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Continuum-Source Molecular Beams

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Introduction

NEARLY fourteen years have passed since Kantrowitz and Grey[1] first suggested that the intensity of classical molecular beams could be substantially exceeded by substituting a continuum nozzle expansion for the conventional free-molecular effusive source. They emphasized that the key problem was the extraction of a working beam from the core of the resulting supersonic stream by a skimmer without degradation of the ordered motion achieved in the expansion. In the companion experimental paper,[2] successful beam formation was not achieved primarily because of skimmer effects. Since that time, however, the work of several groups repeatedly referenced throughout this paper has established not only that continuum-source beams are feasible, but also that they possess advantages beyond those first envisaged. In particular, the fact that they are capable of producing much higher beam velocities really means that they constitute a new member in the family of aerophysical research tools. Their potential remains to be exploited.

In Fig. 1 the approximate domains of simulation open to various types of facilities have been sketched in on top of the familiar flight corridor. The central advantage of the continuum-source beams lies in their ability to produce the energies of interaction corresponding to orbital and even super-orbital flight (approximately 8–15 ev for nitrogen molecules, for example), together with intensities that make measurements feasible. Until this type of facility became available, there existed no practical experimental means of investigating the energy "gap" between the thermal energies available with classical beams and about 20 ev, the lowest energy for charge exchanged ion beams with usable intensity.

Energy simulation is imperative, as was stressed in earlier surveys in this series by Cheng[3] and Wachman.[4] In gas-surface interaction studies, in which the laws governing the reflection of molecules provide the boundary conditions for satellite aerodynamics, the reflection process is known to be energy-dependent. The reaction energies of many chemical processes, of dissociation, excitation and ionization, and of chemisorption on surfaces all lie in this 1–20 ev range. The possibility of studying these processes under closely controllable conditions for the first time has provoked interest in continuum-source beams in many laboratories.

The purpose of this paper is to review the literature that bears directly on the beam-generation problem, with particular emphasis on the recent developments in understanding of the free-jet behavior and the skimming process.

Basic Principles and Problems

The elements of a basic continuum-source beam are shown in Fig. 2. The source is run at relatively high pressures, from hundreds of mm Hg pressure to several atmospheres, and may be heated in a variety of ways. The flow expands through a sonic orifice to form (usually) a hypersonic free jet in the expansion chamber which is separately pumped to pressures in the range 10^{-2} to 10^{-4} mm. The function of the conical skimmer is to allow the highly ordered motion in the core of the free jet to pass on unperturbed into the collimating chamber, where the higher vacuums, typically 10^{-6} to 10^{-7} mm, eliminate background scattering. Expansion may be performed in more than one stage. A final collimator is used to define the working beam, which passes on into the test chamber. The collimation and test chambers may be served by a common pump, or if ultrahigh vacuums are required in the test chamber, as in the case of surface interaction studies, a separate chamber and pumping system may be used.

The over-all performance of the system was idealized by Kantrowitz and Grey under the following assumptions: 1) the expansion is isentropic up to the skimmer, 2) the flow passes through the skimmer without disturbance, and 3)

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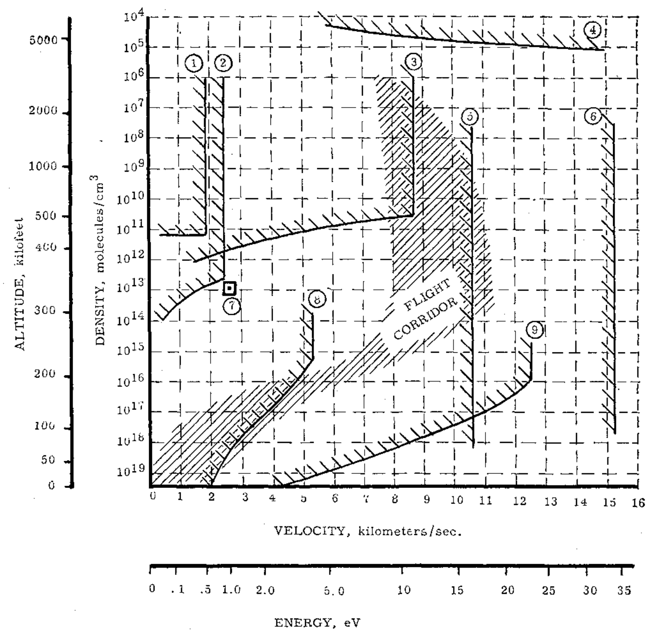

Fig. 1 Simulation capabilities of various facilities: 1) classical oven-source molecular beam, 2) continuum-source beam, pure nitrogen, 3) continuum-source seeded beam, 1% N_2-99% H_2, 4) molecular nitrogen beam from charge-exchanged ion beam, 5) light gas gun, 6) proposed Arnold Engineering Development Center (AEDC) counterflow facility, 7) shock-tube driven monopulse molecular beam,[41] 8) Cornell Aeronautical Laboratory (CAL) shock tunnel, 9) 10,000 psi, combustion-driven shock tube.

downstream of the skimmer no further molecular collisions occur. For Mach numbers greater than three, these yield the following basic beam-performance equations.[5]

Flow through the skimmer N_s (molecules/sec):

$$N_s = n_s a_0 \tfrac{1}{4}\pi d_s^2 M_s\{1 + [(\gamma - 1)/2]M_s^2\}^{-1/2} \qquad (1)$$

where a_0 is the stagnation speed of sound, d_s is the skimmer diameter, and M_s and n_s are the Mach number and number density at the skimmer entry, related isentropically to source conditions. How the Mach number may be obtained for a given sonic orifice-to-skimmer separation will be discussed in the section on free jets.

