# A Mechanical Means to Produce Intense Beams of Slow Molecules

**Manish Gupta and Dudley Herschbach***

*Department of Chemistry and Chemical Biology, Harvard University, Cambridge, Massachusetts 02138*

*Received: October 7, 1999; In Final Form: November 8, 1999*

The flow velocity of gas emerging from a supersonic nozzle mounted on a high-speed rotor can be largely canceled by the rotor velocity, thereby producing an intense beam of molecules traveling in a vacuum with translational speeds slowed to a few tens of meters per second. Centrifugal action significantly enhances the supersonic character of the gas flow from the rotating nozzle, further narrowing the spread of velocities in the emerging beam. These features are demonstrated by model calculations and experimental results for beams of Xe and for $O_2$ and $CH_3F$ seeded in Xe.

## Introduction

The advent of methods for cooling, trapping, and manipulating neutral atoms with laser light has led to dramatic achievements.[1] With atomic vapors cooled below the microkelvin range, Bose−Einstein condensation has been attained,[2] as well as hugely nonlinear optical effects[3] and an atom laser, atom interferometry, and atom lithography, all exploiting coherent matter waves.[4] To pursue such phenomena with molecules is an appealing prospect, since molecules offer a vast range of properties not available with atoms. Energy transfer processes[5] and chemical reactions[6] would exhibit pronounced quantum dynamics if molecular translation can be slowed enough to endow the molecules with deBroglie wavelengths large compared with the size of the molecules.

Since the forces available to trap neutral atoms or molecules are weak, a key requisite for trapping is a means to lower markedly their translational kinetic energy, typically below 1 K. Laser cooling methods effective for alkali atoms fail for molecules because of the complexity of their energy level structure, with its myriad vibrational and rotational components.[7] Recently, two elegant means of trapping molecules have proved successful. One method avoids the molecular cooling problem by creating alkali dimer molecules within an alkali atom trap by photoassociation,[8] but the yield of dimers is very small. The other method employs collisional relaxation by $^3$He buffer-gas, maintained by a dilution refrigerator at about 0.3 K, to produce atoms or molecules slow enough to trap.[9] In this way, Eu and Cr atoms and CaH molecules have been trapped, all species not amenable to laser cooling. Yet the aim is to further cool the trapped species (by evaporation of the fastest particles, as in cooling coffee[4]); this requires that the buffer-gas be pumped away quickly, which so far has not proved feasible before too many molecules escape the trap.

Several other techniques for slowing molecules have been proposed. Among these are momentum transfer induced by dozens of near-resonant laser frequencies,[7] deceleration by means of multiple stages of time-varying electric field gradients,[10,11] and "scooping" by an intense nonresonant laser beam.[12] Other than buffer-gas cooling, the only method thus far implemented employed 63 synchronously pulsed electric field states to slow about 1% of a metastable CO beam (precooled to 225 m/s by seeding in Xe) down to 98 m/s.[10] Here we describe a method that requires less elaborate instrumentation. It employs a supersonic molecular beam, emerging from a nozzle near the tip of a rotor with peripheral velocity high enough to cancel the flow velocity of the beam. This yields an intense beam of molecules cooled by the supersonic expansion and slowed by the contrary rotor.[13] The high-speed rotor also functions as a gas centrifuge, thereby enhancing the supersonic character of the gas flow and further reducing the temperature within the beam. We report calculations and exploratory experiments demonstrating the feasibility of the method.

## Prototype Device

Figure 1 gives a schematic view of our apparatus. Gas (at pressures up to 100 Torr) is introduced into a hollow rotor along its axis and emerges from a pinhole nozzle (50−100 $\mu$m diam) located near the tip of the rotor arm (9.90 cm from the axis). As the rotor spins, a supersonic free jet of molecules sprays out like water from a lawn sprinkler.[14] As pictured, for a narrow angular range in the plane of the rotor sweep, the emitted molecules impinge on a circular aperature (10.5 cm away, 0.32 cm diam). Those passing through the aperature reach a quadrupole mass spectrometer (ionization zone 13.5 cm beyond the aperature), backed by a channeltron ion detector. In preliminary experiments, a fast ion guage was used instead. The rotor position is monitored by a HeNe laser beam and a photodiode to provide a time-zero for time-of-flight measurements. Measurements of TOF distributions, using either the mass spectrometer or the fast ion gauge, were made by observing pulses of molecules delivered from the rotor.

