1-\zeta_o)\frac{1-(1-p^2)^{3/2}}{p}\right], \quad (8)$$

$$\text{for } \tan\theta \leqslant \gamma$$

and

$$j(\theta) = \zeta_o \cos\theta + \frac{4\gamma}{3\pi}(\zeta_1-\zeta_o)\frac{\cos^2\theta}{\sin\theta}, \quad \text{for } \tan\theta \geqslant \gamma \quad (9)$$

In these formulae,

$$R(p) = \cos^{-1}p - p\sqrt{1-p^2} \quad (10)$$

$$p = \tan\theta/\gamma \quad (11)$$

The adequacy of this angular distribution for specified ζ_o and ζ_1 can be verified by Eq. (4). This procedure is not equivalent to direct determination of K, since the formula for $j(\theta)$ is based upon a linear approximation to the wall collision ~~density~~ rate profile.

For short tubes ($\gamma > 1$), Clausing (9) uses the values:

$$\zeta_o = \frac{\sqrt{1+\gamma^2} - 1}{\gamma+\gamma^2/\sqrt{1+\gamma^2}} \quad (12a)$$

$$\zeta_1 = 1 - \zeta_o \quad (12b)$$

~~Note that there is~~ Eq (12b) implies a discontinuity in the collision ~~density~~ rate from the value at the tube entrance (ν_s) to the value on the walls just inside the entrance [$\nu_s(1-\zeta_o)$].

Using these values of ζ_o and ζ_1 in Eq. (8) and (9), Clausing (9) has performed the integration of Eq. (4) for $\gamma=1$ to yield $K = 0.512$, which is in good agreement with the value of 0.5136 obtained by ~~direct means (7, 8)~~ the exact calculations of refs 7 and 8.

In the limiting case of very long tubes ($\gamma \to 0$), the angular distribution is given by Eq. (9). Integration according to Eq. (4) yields:

$$K = \zeta_o + \frac{2}{3}(\zeta_1 - \zeta_o)\gamma \tag{13}$$

It can be shown rigorously that as $\gamma \to 0$, $K \to 4\gamma/3$ (8). The discontinuity in the collision ~~density~~ rate at the tube entrance in short tubes is due to the substantial fraction of the molecules which enter the tube that escape into the vacuum (i.e., the large transmission probability). Increasing the length of the tube is equivalent to reducing the outflow by a restriction at the exit. In the limit as the flow down the tube is completely stopped by closing the exit, the wall collision density in the tube is everywhere equal to ν_s. Consequently, ζ_1 would be expected to approach unity as $\gamma \to 0$. Eq. (13) then requires that as $\gamma \to 0$:

$$\zeta_o = \frac{2}{3}\gamma \tag{14a}$$

$$\zeta_1 = 1 \tag{14b}$$

These limiting values have been deduced by Clausing (10).

The derivation of the transmission probability due to Smoluchowski (11), which assumes only that $(d\nu/dx)$ is constant, yields:

$$\dot{N} = \frac{8a}{3L}\,(\pi a^2)\,\frac{d\nu}{dx} = \frac{4\gamma}{3}\,(\pi a^2)\,\nu_s(\zeta_1-\zeta_o) \tag{15}$$

The proper limiting value of K for long tubes follows if $\zeta_1=1$ and $\zeta_o<<1$. Note that ζ_o need not have the specific value required by Eq. (14a).

Ivanov and Troitskii (4) have attempted to determine the parameters ζ_o and ζ_1 for all γ by using the linear profile, Eq. (7) and requiring that Eq. (6) be satisfied only at x=0 and x=1. This method yields:

$$\zeta_o = \zeta_1 - \frac{(1+2/\gamma)\sqrt{1+1/\gamma^2} - (1+2/\gamma^2)}{\sqrt{1+1/\gamma^2} + 1} \tag{16a}$$

$$\zeta_1 = \left[1+\gamma/(2+\gamma^2)\right]^{-1} \tag{16b}$$

These functions, however, do not agree with Eqs. (12a) and (12b) for large γ nor with Eqs. (14a) and (14b) as $\gamma\to 0$.

In a recent analysis, Zugenmaier (3) made the following assumption covering the behavior of the parameters ζ_o and ζ_1 for tubes of arbitrary length in collision-free flow:

$$\zeta_1 = 1 \tag{17a}$$

$$\zeta_o = K/2 \tag{17b}$$

where K is not the transmission probability calculated by Clausing and Demarcus, but is determined by use of these values of ζ_o and ζ_1 in Eqs. (8) and (9) and integrating according to Eq. (4):

$$K = \frac{4}{2+k} \tag{18}$$

where

$$k = \left(\frac{1}{3}\gamma + 2\gamma u\right)^{-1} \tag{19}$$

$$u = \frac{1}{4\gamma} + \frac{1 - (1+\gamma^2)^{3/2}}{6\gamma^3} \tag{20}$$

Although Eqs. (17a) and (17b) agree with the long tube limits, Eqs. (14a) and (14b), and in the limit $\gamma \to \infty$, $\zeta_o \to 1/2$ in agreement with Eq. (12a), Zugenmaier's ζ_1 does not exhibit the discontinuity calculated by Clausing. Consequently, the transmission probability from Eqs. (18) - (20) is in disagreement with the value calculated by Clausing and Demarcus (by as much as 20% at $\gamma=1$). Recent measurements agreed with the latter values to within several percent (12).

Influence of Intermolecular Collisions on the Angular Distribution

As the pressure in the reservoir driving the flow through the tube is increased, intermolecular collisions become of comparable importance to molecule-wall collisions. The first property to be affected is the angular distribution, $j(\theta)$. Until the driving pressure is sufficiently great to result in significant hydrodynamic effects, however, the total flow can be described by Eq. (1) with a transmission probability based upon collision-free flow.