Flow through collimator N_c:

$$N_c = \frac{1}{2} n_s a_0 \left(\frac{\pi}{4} d_s^2\right)\left(\frac{\pi}{4} d_c^2\right)\frac{M_s(3 + \gamma M_s^2)}{\pi L^2\{1 + [(\gamma - 1)/2]M_s^2\}} \qquad (2)$$

where d_c is the collimator diameter and L the skimmer-to-collimator distance. The gain in intensity possible compared to a classical beam may be seen by noting that, for the latter, the flux reaching the collimator from a source at the skimmer position is

$$N_c = \tfrac{1}{2} n_s a_0[(\pi/4)d_s^2][(\pi/4)d_c^2](\pi L^2)^{-1}(2/\pi\gamma)^{-1/2} \qquad (3)$$

If n_s is chosen to be such that the local mean free path is equal to the skimmer diameter d_s in both cases (which is the normal operating condition for a classical beam and is also a reasonable operating condition for the continuum-source beam), then the intensification gain factor is

$$(\pi\gamma/2)^{1/2}M_s(3 + \gamma M_s^2)\{1 + [(\gamma - 1)/2]M_s^2\}^{-1/2} \qquad (4)$$

which is over 200 at $M = 7$, for example.

The energy gain possible depends on the system used. If a continuum-source beam and a classical beam are compared using the identical gases from sources at identical temperatures, the gains are not spectacular. The classical beam has an rms velocity $\langle c^2\rangle^{1/2}$, which is related to the most probable molecular velocity in the source c_{m_0}

$$\langle c^2\rangle^{1/2} = 2^{1/2}c_{m_0} = (4kT_0/m)^{1/2} \qquad (5)$$

where m is the molecular mass. If we select nitrogen molecules at 2500°K as a basis for comparison, this corresponds to a mean energy per beam molecule of 0.431 ev (1752 m/sec). In the continuum-source beam, it has been shown experimentally that molecular velocities approaching the terminal velocity (i.e., complete conversion to directed motion) can be approached within a few percent. Here,

$$u = \{2[\gamma/(\gamma - 1)]kT_0/m\}^{1/2} \qquad (6)$$

so that, for nitrogen molecules from a 2500°K source, u = 2300 m/sec or 0.755 ev, not a great increase over the classical beam. Of course, as Kantrowitz and Grey stressed, the energy spread will have been greatly narrowed.

However, the continuum-source beams possess two further advantages not open to classical beams. First, the "material limitation" on the source temperature can be overcome by using systems such as shock tubes or arc jets to provide much higher source gas temperatures. Notice that to obtain a beam of 10-ev nitrogen molecules requires, ideally, a source temperature of 33,000°K, so that real gas effects such as dissociation, electronic excitation, and ionization are inescapably involved in the source and expansion, a situation that may have advantages or disadvantages.

A second possibility for energy augmentation is the seeding technique, first proposed by Fenn and Deckers.[6] A low molecular-weight carrier gas such as hydrogen or helium forms the bulk of the expansion, to which the desired seed gas is added in small amounts. Ideally, the terminal velocity will be increased by the root of the ratio of the mean molecular weight of the mixture to that of the seed gas alone. If the seed molecules achieve this velocity (in experiments they come within 10% of it), then their energy is increased directly as the molecular-weight ratio. Thus, if a mixture of 1% nitrogen and 99% hydrogen is used in a 2500°K source, the final nitrogen molecular velocity is 8160 m/sec, corresponding to 9.44 ev. Extreme source temperatures and attendant real gas effects have been avoided at the expense of intensity loss and beam dilution with the carrier gas. However, in one of the rare situations in which nature is benign, there are mechanisms, not yet completely understood, which can be used to good advantage to eliminate almost completely the carrier gas from the beam, once it has done its job of acceleration of the heavy molecules in the expansion.

Intensive study over the past few years of all the phenomena occurring in the formation of beams from continuum sources has led to performances that are approaching the ideal behavior outlined previously. The problems involved in attaining such performance may be divided conveniently into two groups. First, there are those problems that are intrinsic in the expansion process itself, the theoretical description of ideal behavior, departures from ideal behavior because

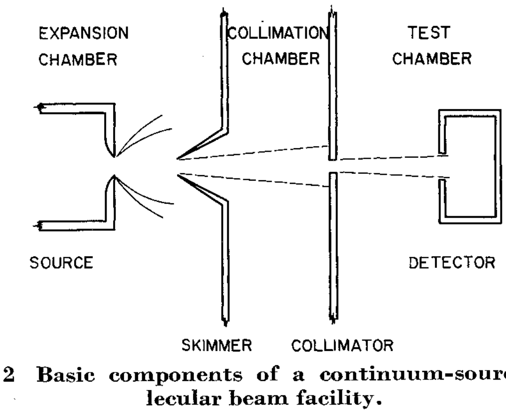

Fig. 2 Basic components of a continuum-source molecular beam facility.

of rapid expansion and freezing within the jet, diffusion from the boundaries, gas-separation phenomena in jets composed of mixed gases, and other effects such as condensation. Second, there are those problems that also involve the action of the skimmer, the basic modes of operation possible, effects of skimmer geometry, and separation effects due to the skimmer. These effects are discussed in turn in the next sections.