The rotor, its gas feed, and the drive mechanism are shown in Figure 2. The material and shape of the tubular rotor are important factors governing its durability at high speeds.[13,15] It is made of an aluminum alloy (7075-T6), with tensile strength/density ratio ($1.8 \times 10^5$ m$^2$ s$^{-2}$) nearly as high as titanium. For ease of fabrication, the rotor arm (weight 29 g) consists of stepped cylindrical segments, approximating an optimal Gaussian profile. An adjustable screw (shown shaded) which seals the open end of the rotor tube also enables balancing the rotor. Near the rotor tip, flat on both sides, a tangential hole (1.5 mm diam) was drilled. This hole was covered by shingle patches attached by epoxy; one patch is thin stainless steel (0.012 mm thick) and bears the laser-drilled pinhole nozzle, the other is a transparent Lexan sheet (0.25 mm thick) that transmits the monitor beam from the HeNe laser. The gas inlet is a stationary

* Corresponding author.

10.1021/jp993560x CCC: $18.00 
Published on Web 11/30/1999

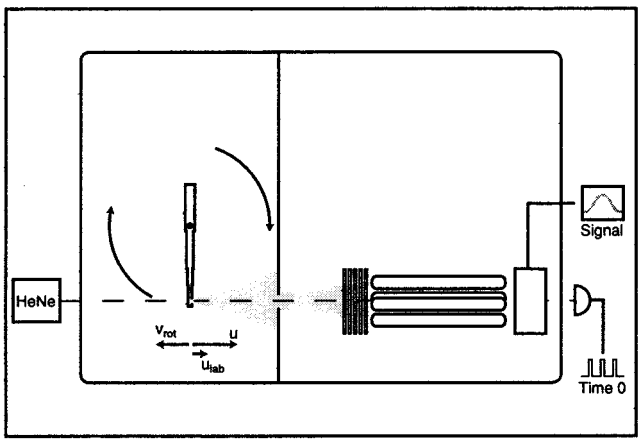


**Figure 1.** Schematic of apparatus (top view). Gas molecules (shaded cone) emerge from pinhole nozzle in rotor arm; those passing through an observation slit are detected by a quadrupole mass filter (or fast ion guage). Rotor position is monitored by a light beam (HeNe laser). Velocity of gas along centerline of the beam with respect to the nozzle is denoted by $u$, peripheral velocity of the rotor by $v_{rot}$, and resultant centerline flow velocity with respect to laboratory coordinates by $u_{lab}$.

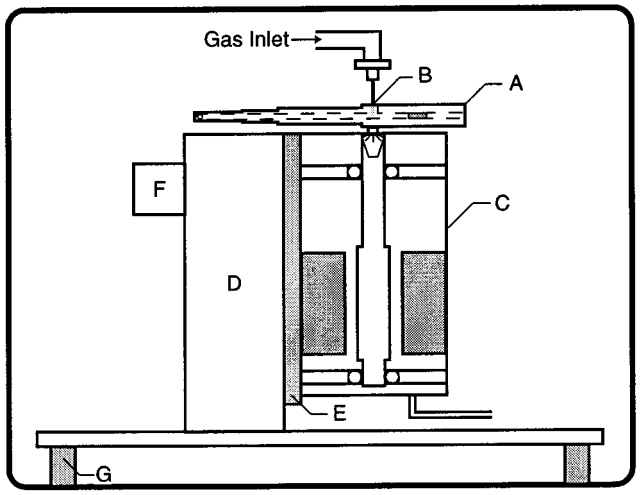


**Figure 2.** Plan view of rotor, gas feed, and drive mechanism. (For the latter, the vertical scale is reduced by a factor of 1/2). Components: A, rotor; B, needle gas inlet; C, motor; D, mounting block; E, cooling plate; F, accelerometer; G, neoprene vibration dampers. The ratio of the peripheral velocity $v_{rot}$(m/s) of the rotor to its angular velocity $\omega$-(RPM) is $1.04 \times 10^{-2}$.

tefzel needle (1.6 mm o.d.; 0.25 mm wall) which passes through a slightly larger hole in the topside of the rotor.