Quantitatively, the effect of intermolecular collisions is to add another dimensionless parameter to the theoretical expression for $j(\theta)$. For short tubes ($\gamma>1$), the most convenient parameter is the Knudsen number based upon tube diameter:

$$Kn_D = \frac{\lambda_s}{2a} = \frac{n^*}{n_s} \tag{21}$$

For long tubes, the Knudsen number based upon tube length is important

$$Kn_L = \frac{\lambda_s}{L} = \gamma\frac{n^*}{n_s} \tag{22}$$

In these formulae, λ_s is the mean free path of the molecules in the source reservoir and n^* is the density at which the mean free path is equal to the tube diameter.

For tubes with $\gamma \ll 1$, the angular distribution and centerline intensity begin to deviate from the collision-free values at $Kn_L < \sim 10$. The transmission probability of Clausing, however, predicts the total flow to $Kn_D \sim 1$ (~~12~~ 13, ~~13~~ 14). Thus there is a wide range of Knudsen numbers ($\gamma < Kn_L < 10$) in which intermolecular collisions affect the angular distribution but do not significantly influence the total flow. The considerations here are restricted to this range, which represents the normal operating region of molecular beam sources (excluding nozzle beams). Hydrodynamic effects are not considered.

In the Knudsen number region of interest, the transmission probability is given by the Clausing-Demarcus calculations, but the angular distribution is no longer given by Eqs. (8) and (9). The theoretical angular distribution is determined by three contributions (2,3,4):

(1) Molecules from the source reservoir which pass through the tube without colliding with the walls or with other molecules.

(2) Molecules re-emitted in a diffuse manner from the walls.

(3) Molecules scattered out of the tube exit after intermolecular collisions within the tube.

The third contribution is absent from the collision-free flow discussed previously ($Kn_L \to \infty$). The second component does not contribute to the intensity along the axis ($\theta = 0$).

In the analysis of collision-free flow, the number density of molecules within the tube is not required; knowledge of the wall

collision density, $\nu(x)$, is sufficient to fix all properties of the flow. Introduction of intermolecular collisions, however, means that the number density is required to describe the gas-phase collision frequency (in component 3). Because of attenuation in the gas phase, the wall collision ~~density~~ rate required for component 2 can no longer be described by the purely geometrical considerations which sufficed in free-molecule flow. The angular distribution is computed by guessing a number density profile along the tube and calculating the intermolecular collision frequency by the formula:

$$\nu_i(x) = \frac{1}{2}\sqrt{2}\pi\sigma^2[n(x)]^2\bar{v} \tag{23}$$

where $\nu_i(x)$ is the intermolecular collision rate (sec^{-1} - cm^{-3}) and $\pi\sigma^2$ is the collision cross section. The wall collision rate is assumed related to the number density by:

$$\nu(x) = \frac{1}{4}\, n(x)\bar{v} \tag{24}$$

From the start, the calculation is on a much more tenous basis than the analogous collision-free case, since the number density profile is simply guessed rather than calculated in an exact manner, as is $\nu(x)$ in collision-free flow (by Eq. (6)). In addition, neither Eq. (23) nor (24) are valid relations between density and collision rates; these formulae apply only to an equilibrium gas, whereas the gas flowing through a tube is neither Maxwellian in speed distribution nor isotropic in angular distribution.

Although Eq. (24) is not valid even in collision-free flow (because of non-isotropy) it very often appears in such calculations (3, 11). Its utilization is unnecessary, but does not

affect the validity of the final transmission probability or angular distribution; these quantities are proportional to the wall collision rate in the source reservoir (ν_s of Eq. (2)), which is valid. Since n(x) appears in the analysis only to determine the wall collision rate in the tube by Eq. (24), and since the governing equations are linear in n(x) [or $\nu(x)$], the constant factor $\bar{v}/4$ cancels out. Consequently, the analysis of collision-free flows rests upon the accuracy of the function $\nu(x)$, not upon the validity of the connection between $\nu(x)$ and n(x).

In order to proceed with the computation of the angular distribution, the function n(x) must be prescribed. By analogy to collision free flow, where $\nu(x)$ is known to be very nearly linear, all analyses of the flow in tubes with intermolecular collisions have assumed a function of the form:

$$\frac{n(x)}{n_s} = \xi_1 - (\xi_1 - \xi_o)x \tag{25}$$

The parameter ξ_1 denotes the ratio of the number density just inside the tube entrance to the number density in the source reservoir. ξ_o is the ratio of the number density at the tube exit to the source density. These parameters cannot be calculated from theory, as can the analogous constants ζ_o and ζ_1 appearing in Eq. (7) for the wall collision rate in free molecule flow.

The variation of ξ_1 and ξ_o with γ has been determined by a variety of approximations. Table 1 shows the transmission probabilities and normalized entrance and exit densities utilized by the five studies which have considered this problem.

Zugenmaier identified ξ_o and ξ_1 with ζ_o and ζ_1. He did not

TABLE 1. PARAMETERS OF THE LINEAR NUMBER DENSITY PROFILE USED BY VARIOUS INVESTIGATORS FOR COMPUTATION OF THE ANGULAR DISTRIBUTION FOR CYLINDRICAL TUBES WITH INTERMOLECULAR COLLISIONS

Investigators	Transmission Probability, K	ξ_1 [a]	ξ_o [a]
Giordmaine & Wang (2)	$4\gamma/3$	1	0
Zugenmaier (3)	Eqs. (18)-(20)	1	K/2
Ivanov & Troitskii (4)[b]	---	Eq. (16b)	Eq. (16a)
Jones et al (5)[c]	$(1 + 3/4\gamma)^{-1}$	$3K/4\gamma$	K/2.8
Becker (6)[c]	$(1 + 3/4\gamma)^{-1}$	1	K

[a]The transmission probabilities appearing in these columns refer to those given in the second column.

[b]Considered angular distribution, but not centerline intensity.

[c]Considered centerline intensity, but not angular distribution.

allow for a discontinuity in the number density at the tube entrance ($\xi_1 \neq 1$) despite the fact that Clausing has demonstrated an analogous discontinuity in the wall collision rate.