Free Jet

Except for early work, attention has been concentrated on the free jet as the basic expansion mechanism in the generation of aerodynamic molecular beams, for three reasons. First, since only the centerline flow is used, there is no need to generate the uniformity of flow field which is desirable in wind-tunnel applications. Second, the overexpansion produced in front of the first Mach disk permits much higher Mach numbers and better beam collimation than could otherwise be obtained for a given pumping capability. Third, the theoretical treatments of free jets now available permit convenient relation of the position along the centerline with the flow properties, within the limitation imposed by the usual isentropic assumption.

Ideal Free-Jet Behavior

The first to consider the free jet theoretically were Owen and Thornhill.[7] They obtained a method-of-characteristics solution for a $\gamma = \frac{7}{5}$ gas expanding into a perfect vacuum. Using a starting condition of Mach 1.0038 flow expanding radially from a 15° conical nozzle, they obtained the downstream axial Mach number variation indicated in Fig. 3. If the jet expands into a region of finite pressure rather than into a vacuum, then the typical free-jet structure shown in the insert in Fig. 3 will develop.[8, 9] The Owen and Thornhill solution will still be valid up to the position of the first Mach disk, since flow within the shock barrel is in a "zone of silence," which is uninfluenced by pressure changes along the jet boundary. In 1963 Wolff obtained similar method-of-characteristics solutions, published in Ref. 10, for the different specific-heat ratios shown in Fig. 3 and extended these to much higher pressure ratios. The small discrepancy with the Owen and Thornhill solution at low Mach numbers is because Wolff's characteristic net started from a uniform Mach 1.10 flow.

Earlier, Love and his associates at NACA had also studied the free jet theoretically and experimentally, although through smaller pressure ratios.[11, 12] Their method-of-characteristics solution described the whole flow field, since they incorporated a matching procedure between the jet boundary and the surrounding gas. They calculated the position of the jet boundary, barrel shock, and Mach disk for a variety of conditions, but somewhat curiously refrained from presenting data concerning flow parameters within the shock barrel, although the program could have provided this.

Adamson and Nicholls[8] also developed an approximate analysis giving the position of the first Mach disk but valid only for stagnation-to-ambient pressure ratios less than 140.

In general, the experimental results of several groups confirm the axial Mach number distribution and Mach disk position predicted by these theories up to pressure ratios approaching 1000. Comparison of the experimental data on Mach disk position obtained by Adamson and Nicholls, Love et al., and Bier and Schmidt[13] is summarized in the paper by Bier and Schmidt. Axial Mach number variation has been

Table 1 Free-jet parameters

γ	A	x_0/D	x_0'/D	B	C
$\frac{5}{3}$	3.26	0.075	0.04	0.643	1.365
$\frac{7}{5}$	3.65	0.40	0.13	0.357	1.662
$\frac{9}{7}$	3.96	0.85	...	0.246	1.888

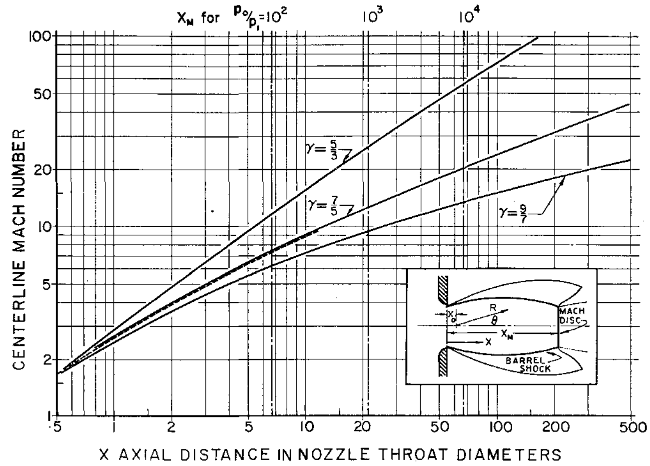

Fig. 3 Free-jet parameters: dashed curve, Owen and Thornhill[7]; solid curves, Wolff.[10]

measured by Reis and Fenn[14] to be closely in agreement with theory up to Mach 10 for a stagnation pressure as low as 200 mm.

Two recent papers on free jets by Sherman[10] and Ashkenas and Sherman[15] serve admirably to summarize the present picture of free-jet behavior. In these papers, Sherman studied the available method-of-characteristics solutions, and noticed that, at large enough pressure ratios, a region of "inertia-dominated flow" existed within the barrel shock and Mach disk. This is flow in which the conversion from thermal to directed motion is so nearly complete that the internal energy and static pressure are negligible in comparison to the kinetic energy of directed motion and the corresponding dynamic pressure, respectively. In the ideal expansion, the flow velocity in this region will be constant at nearly the terminal velocity. In this region the method-of-characteristics solutions showed that, in the words of Ashkenas and Sherman, "the flows exhibit a relatively simple and self-similar development. The streamlines appear to radiate from a 'source' at a distance x_0 (measured in orifice radii) downstream of the orifice, the flow speed has very nearly attained its adiabatic flow limit, and density decreases along each streamline in proportion to the inverse square of distance from this source. The variation of density from streamline to streamline (i.e., with polar angle θ, at constant distance R from the "source") is approximately independent of R." These observations suggested the following fitting formulas. For the centerline Mach number at a distance x downstream of an orifice of diameter D,