The rotor shaft is stainless steel, insulated with a G-10 fiberglass sheath to minimize conducting heat from the bearings to the rotor. The shaft is grasped by a collet connected to a spindle driven by a vacuum motor[16] equipped with ceramic bearings (silicon nitride) that are lubricated with vacuum grease. The entire driving unit is mounted on neoprene spacers to damp vibrations. In operation, the motor has been driven up to 42000 RMP, corresponding to a peripheral velocity of 436 m/s for the rotating nozzle.

## Velocity Distributions

The velocity distribution in a supersonic beam has the approximate form[17]

$$P(v) = v^2 \exp\{-[(v-u)/\Delta v]^2\} \quad (1)$$

Here, $u$ denotes the flow velocity along the centerline of the beam, and $\Delta v$ represents the velocity spread (fwhm $\approx 1.65\Delta v$). In terms of these parameters, the most probable velocity at the peak of the distribution is given by

$$v_{mp} = \{u + [u^2 + 4(\Delta v)^2]^{1/2}\}/2 \quad (2)$$

The velocity spread is related to the local translational temperature for relative motion of molecules within the beam by

$$\Delta v = (2k_B T/m)^{1/2} \quad (3)$$

where $k_B$ is Boltzmann's constant and $m$ is the molecular mass. When the nozzle rotates with peripheral velocity $v_{rot}$, contrary to the gas flow (as depicted in Figure 1), the resultant centerline flow velocity with respect to laboratory coordinates is reduced to $u_{lab} = u - v_{rot}$, which replaces $u$ in eqs 1 and 2. The terminal translational temperature $T$, and hence the velocity spread, is governed by the strength of the supersonic expansion; theoretical models give[17]

$$T/T_o = A(P_o d)^{-\alpha} \quad (4)$$

where $P_o$ and $T_o$ are the pressure and temperature of the gas behind the nozzle, and $d$ is the nozzle diameter; the prefactor $A$ and exponent $\alpha$ depend on molecular parameters, particularly the specific heat ratio $\gamma = C_p/C_v$. When the nozzle is rotated, centrifugal force increases the gas density at the tip of the rotor. If the "leak" out of the pinhole nozzle is small enough, the gas within can be regarded as in thermal equilibrium. Then $P_o$ near the rotor tip is larger than $P_{in}$, the input pressure of gas entering along the axis of rotation, by an exponential factor,[18]

$$P_o = P_{in} \exp[m v_{rot}^2/(2k_B T_o)] \quad (5)$$

This centrifugal effect, via eq 4, thus can substantially reduce the terminal temperature attained in the supersonic expansion.

Figure 3 shows experimental velocity distributions for beams of Xe and for $O_2$ and $CH_3F$, pure or seeded in Xe, with the nozzle stationary or rotating. Table 1 gives the corresponding values of $v_{rot}$, $P_o d$, and parameters derived from fitting eqs 1−3 to the TOF data. The amplitude of the pulsed signals obtained with the nozzle rotating corresponds to an intensity above $5 \times 10^9$ molecules/pulse, or $10^{13}$ molecules cm$^{-2}$ s$^{-1}$, for the pure Xe beam; it is about 50-fold lower for the $O_2$ and $CH_3F$ components in the seeded beams.

## Discussion

From calculations based on theoretical and empirical results for high-speed rotation[13] and supersonic expansions,[17] we expect that beams with $u_{lab} = 10$ m/s, $T = 0.3$ K, and intensity above $10^{14}$ molecules cm$^{-2}$ s$^{-1}$ can be attained. This benchmark pertains to molecular masses, rotor speeds, and input pressures such that $P_o d \geq 20$ with the aid of the centrifugal effect. Rotors have been operated up to about $v_{rot} = 2$ km/s, so cancellation of molecular flow velocities is feasible over a wide range. We specify a lower limit for $u_{lab}$ of about 10 m/s to permit molecules emitted by the nozzle to escape from the path of the rotor arm before it returns and swats them.[19] The attainable $T$ and beam intensity, both largely determined by pumping capacity, might be improved beyond the benchmark if a means to pulse the rotating nozzle can be devised.[14,20]