Although Ivanov and Troitskii computed ζ_o and ζ_1 as a function of γ for collision free flow, it is of interest to note that the value of $\xi_o = 0.6\gamma$ utilized by these authors in the long tube limit represents a compromise: as $\gamma \to 0$, ζ_o from Eq. (16a) approaches 0.5γ but a limiting value of 0.67γ is required to satisfy the restriction that the transmission coefficient for long tubes be $4\gamma/3$. Apparently the value of 0.6γ represents the average of these two figures.

The ξ_1 value used by Jones et al (5) was based upon the same considerations used in deriving the transmission probability by the Dushman method (i.e., an ideal orifice and a long tube represent series flow resistances). The exit density used by these authors was based upon experimental measurements of the average cosine of the flux from the tube.

If ξ_o and ξ_1 in Eq. (25) are considered specified, the angular distribution function $j(\theta)$ can be calculated by extending Clausing's collision-free calculation (9) to include the contribution due to intermolecular collisions. Giordmaine and Wang and Ivanov and Troitskii both utilized this technique. The general form of the angular distribution function may be written as:

$$j(\theta) = \xi_o \cos\theta + \frac{2}{\sqrt{\pi}} \frac{\xi_o \cos\theta \; e^{\delta'^2}}{\delta'} \left\{ \frac{1}{2} R(p) \left[\mathrm{erf}\left(\frac{\xi_1}{\xi_o} \delta' \right) - \mathrm{erf}(\delta') + \frac{2}{\sqrt{\pi}} \delta' \frac{\xi_1}{\xi_o} \left(\frac{1}{\xi_1} - 1 \right) e^{-(\delta' \xi_1/\xi_o)^2} \right] + S(p) \right\}, \tag{26}$$

for $\tan\theta \leq \gamma$

and

$$j(\theta) = \xi_o \cos\theta + \frac{2}{\sqrt{\pi}} \frac{\xi_o \cos\theta \; e^{\delta'^2}}{\delta'} S(1), \tag{27}$$

for $\tan\theta \geqslant \gamma$

Here p and R(p) have the same meaning as in Eqs. (10) and (11) and S(p) is given by:

$$S(p) = \int_0^p \sqrt{1-z^2} \left\{ \mathrm{erf}\left[\delta'\left(1 + \frac{\gamma z}{\tan\theta}\left(\frac{\xi_1}{\xi_o} - 1\right)\right)\right] - \mathrm{erf}(\delta') \right\} dz \tag{28}$$

$$\delta' = \delta/\sqrt{\cos\theta} \tag{29}$$

$$\delta = \left[\frac{1}{2\gamma Kn_D} \frac{\xi_o^2}{\xi_1 - \xi_o}\right]^{1/2} = \tag{30a}$$

or

$$\delta = \left[\frac{1}{2Kn_L} \frac{\xi_o^2}{\xi_1 - \xi_o}\right]^{1/2} \tag{30b}$$

The centerline intensity, j(0), is no longer unity as it is for collision-free flow, but is given by:

$$j(0) = \xi_o + \frac{\sqrt{\pi}}{2} \xi_o \frac{e^{\delta^2}}{\delta}\left[\mathrm{erf}\left(\frac{\xi_1}{\xi_o}\delta\right) - \mathrm{erf}(\delta)\right] + \left(\frac{1-\xi_1}{\xi_o}\right) \exp\left\{-\delta^2\left[\left(\frac{\xi_1}{\xi_o}\right)^2 - 1\right]\right\} \tag{31}$$

The peaking factor, utilized as a measure of centerline beam intensity in the preceeding papers of the series, is

$$\chi = j(0)/K \tag{32}$$

The angular distribution is a function of two parameters, γ and Kn_D (or Kn_L). ~~The end densities ξ_1 and ξ_o are functions of γ.~~ As Kn_D or Kn_L becomes large, Eqs. (26) and (27) reduce to Eqs. (8) and (9) and Eq. (31) becomes unity.

The angular distribution and centerline intensities derived by the first four works in Table 1 may be obtained by utilizing the particular ξ_1 and ξ_o values shown in the table (~~14~~ 15).

Integral Constraints on the Angular Distribution

As in the case of collision-free flow, the angular distributions of Eq. (26) and (27) must satisfy the total flow criterion of Eq. (4), which amounts to a constraint on the parameters ξ_o and ξ_1. None of the angular distributions based upon the ξ_o and ξ_1 values in Table 1 satisfy Eq. (4) over the entire range of γ.

Because of the prescription of the number density at the exit of the tube, an additional integral condition is required of the angular distribution: the average cosine of the flux at the exit, implied by the value of ξ_o selected, must be equal to that obtained from the angular distribution. This requirement stems from the assumption (implicit in all analyses listed in Table 1) that the flow is collision-free downstream of the tube exit. The angular distribution of the molecular beam is therefore directly related to the angular distribution of the number density at the tube exit.

By the definition of a particle current, the total flow rate per unit cross sectional area of the tube is the product of the exit density ($n_o=\xi_o n_s$) and the mean molecular velocity component along the tube axis ($\bar{v}_{xo}$)

$$\frac{\dot{N}}{\pi a^2} = n_o \bar{v}_{xo} \tag{33}$$

Let $\bar{\mu}_o$ denote the average cosine relative to the tube axis of the molecular distribution function (not the molecular beam) at the tube exit. Since the flow process is assumed not to alter the speed distribution,

$$\bar{v}_{xo} = \bar{\mu}_o \bar{v} \tag{34}$$

Combining Eqs. (1), (2), (33), and (34) yields:

$$\bar{\mu}_o = \frac{1}{4}\frac{K}{\xi_o} \tag{35}$$

The average cosine of the molecular distribution may be obtained from the angular distribution of the molecular beam:

$$\bar{\mu}_o = \frac{\int_0^1 J(\theta)\,d(\cos\theta)}{\int_0^1 \frac{J(\theta)}{\cos\theta}\,d(\cos\theta)} = \frac{K}{2\int_0^1 \frac{j(\theta)}{\cos\theta}\,d(\cos\theta)} \tag{36}$$

Equating (35) and (36) yields the condition:

$$\xi_o = \frac{1}{2}\int_0^1 \frac{j(\theta)}{\cos\theta}\,d(\cos\theta) \tag{37}$$

A detailed development of Eq. (37) is presented in the appendix.