$$M = A\left(\frac{x - x_0}{D}\right)^{\gamma-1} - \frac{1}{2}\left(\frac{\gamma+1}{\gamma-1}\right)\left[A\left(\frac{x - x_0}{D}\right)^{\gamma-1}\right]^{-1} \tag{7}$$

where the constants A and x_0 are given in Table 1. Impact pressure is given by

$$\frac{P_i}{P_0} = \left(\frac{\gamma+1}{\gamma-1}\right)^{\gamma/\gamma-1}\left(\frac{\gamma+1}{2\gamma}\right)^{1/\gamma-1} A^{-2/\gamma-1}\left(\frac{x - x_0'}{D}\right)^{-2} \tag{8}$$

where P_0 is the stagnation pressure, and x_0' is a slightly adjusted apparent-source distance parameter given in Table 1. The density field is described by

$$\rho/\rho_0 = B\cos^2(\pi\phi/2C)R^{-2} \tag{9}$$

where ρ_0 is the stagnation density, ϕ is the polar angle connecting the point of interest to the apparent source a distance R apart (in units of nozzle-exit radius), and B and C are given in Table 1. The formulas for Mach number and impact pres-

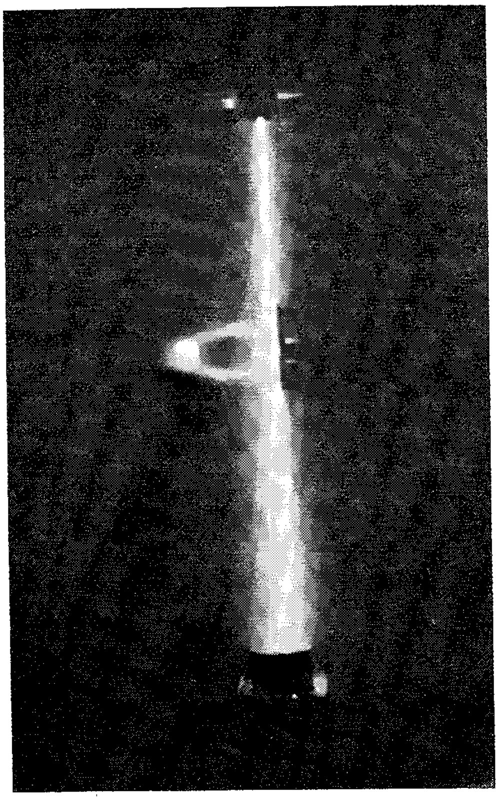

Fig. 4a Free-jet visualization by afterglow, using electron beam excitation at the orifice exit. Conditions: unheated argon, P_0 = 1038 mm, P_1 = 50μ, and D = 0.013 in. The electron-beam entry, jet orifice plate, and electron-beam Faraday cup are visible.

Fig. 4b Free-jet density field obtained by traversing electron beam down the jet axis. Conditions: unheated argon, P_0 = 1200 mm, and P_1 = 10 μ.

sure fit the method-of-characteristics data to within 1%, and the density formula is good to within 3 to 4%. Finally, the position of the Mach disk x_M is given to very good accuracy, independent of the value of γ, by

$$x_M/D = 0.67(P_0/P_1)^{1/2} \tag{10}$$

where P_1 is the background pressure. This formula has been checked experimentally by Ashkenas and Sherman for pressure ratios up to 17,000.

Departures from Ideal Behavior

The previous formulas are very useful in the beam-generation problem because, in principle, they provide the information necessary to characterize the final beam produced after the skimming process and collimation: the n_s and M_s of Eq. (2). That is, the properties of a continuum-source beam could be calculated in as straightforward a manner as those of a classical beam, provided that the skimmer action produced a collisionless beam without disturbance. However, the free jet in itself may depart from ideal behavior in several ways, and the various possibilities for nonideal behavior are now examined, in turn, to see how these may affect the generated beam.

Viscous effects ahead of the orifice have small but measurable effects on the discharge coefficient. For example, Anderson et al.[16] quote an effective flow diameter of 0.94 times the actual throat diameter for argon expanding through a 1.71-mm orifice from 50 torr and room temperature. Ashkenas and Sherman[15] have investigated the variation of effective orifice diameter D^* with Reynolds number over the range $13 < Re < 2500$, given in their Fig. 8, and have shown a simple D^*/D dependence on Re. The effects were generally very small for throat Reynolds numbers greater than a few hundred. Beam formation is affected only in that the scale of the free jet is based on the effective throat diameter.