The results in Figure 3 are quite short of the benchmark. However, our current apparatus, intended only for exploratory experiments, is far from optimal. Inputting the gas via a needle becomes quite leaky when the rotor is spinning and in any case

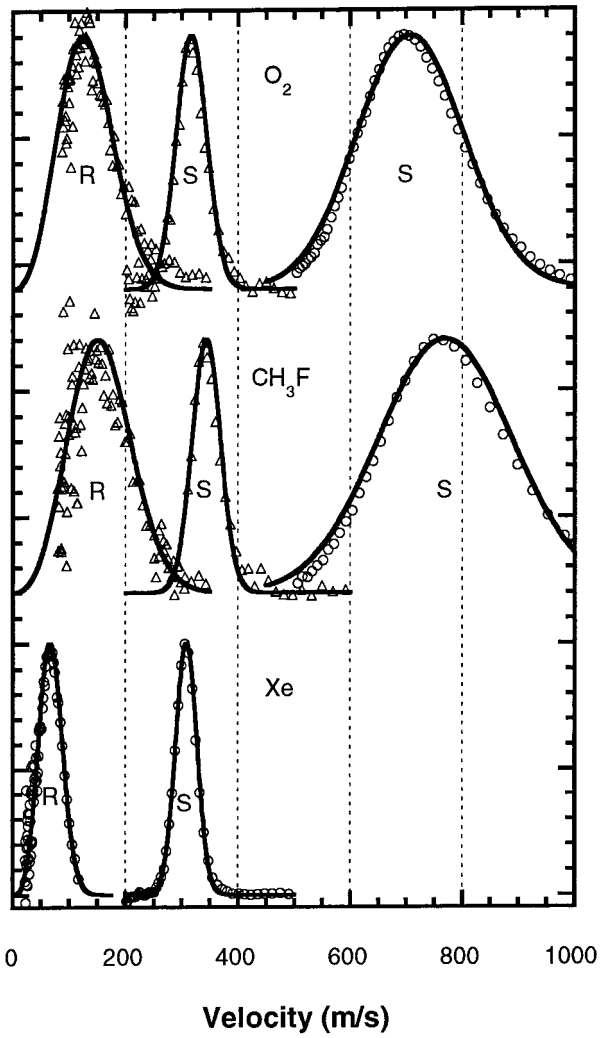


**Figure 3.** Experimental velocity distibutions for beams of Xe, $O_2$, and $CH_3F$, with nozzle stationary (S) or rotating (R). Data points for pure beams are shown by circles (○), those for $O_2$ and $CH_3F$ seeded in Xe by triangles (△). Curves show fits to eq 1 with parameters given in Table 1.

**TABLE 1: Parameters for Velocity Distributions**[a]

| species | $P_o d$ torr-cm | $v_{rot}$ m/s | $v_{mp}$ m/s | $u_{lab}$ m/s | $\Delta v$ m/s | $T$ K |
|---|---|---|---|---|---|---|
| $O_2$ | 2.00 | 0 | 707 | 680 | 138 | 37 |
| $O_2$ in Xe | 1.12 | 0 | 317 | 312 | 39 | 2.9 |
| $O_2$ in Xe | 1.83 | 248 | 125 | 67 | 85 | 14 |
| $CH_3F$ | 1.05 | 0 | 769 | 730 | 173 | 61 |
| $CH_3F$ in Xe | 1.52 | 0 | 344 | 340 | 36 | 2.6 |
| $CH_3F$ in Xe | 1.66 | 248 | 150 | 91 | 94 | 18 |
| Xe | 1.48 | 0 | 308 | 306 | 27 | 5.6 |
| Xe | 1.04 | 273 | 67 | 49 | 34 | 9 |

[a] For curves shown in Figure 3; parameters pertain to eqs 1−3; for rotating source, nominal $P_o$ computed from eq 5.