The internal consistency condition represented by Eq. (37) applies only when the relative number density at the exit, ξ_o, is specified. There is no analogous restriction on the relative wall collision ~~density~~ rate at the exit, ζ_o.

Eqs. (4) and (37) represent integral constraints on the angular distribution function involving the zeroth and minus one moments, respectively. No arbitrarily chosen pair of parameters

ξ_o and ξ_1 can satisfy both of these restraints. The only way that a two-parameter density profile (linear or not) can be utilized in a collision model such as that which generated Eqs. (26) and (27) is to allow the total flow and average cosine restrictions to determine ξ_o and ξ_1 rather than specify these parameters a priori. In this case, the angular distribution is regarded as parametric in γ, Kn_L (or Kn_D) and two adjustable constants, ξ_o and ξ_1. Eqs. (4) and (37) provide two independent relations for the determination of ξ_o and ξ_1:

$$K = 2 \int_0^1 j(\theta, \gamma, Kn, \xi_o, \xi_1) d(\cos\theta) \tag{38}$$

and

$$\xi_o = \frac{1}{2} \int_0^1 \frac{j(\theta, \gamma, Kn, \xi_o, \xi_1)}{\cos\theta} d(\cos\theta) \tag{39}$$

The function $j(\theta, \gamma, Kn, \xi_o, \xi_1)$ is given by Eqs. (26) and (27).

For specified values of γ and Kn_L or Kn_D, Eqs. (38) and (39) can be solved simultaneously for ξ_o and ξ_1. The transmission probability is regarded as a known function of γ, given by the Clausing-Demarcus calculations. Figs. 1 and 2 show the values of ξ_1 and $\bar{\mu}_o$ determined by this method for $10^{-3} \leq \gamma \leq 20$. The average cosine $\bar{\mu}_o$ has been shown in place of ξ_o to reduce the extent of the variation (these two parameters are related by Eq. (35)). The curves are parametric in the Knudsen number based upon tube diameter, which has been restricted to values ≤ 1.

The values of ξ_1 and ξ_o which simultaneously satisfy Eqs. (38) and (39) approach limits at large and small γ. The limit as $\gamma \to 0$

can be obtained analytically from the angular distribution function and the integral restrictions if it is noted that $K \to 4\gamma/3$ and if ξ_o is assumed to become proportional to γ. As $\gamma \to 0$, Eqs. (27), (38), and (39) yield:

$$\xi_o = \left[\frac{4/3}{1+\pi/2}\right]\gamma = 0.515\ \gamma$$

$$\xi_1 = \frac{\pi}{1+\pi/2} = 1.223 \tag{40}$$

These limits are different from the values of the analogous wall collision density parameters, ζ_o and ζ_1 given by Eq. (14). The reason for this difference is not due to the presence of intermolecular collisions in the former instance, since Eq. (40) is valid for any Knudsen number. The difference is due to the fact that ξ_o and ξ_1 represent number densities, so that Eq. (39) must be satisfied; ζ_o and ζ_1, on the other hand, represent parameters of a wall collision ~~density~~ rate profile, which is not subject to the constraint of Eq. (39). The value of ξ_1 greater than unity does not make sense physically. Either the number density cannot be approximated by the linear function of Eq. (25) over a sufficiently large fraction of the tube or the relation between $\nu(x)$ and $n(x)$ based upon equilibrium considerations (Eq. (24)) is not applicable. It is probable that neither of these assumptions are valid.

In the limit as $\gamma \to \infty$ (orifice), ξ_o, ξ_1, ζ_o, and ζ_1 all approach 1/2. Clausing (9) has discussed the reasons why ζ_o and ζ_1 behave so. That ξ_o and ξ_1 should also approach 1/2 can be seen by considering two chambers separated by a small orifice. If both vessels contain a gas at a density n_s, the density in the orifice is also n_s. If one

of the chambers is evacuated, those molecules which were in the orifice by virtue of effusion from the now-evacuated chamber disappear, but the contribution from the chamber still containing a density n_s remains unaltered. Consequently, the number density in an orifice separating a vessel containing a gas from a vacuum is one-half of the density in the reservoir.

Fig. 2 shows that for tubes of $\gamma<0.1$ the average cosine is very close to the long tube limit of 0.643 and only weakly dependent upon Knudsen number. Jones et al (5) determined the average cosine from all of their angular distribution data and found a constant value of 0.70 ± 0.02 under all conditions. Although the near-constancy of $\bar{\mu}_o$ predicted by the theory is verified, the magnitude is not. This may be due to the experimental difficulty of obtaining accurate measurements of the weak flux at large polar angles, which are weighted very heavily in the integrals of Eqs. (38) and (39). Zugenmaier's theory employs a $\bar{\mu}_o$ of one half for all γ although this value is characteristic of a cosine distribution and is attained only for very short tubes.

Comparison of Various Theories

The peaking factor (or centerline intensity) from the Giordmaine and Wang and Zugenmaier theories and the present analysis are plotted in Fig. 3. The results of the Ivanov and Troitskii theory are not shown, but fall close to Giordmaine and Wang and Zugenmaier curves. The peaking factors from the present theory are greater than those from the earlier works, especially at small Kundsen numbers. This effect can be seen more clearly by considering the limiting case of $Kn_D \approx$ order unity, $\gamma \to 0$, $Kn_L << 1$ (which represents

a long opaque tube). Since in the limit of long tubes, Eq. (40) shows that $\xi_o \sim \gamma$, the term δ of Eq. (30a) approaches zero but $\xi_1\delta/\xi_o$ becomes large. The centerline intensity of Eq. (31) reduces to:

$$j(0) = K\chi = \frac{\sqrt{\pi}}{2}\frac{\sqrt{\xi_1}}{\sqrt{1/2Kn_L}} \tag{41}$$

which is larger than the peaking factor predicted by the Giordmaine and Wang model by $\sqrt{\xi_1} = \sqrt{1.223}$. This constant factor of $\sim$ 11% is evident in the Kn_D=10 and 1 sets of curves at small γ in Fig. 3. Although this difference is of the same order as the accuracy of the experimental measurements, the results presented in the first paper of this series (1) generally showed higher peaking factors than the theoretical prediction of the Giordmaine and Wang theory. Moreover, the discrepancies were most evident at smaller Kn_D, which is consistent with the present theory (the $\sim$ 11% difference between Giordmaine and Wang theory and the present theory is attained only when Kn_L is small; as $Kn_L \to \infty$, the peaking factor approaches 1/K in all theories).