Sherman and Ashkenas also considered the possibility of viscous effects in the core of the free jet due to the intrinsic velocity and temperature gradients. Using the expansion

$$M = M_0[1 + m_1/Re + \ldots] \tag{11}$$

where M_0 refers to the inviscid approximation, and m_1 is the first viscous perturbation, they found that, at large distances from the throat, the perturbation became

$$m_1 = \frac{-2\gamma(\gamma-1)(\lambda/\mu+1)A^{[2\gamma/(\gamma-1)]-2\omega}}{1+2(\gamma-1)(1-\omega)} \times \left(\frac{\gamma+1}{\gamma-1}\right)^{\omega-[(\gamma+1)/2(\gamma-1)]}\left(\frac{x-x_0}{D}\right)^{1+2(\gamma-1)(1-\omega)} \tag{12}$$

in which μ and λ are the normal and bulk viscosity coefficients, ω is the index (assumed constant) in the viscosity-temperature relationship $\mu \propto T^\omega$, and A is given by $[(\gamma+1)/(\gamma-1)]^{(\gamma+1)/4}$ in the simple source flow approximation. Near the orifice the main contribution to the perturbation comes from heat conduction, but at larger distances the dominant mechanism is viscous dissipation due to lateral stretching of the fluid elements.

The net result is a rise in internal energy which is significant compared to the small amounts of internal energy present in the highly expanded gas in the inviscid case, but the concomitant decrease in the bulk-stream kinetic energy is relatively negligible. The implication of this for beam formation is that, whereas the collimation of the final beam will be affected as a result of the increased lateral spreading of the beam from the random velocity components, the other factors that affect beam formation, i.e., the bulk velocity and the mass flux bombarding the front of the skimmer, will be almost unaffected.

This last remark also applies to the phenomenon of kinetic freezing, which occurs when the collisional frequency drops so low that further conversion of energy in the translatory degrees of freedom to directed motion is negligible. From this point on, the gas continues to expand with density decreasing as the inverse radius squared but at constant speed ratio and velocity. This process was considered theoretically by Brook and Oman,[17] who used a kinetic model of the Boltzmann equation in studying spherical and cylindrical expansions. For a spherical expansion of argon, they found that the temperature T_F at which the expansion freezes, expressed in terms of the temperature at the sonic line T_*, is primarily a function of the sonic Reynolds number Re_* and to a lesser extent is a function of the stagnation temperature. The results of their program may be expressed in the very approximate form

$$T_F/T_* = 7.36(\mathrm{Re}_*)^{-0.505} \tag{13}$$

for a stagnation temperature of 3000°K, and

$$T_F/T_* = 33.7(\mathrm{Re}_*)^{-0.732} \tag{14}$$

for the 1000°K case. The transition from equilibrium flow to fully frozen flow was very abrupt, suggesting that the region in a real expansion over which the flow changes from being describable in continuum terms to a completely collisionless radially expanding flow is very narrow. This lends support to the notion of a surface of last collisions in the expansion suggested by Anderson et al.[16] This surface acts as a distribution of highly nonisotropic sources of molecules for a skimmer that may be positioned downstream. Another factor suggested by the Brook and Oman results stems from the apparent sensitivity of the freezing point in the expansion to the initial gradients chosen in the computer program. This implies that, if it is desired to squeeze the maximum possible conversion of thermal to directed motion out of an expansion, it may be advantageous to program the initial stages of the expansion by using a short divergent section instead of the completely free jet so popular at present.*

Anderson et al.[16] have studied the kinetic freezing problem experimentally, and, by measuring the velocity distribution of the molecules emanating from the plane of last collisions in the jet, have determined the functional dependence of the limiting Mach number on a characteristic Knudsen number Kn_0 for the expansion based on the stagnation mean free path and the throat diameter. They found that for an argon free jet expanding from room temperature the Mach number at which the expansion froze was approximately

$$M = 1.18(Kn_0)^{-[(\gamma-1)/\gamma]} \tag{15}$$

Another factor that can hinder the utilization of these highly directed streams arises from the rapid growth of the mixing layer at the free-jet boundary. For high jet-pressure ratios and low ambient pressures, the shock waves forming the sides of the cell thicken, merge with the mixing region, and may, under extreme conditions, converge on the centerline, eliminating traces of the first Mach disk. There are no theoretical assessments of this process, but experimental studies have been reported by Bier and Hagena,[18] Ashkenas and Sherman,[15] and Fenn and Anderson.[19] Bier and Hagena suggest that the Mach disk will disappear when the mean free path behind it (which is close to the mean free path in the surrounding chamber) approximately equals the Mach disk diameter. Ashkenas and Sherman cite impact probe data and flow visualization studies, which support this conclusion. Figures 4a and 4b taken at the University of Toronto Institute for Aerospace Studies (UTIAS) also suggest agreement, and we thank D. E. Rothe and G. R. McMichael for permission to reproduce them. Figure 4a shows typical free-jet structure made visible by crossing the flow just downstream of the throat with an electron beam, which is badly overexposed. At a background pressure of 50 μ theMach disk is clearly discernible and is positioned at 96 throat diameters downstream, in almost exact agreement with the prediction of Eq. (10) for the experimental pressure ratio of 20,700.

In contrast, Fig. 4b shows the density field of a free jet expanding into a chamber at 10 μ. The local density was mapped by moving the electron beam downstream to "paint" a diametral cut along the axis of the jet. Taking the distance along the axis, at which the density returns to that of the background, as the shock position (240 throat diameters) gives very good agreement with the prediction of Eq. (10) (232 throat diameters), even for the extreme pressure ratio of 120,000 used here. The dark central region of overexpansion is clearly evident, but the Mach disk is diffuse to the point of disappearance. This agrees with Bier and Hagena's criterion, since a rough estimate of the ratio of chamber mean free path to Mach disk diameter is of order unity. The dark central region of the Mach cell in Fig. 4b suggests that a beam could be extracted, under these conditions, without deleterious effect from background gas, but Fenn and Anderson[19] present convincing evidence that, under lower jet stagnation pressures, the background gas can invade the Mach cell far ahead of the predicted position of the Mach disk. Their data, taken over a wide range of conditions, suggest that the distance L/D in orifice diameters at which the background gas has achieved ambient density on the jet centerline is given by the empirical correlation

$$\log_{10}\left(\frac{L}{D} + \frac{33}{Kn_0\, P_0/P_1}\right) = -1.07 \log_{10}\left[\frac{(10^3\, Kn_0)^{2(\gamma-1)/\gamma}}{\gamma}\right] + 1.26 \tag{16}$$

where Kn_0 is the source Knudsen number given by the source mean free path divided by the orifice diameter.

Mention should be made of condensation, another possible phenomenon, which can occur during expansion. Extensive experiments by Becker and his associates have shown that it is possible to generate extremely dense slow beams by making use of cold high-pressure source conditions so that a condensed phase appears during expansion and passes through the skimmer.[20-23] Such beams have applications in several physics and plasma-physics experiments. Onset of condensation has also been studied recently by Greene, Brewer, and Milne,[24, 25] and Anderson et al.[16] have discussed a theoretical model for dimer formation in the expansion. It would appear that, in general, condensation is not a serious problem in typical beam-generation situations.

Skimming Process and Beam Formation

The original Kantrowitz-Grey concept of the skimmer action contains a contradiction in requiring a continuum-like behavior with an attached shock external to the skimmer and at the same time a collisionless beam from the skimmer entry plane onwards. Both Troitskii[26] and Valleau and Deckers[27] have implied that the contradiction is not as stark as it seems, because the act of stripping away the surrounding gas from the beam greatly reduces the frequency of collisions within a volume element in the beam, so long as the mean free path at the skimmer entrance is of the same order as the beam diameter.

However, this concept is not adequate in general because the skimmer can operate under a variety of conditions as its axial position in the free jet is varied. Many groups have measured experimentally the beam flux arriving at a detector as the orifice-to-skimmer distance is varied.[6, 18, 28-30] The theoretical flux is the monotonically decreasing value given by Eq. (2). Experimentally, the flux (as the orifice-skimmer separation is increased) falls rapidly to a minimum, which is perhaps 1% of the theoretical value, rises to a maximum somewhat less than the theoretical value, and then decreases with further increase in orifice-skimmer separation, almost always remaining lower than the theoretical value. The commonly accepted explanation of this behavior is as follows.

* A publication by Knuth,[53] received after this manuscript was completed, deals with the freezing problem in more generality, including treatment of rotational freezing for diatomic gases. This material is also covered somewhat more briefly in a recent review by the same author.[54] The latter paper also contains a useful summary on molecular-beam speed determination and detection, topics not covered in the present review.

Even when the skimmer is only a few orifice diameters downstream, the Mach number is large enough so that the skimmer will operate with an attached external shock, and the internal flow in the skimmer will undergo a second expansion with internal shock waves. This condition has been extensively investigated by Becker and his associates at Karlsruhe.[13, 18, 31] The observed minimum at increased orifice-skimmer separation is ascribed to the formation of a detached shock wave in front of the skimmer, caused by viscous effects, which becomes increasingly important at the lower density. At greater separations the skimmer might be said to be operating in a transition regime in which a cloud of reflected molecules exists in front of, and perhaps just inside of, the skimmer, partially scattering the beam. This separation is the smallest that is of real interest in beam formation. An analytical model of this region has been proposed by Oman.[32]

At still larger separations the importance of molecules reflected externally from the skimmer is reduced rapidly, producing the increase in the experimental curve of beam intensity. This change in importance of external skimmer effects has been illustrated experimentally by Bier and Hagena[33] using a pulsed-beam technique and by Anderson et al.[16] and McGinn[34] using cryogenically cooled skimmers to eliminate reflected molecules. Finally, the reduction in beam intensity below the maximum at still larger separations has been demonstrated convincingly by Fenn and Anderson[19] to be due to the penetration of the jet by background gas, as was mentioned previously.

This last situation will only develop when over-all pressure ratios are large enough so that the position of the Mach disk given by Eq. (10) remains well downstream of the tip of the skimmer. Bier and Hagena[18] have shown, under conditions of lower over-all pressure ratios (100–2000) and higher background pressures (0.1–2 torr), that, when the conical skimmer is retracted beyond the position where its tip lies in the plane of the largest diameter of the shock barrel, the Mach disk-skimmer interaction is such that the shock moves to a detached position in front of the skimmer, drastically lowering beam intensity. Thus the Mach numbers attained in the free jet beyond the position of the maximum shock barrel diameter are not available for beam extraction.

An alternate explanation of the rise of beam intensity to a maximum value with increasing orifice-skimmer separation has been proposed recently by Valleau and Deckers[27, 35] who have stressed the importance of collisions among the beam molecules after skimming. Naming an effective molecular diameter σ for collisions that deflect molecules enough to miss the collimator, and assuming that the Mach number and density of molecules coming through the skimmer are known and related isentropically to conditions in the source, Valleu and Deckers show that, under approximations that are reasonable in typical beam-generation situations, the number density of molecules arriving at the collimator n_c is given by

ming can be important. Perhaps the only safe conclusions are that there is nothing mutually exclusive about the two mechanisms, and that more sophisticated experiments using, for example, electron-beam probing external to the skimmer may be of assistance in assessing their relative importance under various conditions.