Fig. 4 compares the angular distribution from the four theories with data obtained by Giordmaine and Wang (2) for their source B, (γ=0.0152). At the 0.25 torr source pressure for which the data in Fig. 4 were reported to have been taken, Kn_D is calculated to be 2.84. The measured total flow rate under these conditions was reported as 1.78×10^{14} molecules/sec-channel, yet the flow rate computed from Eq. (1) is 2.66×10^{13} molecules/sec-channel. The peaking factor calculated from the centerline

intensity data reported by Giordmaine and Wang is 5.1, while that computed from Eq. (41) is 13.2 - 14.6, depending upon whether ξ_1 is taken as unity or 1.223. Since the measured flow rate is too large by a factor of ~ 6.7 and the centerline intensity is ~~high~~ low by a factor of ~ 2.6 (or $\sim \sqrt{6.7}$), it appears that the source pressure reported by Giordmaine and Wang for this experiment was too small by a factor of ~ 6.7. If the driving pressure were 1.7 torr instead of 0.25 torr, both the total flow rate and the centerline intensity measurement would have been in accord with theory. In computing the various theoretical curves in Fig. 4, therefore, the Knudsen number was taken to be 2.84/6.7 = 0.42.

It is impossible to distinguish between the various theories at small polar angles; the half widths at half maximum are within 1° of each other. At larger polar angles, the Giordmaine and Wang theory is substantially below the three theories which account for end density effects. The latter, however, all predict angular distributions which are within $\sim$ 10% of each other. The theory developed in the present work falls between those of Zugenmaier and Ivanov and Troitskii. The data are not sufficiently precise to permit a decision concerning the validity of anyone of the three theories to be made.

The same indistinguishability of the three theories appears at other values of the parameters γ and Kn_D. Ivanov and Troitskii have found agreement between their theory and the measurements of Naumov (~~15~~ 16) comparable to the accord shown in Fig. 4.

Conclusions

Previous theoretical studies of the angular distribution of a molecular beam from cylindrical tubes have been shown to differ only in the choice of the entrance and exit densities which fix the linear density profile upon which the calculation is based. The approximations used by Giordmaine and Wang (2), Zugenmaier (3), Ivanov and Troitskii (4) and Jones et al (5) are internally inconsistent, since the integral of the angular distribution does not equal the total flow rate and the average cosine of the distribution is not consistent with the exit density selected.

These two internal inconsistencies have been rectified by selecting ξ_o and ξ_1 to satisfy integral constraints on the angular distribution. In contrast to previous studies, the total flow rate is eliminated as a parameter of the angular distribution by utilizing the transmission probability of the tube. While this method restricts the calculation to driving pressures where the Knudsen number based upon tube diameter is of the order of unity or greater, the angular distribution and centerline intensity are functions only of the diameter-to-length ratio of the tube and the Knudsen number.

The parameters ξ_o and ξ_1 computed by the method of integral constraints agree with the analogous parameters derived by Clausing for collision-free flow only in the orifice limit.
The lack of agreement for long tubes is believed due to failure of the equilibrium relation $\nu=n\bar{v}/4$ used in the calculation or to the inapplicability of a linear number density profile, even in collision free-flow.

Despite substantial differences in the end conditions ξ_o and ξ_1 in the various theoretical models, the angular distribution from all theories are so similar that existing experimental data cannot demonstrate the superiority of any one. It appears that selection of $\xi_1 \approx 1$ and ξ_o anywhere between 1/3 and 1/2 of the transmission probability will generate an angular distribution adequate for most practical purposes. However, the angular distribution based upon the Giordmaine and Wang end conditions (which sets $\xi_o=0$) is distinctly different from the theories involving non-zero exit densities.

The peaking factors from the integral constraint method developed here, however, can be as much as 10% greater than predicted by the earlier methods at low Knudsen numbers. For practical purposes, the centerline beam intensity may be estimated by utilizing the simple Giordmaine and Wang theory and adding an additional 10% at Knudsen number based on diameter of less than ~ 10.

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Appendix

Relation Between the Exit Density and the Average Cosine of the Angular Distribution

Consider the point within the tube at downstream position x and radial position r. A direction vector $\vec{\Omega}$ from this point may be described by the polar angle $\theta = \cos^{-1}\mu$ with respect to a normal parallel to the tube axis and an azimuthal angle ϕ on the cross section at x. Let $n(r,x,\mu,\phi)d\Omega$ denote the number of molecules per unit volume at location r,x with velocities in the directions in $(\vec{\Omega},d\Omega)$. By symmetry, this distribution function does not depend upon the azimuthal location of the point on the cross section. Since the speed distribution within the tube is assumed Maxwellian, all molecules may be considered to be travelling with the mean speed $\bar{v}$.