Effect of Skimmer Geometry

Kantrowitz and Grey suggested that the central problem was the skimmer design, and the first study of the effects of skimmer geometry by Zapata, Parker, and Bodine[36] showed that for the supersonic flows then attainable it was necessary to use a skimmer opening relatively large compared to the mean free path and to provide a sharp skimmer edge, in order to allow any supersonic flow at all through the skimmer. Since this forces the skimmer into the role of a second-stage expansion nozzle, the problem still remains of extracting from this continuum expansion a working beam.

Using a two-stage expansion and higher pressure ratios, Becker and his associates[37–39] were able to separate a working beam using a sharp-edged skimmer of 35° external half-angle and approximately 1-min entry-hole diameter, and similar skimmer geometry has been used successfully many times since. For the hypersonic Mach numbers now customary in the expansion process (whether it be one or two stage), elimination of external shock effects on the skimmer is no longer a problem at sufficiently large orifice-skimmer separations, even for skimmers with external half-angles as large as 52°, as shown by Bier and Hagena.[33] This paper provides the most recent study of skimmer geometry and indicates experimentally that making the internal divergence angle as large as possible is very necessary, since 20° internal half-angle drastically reduced the beam, whereas 50° did not.

Both these angles were larger than the old Kantrowitz-Grey criterion of the angle whose tangent is the inverse speed ratio of the molecules coming through the skimmer entry, the most probable thermal speed divided by the bulk velocity. Skinner[40, 41] suggests an explanation for the importance of the internal divergence angle, which is appropriate for large-flux-density highly collimated streams. The role of small numbers of slow-moving background molecules in the region of the skimmer entry may be very significant in forming collision-product pairs, which collide with the internal skimmer walls to become further low-energy scatterers in a chain mechanism, which rapidly builds up to choke off the beam. Thus, rapidly diverging internal skimmer walls should be important in allowing easy escape of the scattered molecules into the collimation chamber pump.

Separation Effects in Binary Mixtures

Because of the large possible velocity augmentation using the seeding technique mentioned earlier, binary gas mixtures are of basic interest in beam generation. Beams isolated from

$$n_c \approx \frac{n_0 M_s{}^2 d^{s2}\gamma/8L}{\{1 + [(\gamma - 1)/2]M_s{}^2\}^{1/(\gamma-1)} + (\pi\sigma^2 d_s n_0/2^{1/2})[1 - (\gamma/2\pi)^{1/2}(M_s d_s/2L)]} \tag{17}$$

At relatively low Mach numbers, the second half of the denominator, which represents the collisional effects, dominates. If skimming takes place at high Mach numbers, the first term only is important, and thus, a maximum in the beam intensity $n_c u$ is predicted. Using σ as a curve-fitting parameter to match the prediction of Eq. (17) with the data of Deckers and Fenn[28] at the intensity maximum, Valleau and Deckers obtain a reasonable agreement between the experimental and theoretical curves. Thus the situation at present is that two explanations are available to explain the intensity curve shape in the interesting region near the maximum, with experimental evidence available which supports the idea of a shock disturbance external to the skimmer and quantitative analysis available which clearly shows that collisions after skim-

binary jets have been shown to have the calculated velocity but in addition can be much enriched in the heavier component. Many studies of these separation phenomena, not all directed at beam generation, show that the degree of enrichment depends markedly on gas conditions and geometry. A skimmer or sampling probe, moved along the free-jet axis usually indicates an increasing enrichment with increasing distance to some maximum value. Further increase in distance results in a decrease in the observed enrichment sometimes to the point that the enrichment is reversed, and an excess of the light component is observed.

Becker and his associates have studied this separation extensively in the "Trenndüse" series of papers, with a view toward beam formation as well as toward other applications, such as isotope separation.[31, 37–39, 42–46] Separations achieved in the expansion and skimming process are surprisingly large. In the paper by Becker and Henkes,[39] the results indicate that an almost completely pure beam of argon was obtained from a source mixture of 2-mole-% argon in hydrogen. In the recent paper by Klingelhofer and Lohse,[46] a 1% argon-99% hydrogen mixture in a source at 3.15 atm pressure was reported to have produced a beam that was 40% argon. (The other beam parameters were 1.7×10^{17} molecules/cm^2-sec and a beam velocity of 2130 m/sec or 1.1 ev for argon, which is 94% of the terminal velocity.)

Differing explanations of the separation effects have been offered. Becker[38] at first noticed that the gas in the jet had to reach relatively low density before separation effects occurred, and he proposed that, if both species in the expansion reached the same bulk velocity and temperature before collisions became unimportant, then the larger thermal velocities of the lighter species would cause it to spread radially more rapidly. He later abandoned this explanation in favor of a mechanism based on continuum concepts, the net effect of pressure, temperature, and concentration diffusion acting because of the gradients existing in the free jet,[42] although no quantitative analysis was attempted. However, Waterman and Stern[47] and Stern, Waterman, and Sinclair[48] expanded the free-molecular separation argument. Assuming a homogeneous jet up to a cut-off plane at which collisions ceased, they calculated the flux of each species that would pass through the skimmer downstream by treating the cut-off plane as a distribution of sources of both gases. Using the axial position of this plane as a data-fitting parameter, they were able to achieve partial agreement with their experimental data.