The number of molecules with direction in $(\vec{\Omega},d\Omega)$ passing a unit area of the tube cross section at r,x per second is $\mu\bar{v}n(r,x,\mu,\phi)$. The net flow rate is obtained by integrating this expression over all directions and then integrating over the tube cross section:

$$\dot{N} = 2\pi\bar{v}\int_0^a rdr\int_{-1}^{1}\mu n(r,x,\mu)d\mu \qquad \text{(A-1)}$$

where $n(r,x,\mu)d\mu$ is the number of molecules per unit volume at r,x with direction in the cone angle $(\mu,d\mu)$:

$$n(r,x,\mu) = \int_0^{2\pi} n(r,x,\mu,\phi)d\phi \qquad \text{(A-2)}$$

The average cosine of the distribution at location r,x is defined by:

$$\bar{\mu}(r,x) = \frac{\int_{-1}^{1} \mu n(r,x,\mu)d\mu}{\int_{-1}^{1} n(r,x,\mu)d\mu} \tag{A-3}$$

The radially-averaged average cosine at downstream location x is defined by:

$$\bar{\bar{\mu}}(x) = \frac{\int_{0}^{a} \bar{\mu}(r,x)n(r,x)rdr}{\int_{0}^{a} n(r,x)rdr} \tag{A-4}$$

where n(r,x) is the total density at r,x

$$n(r,x) = \int_{-1}^{1} n(r,x,\mu)d\mu \tag{A-5}$$

The average density across the tube cross section at x is:

$$\bar{n}(x) = \frac{2}{a^2}\int_{0}^{a} n(r,x)rdr \tag{A-6}$$

Using Eqs. (A-2) - (A-6) in Eq. (A-1) yields:

$$\dot{N} = (\pi a^2)\bar{v}\ \bar{n}(x)\bar{\bar{\mu}}(x) \tag{A-7}$$

Equating this expression to Eq. (1) of the text yields:

$$\bar{\bar{\mu}}(x) = \frac{K}{4[\bar{n}(x)/n_s)]} \tag{A-8}$$

Eq. (A-8) applies to all downstream locations in the tube. If at the tube exit, $\bar{\bar{\mu}}(L)$ is denoted by $\bar{\mu}_o$ and $\bar{n}(L)/n_s$ by ξ_o, Eq. (A-8) reduces to Eq. (35) of the text.

The angular distribution of the molecular beam is

$$J(\mu) = \mu\bar{v}\int_0^a n(r,L,\mu)\,r\,dr \tag{A-9}$$

The integral of Eq. (A-9) over all polar angles is:

$$\int_{-1}^{1} J(\mu)\,d\mu = \bar{v}\,\bar{\bar{\mu}}(L)\int_0^a n(r,L)\,r\,dr \tag{A-10}$$

where Eqs. (A-3) and (A-4) have been used.

Division of Eq. (A-9) by μ and integration over all polar angles yields:

$$\int_{-1}^{1} \frac{J(\mu)}{\mu}\,d\mu = \bar{v}\int_0^a n(r,L)\,r\,dr \tag{A-11}$$

Division of Eq. (A-10) by Eq. (A-11) gives:

$$\bar{\bar{\mu}}(L) = \int_{-1}^{1} J(\mu)\,d\mu \Big/ \int_{-1}^{1} \frac{J(\mu)}{\mu}\,d\mu \tag{A-12}$$

Since $J(\mu)$ is zero for $\mu<0$, Eq. (A-12) is equivalent to Eq. (36) of the text.

For an ideal orifice, $n(r,L,\mu,\phi) = n_s/4\pi$ for $\mu>0$, and zero otherwise. Eq. (A-9) reduces to the cosine effusion law, Eq. (A-12) gives $\bar{\bar{\mu}} = 1/2$, and Eq. (A-8) (with K=1) shows that the density in the orifice is $n_s/2$.

In collision-free flow, the wall collision rate, $\nu(x)$ is

given by a solution to Eq. (6) of the text. The rate at which molecules enter each unit area of the tube cross section at $x=0$ is ν_s. Knowledge of these supply rates and assumption of a cosine distribution of $\nu(x)$ permits the number density distribution function $n(r,x,\mu,\phi)$ to be computed by the same purely geometrical considerations from which Eq. (6) was derived. To our knowledge, this computation has not been performed. Determination of $n(r,x,\mu,\phi)$ and hence $\bar{n}(x)$ by this exact method would demonstrate whether Eqs. (24) and (25) are valid approximations in the absence of intermolecular collisions.

FOOTNOTES

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15. Becker's centerline intensity was derived from a cruder model than that upon which the other four are based. His peaking factor (for a long cylindrical tube) differs from Giordmaine and Wang's (2) by a factor of $2^{3/4}/\sqrt{\pi} \equiv 0.95$. Ivanov and Troitskii's angular distribution formula did not have the term which contains $(1/\xi_1-1)$ in Eq. (26), even though ξ_1 was not unity. This was probably the result of using $\xi_1 n_s$ instead of n_s in computing the contribution from the reservoir, which is incorrect.

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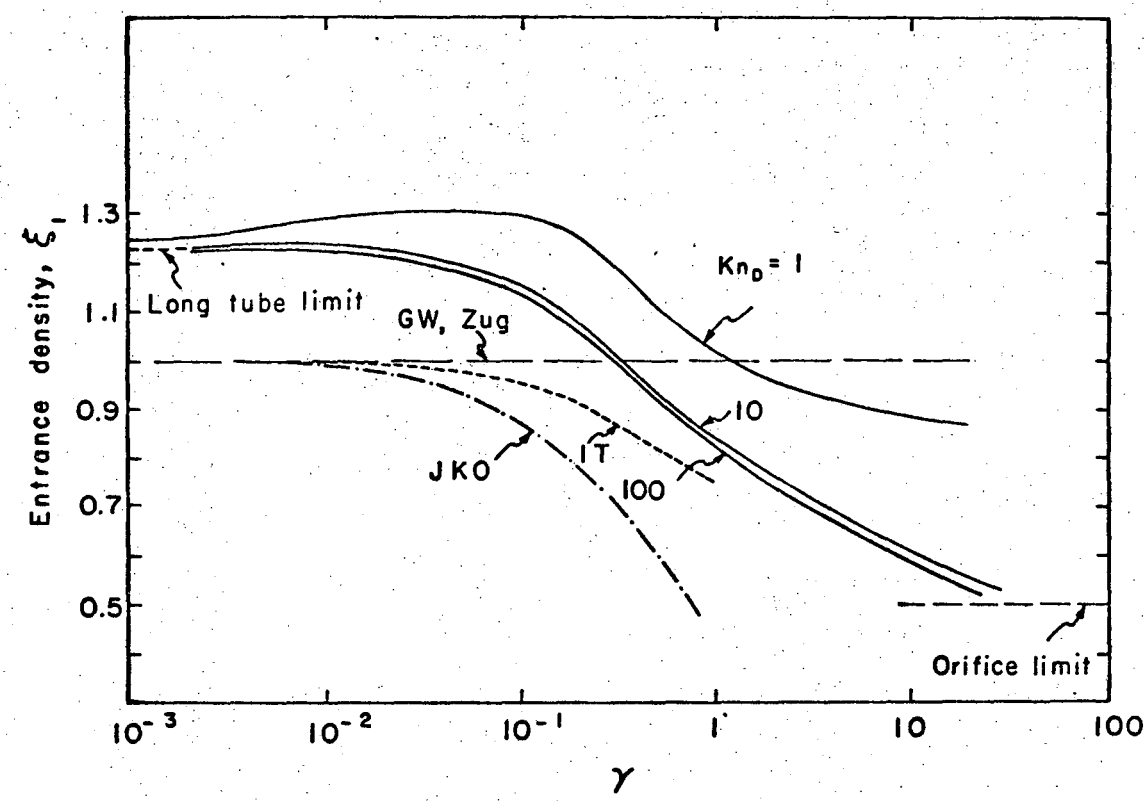
FIGURE CAPTIONS