In another study, Chow[49] suggested that diffusive separation was responsible for the effects he apparently observed in relatively large low-pressure-ratio free jets of air, using a small mass-sampling probe. Again, no quantitative analysis was attempted. However, because systematic probe effects were suspected, Masson[50] at the same laboratory attempted to analyze separation effects possible due to the gradients in the stagnation region at the tip of a probe in a supersonic stream.

The real difficulty in assessing exactly which mechanism is responsible for the observed separation lies in the fact that all experiments have introduced a skimmer or probe as a disturbance, so that it is difficult to tell how much real separation existed in the jet without them. In fact, in their recent study, Reis and Fenn[14] have proposed that all separation effects are caused by the skimmer or probe itself, citing evidence that a sampling impact probe with a detached shock indicated separations, whereas a cone-static probe configuration indicated no separation anywhere in the jet.

Finally, the second paper by Valleau and Deckers[35] suggests and analyzes a different mechanism and obtains encouraging agreement with experimentally observed beam enrichment. The arguments parallel those mentioned earlier which ascribe the beam-intensity maximum to self-scattering of the beam downstream of the skimming plane. Laying aside the question of a shock or external disturbance in front of the skimmer, they consider a gas passing through the skimmer which is radially expanding and has a nonnegligible self-collision frequency after skimming. Assuming that collisions between light and heavy molecules will result only in scattering the light molecule out of the beam, a result is obtained for n_{1c}, the density of the heavy component 1 at the collimator,

$$n_{1c} \approx \frac{x_1 n_0 M_s^2 \langle\gamma\rangle d_s^2 m_1/8\langle m\rangle L^2}{\{1 + [(\gamma - 1)/2]M_s^2\}^{1/\gamma-1} + \pi\sigma_1^2 d_s^2 x_1 n_0/2^{1/2}} \tag{18}$$

where x_1 is the mole fraction of component 1 as the source, m_1 is the molecular weight of component 1, and $\langle m\rangle$ is the mean molecular weight in the source.

Valleau and Deckers have compared this equation with experiment for various mixtures of nitrogen and hydrogen expanded from a room temperature source at 20 torr pressure, with an orifice-skimmer distance of 5.5 orifice diameters. The comparison shows remarkable agreement, both theory and experiment showing a very strongly peaked maximum in nitrogen beam intensity for 0.9-mole fraction hydrogen in the source.

As previously, it may only be concluded, in the absence of definitive experiments, that shock phenomena external to the skimmer and self-scattering downstream of the skimmer could both play a part in the enrichment observed at orifice-skimmer separations at which the beam molecules are still colliding. At very large separations where no collisional effects are left, either in the beam or as a result of the skimmer, the enrichment factor predicted from Eq. (18) (beam-flux-intensity ratio observed divided by mole ratio in the source) approaches the molecular-weight ratio m_1/m_2, as observed by French and O'Keefe,[30] Greene, Brewer, and Milne,[25] and others. Thus, if optimum separation is required, it appears to be very advantageous to perform the skimming at a point in the expansion where collisions, whatever the mechanism, will be of assistance in eliminating the lighter unwanted gas.

Conclusion

The feasibility and advantages of producing molecular beams from continuum sources have been clearly established. Although understanding of all aspects of the formation process is still incomplete, the advances made over the past three years (as evidenced by the dates of many of the references) have been remarkable. Many aspects of the expansion process of a free jet and the behavior of the Mach cell, even for extreme pressure ratios, have been investigated and clarified. It has been shown that the formation of a beam by skimming can be successfully accomplished, although the mode of operation of the skimmer in the interesting transition region is not completely understood. This is particularly true of the role of flow disturbances due to external shocks, internally reflected molecules, and self-scattering of the beam immediately after skimming in affecting final beam intensity and the separation of gas mixtures.

A catalog of beam designs and performances achieved to date would serve little purpose. To summarize briefly the various beam parameters quoted, the best intensity performances have been achieved by Campargue[51] (10^{19} hydrogen molecules/cm^2-sec) and Skinner[41] (2.4×10^{18} nitrogen molecules/cm^2-sec) using primarily empirical development approaches. The highest energies to date have been reported by Klingelhofer and Lohse[46] (1.1-ev argon atoms) using a seeded beam, and by Skinner[41] (1.2-ev nitrogen molecules) using a shock-tube driven monopulse beam. To extend the capabilities into the 10-ev range, heated-seeded beam sources[30] and arcjet heaters[34, 52] are being tested. Campargue's results[51] and those of Greene, Brewer, and Milne[24] also indicate that, by careful optimization, first- and second-stage pumps of hundreds of liters/sec pumping capacity rather than many thousands of liters/sec can be employed, so that much more compact source designs can be expected for many future applications. Exploitation of the pulsed-source technique of Bier and Hagena[33] also offers attractive reductions in pumping capacity.

Each of these various approaches has its own peculiar advantages and disadvantages, and, as in the case of wind tunnels and shock tubes, a variety of specialized forms of continuum-source beams will undoubtedly prove inevitable and desirable. The only certainty is that, with the capabilities and potential now proved, the immediate future will see these beams being applied to basic investigations of many gas-phase and surface phenomena not previously open to experiment.

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