1. Normalized entrance density as a function of tube diameter-to-length ratio. Curves representing previous studies are denoted by: "Zug", ref. 3; "IT", ref. 4; "JKO", ref. 5; and "GW", ref. 2.

2. Average cosine at the tube exit (related to the normalized exit density by Eq. (35)). Curves representing previous studies are denoted by: "Zug", ref. 3; "IT", ref. 4; "JKO", ref. 5.

3. Peaking Factor as a function of the tube diameter-to-length ratio for various Knudsen numbers.

4. Angular distributions for $\gamma + 0.0152$, $Kn_D = 0.42$. Data from Giordmaine and Wang (2), source B at a pressure of 0.25 torr.



XBL6912-6357

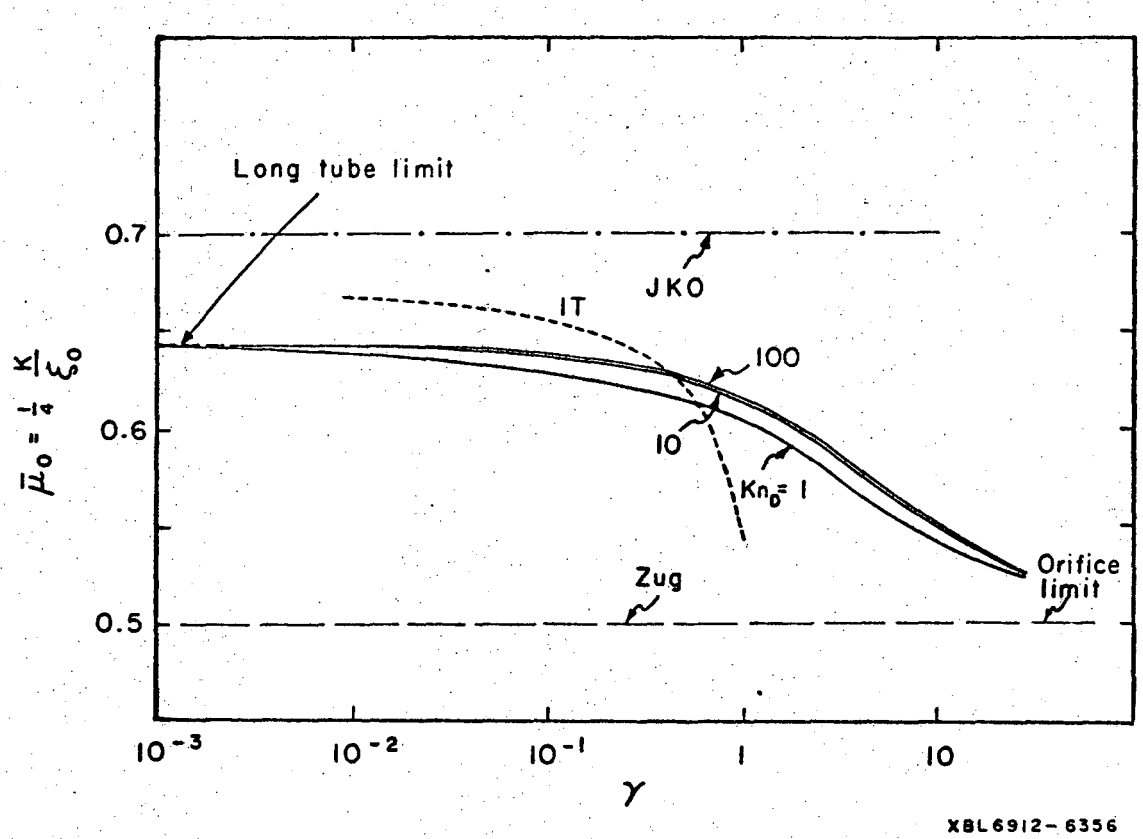


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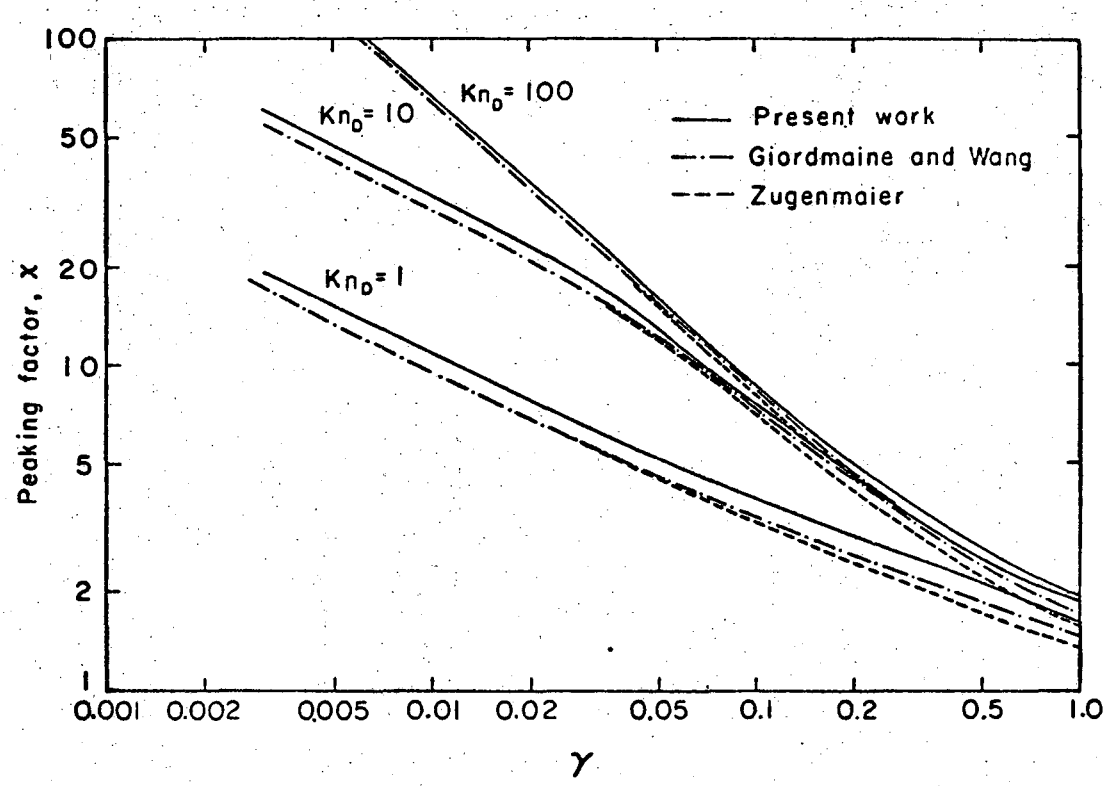
Fig 2



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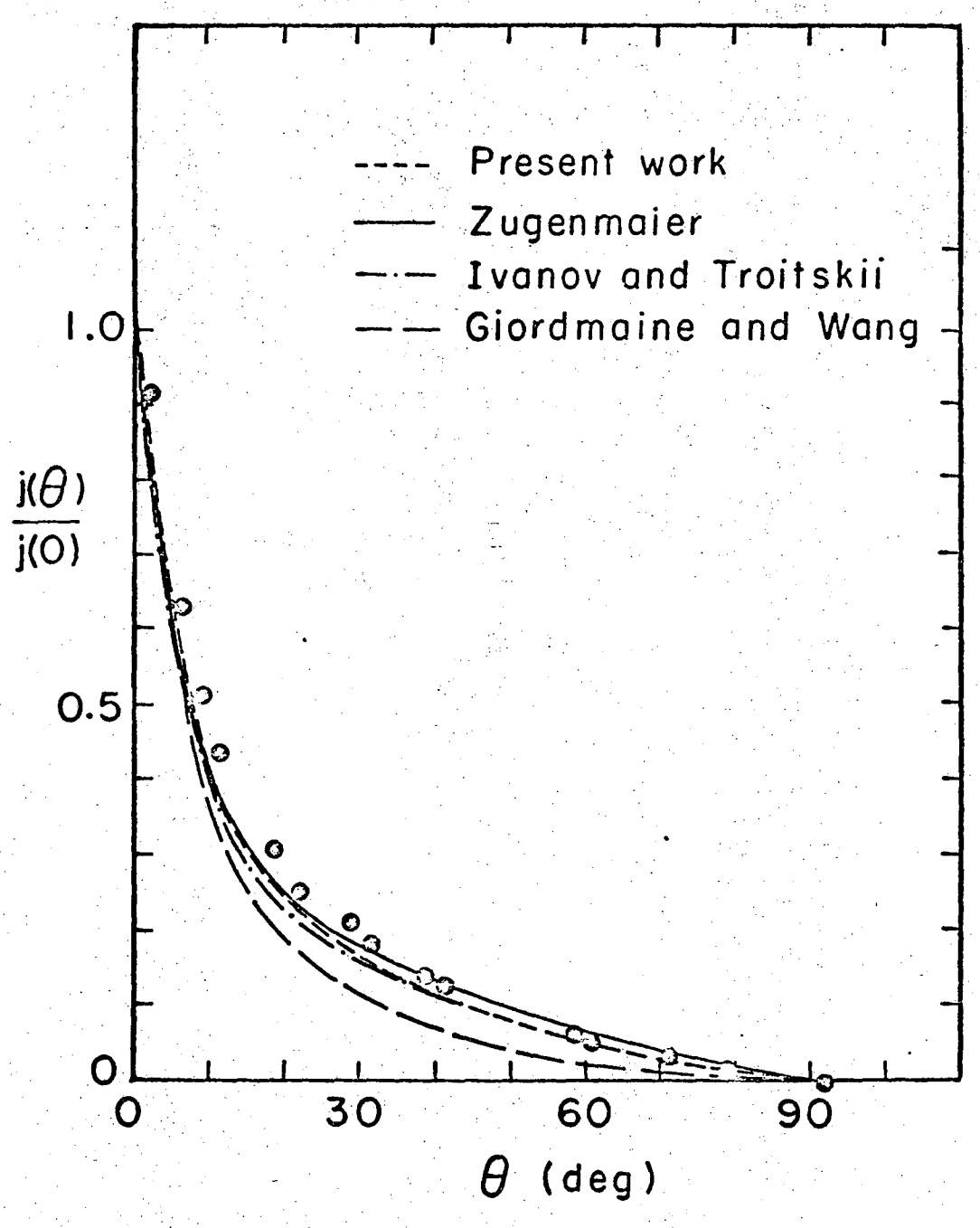

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Fig 4

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