our pumping capacity is inadequate for strong supersonic beams. With the rotor stationary (which allowed the gas inlet tube to be sealed to the rotor) we could take $P_{in}$ (= $P_o$) up to 200 Torr, but with the rotor spinning (and the unsealed inlet leaky) we could only go up to $P_{in}$ = 40 Torr. Also, due to overlap of adjacent molecular pulses at low velocities ("wrap-around"), our TOF analysis could not reliably measure velocities below 100 m/s. For the pure Xe beams, we were able to extend the velocity analysis down to 40 m/s, by installing a chopper wheel between the observation slit and fast ion guage (Figure 1), which much reduced wrap-around. This was not feasible for the seeded $O_2$ or $CH_3F$ because of the reduced intensity. Augmenting the

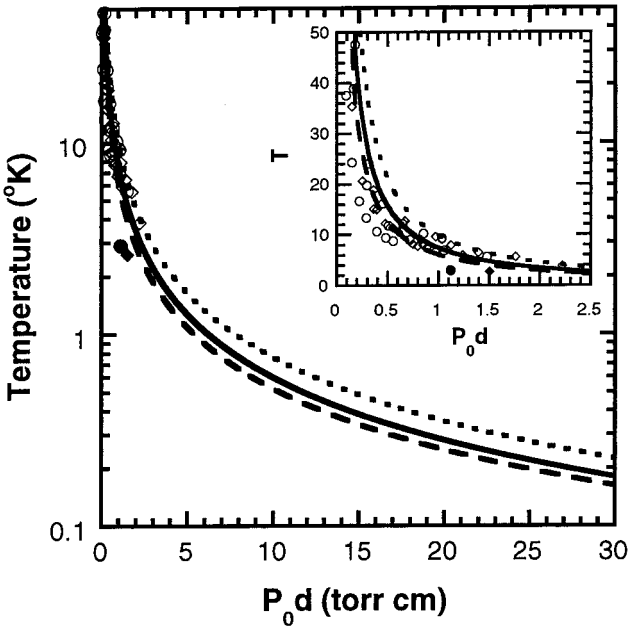


**Figure 4.** Variation of terminal translational temperature with product of nozzle diameter and stagnation pressure. Curves from eq 4: full curve for pure Xe ($A$ = 0.0246; α = 1.09), dashed ($A$ = 0.0202; α = 1.06), and dotted ($A$ = 0.0328; α = 1.11) for $O_2$ (5%) and Xe components of seeded beam, respectively. Points are obtained via eq 3 from widths of velocity distributions for pure Xe beams with nozzle stationary (○), or rotating (◇), and for seeded $O_2$ (●) or $CH_3F$ (◆) with nozzle stationary. Pressure $P_o$ includes centrifugal contribution of eq 5. Insert shows an expanded view of the low $P_o d$ range (with ordinate scale linear rather than logarithmic).

pumping capacity and eliminating the leaky input (by use of an oil gland[18] or sealing within the rotor a sample sufficient for several hours) should much improve performance. Likewise, other means (REMPI, laser fluorescence) can measure slower velocities. In auxiliary experiments (with the detector switched to the left side of Figure 1) with $v_{rot} > u$, we observed a Xe beam traveling backward at 120 m/s, demonstrating that the rotor is indeed capable of more than offsetting the flow velocity.

Figure 4 compares the variation of $T$ with $P_o d$ predicted from supersonic beam theory[17] with the terminal translational temperatures derived via eq 3 from the widths of our experimental velocity distributions (including some runs not displayed in Figure 3 and Table 1). Although indicative, this comparison is provisional. Most of our values for $T$ are somewhat high since we have not made convolution corrections for wrap-around. Also, we used a nominal $P_o$ obtained from eq 5, which likely overestimates the centrifugal effect, particularly as the gas flow increases at higher rotor speeds. In runs varying $v_{rot}$ for pure Xe and Kr beams, we found the variation of $T$ to be consistent with this nominal $P_o$. However, our more limited data for seeded beams indicate $T$ for both components is appreciably higher than expected from the nominal $P_o$. This could arise if the centrifugal factor for the diluent Xe gas is not fully effective for seeded beams or if rotational and/or vibrational relaxation alter the expansion properties.[17] These aspects require further study, to assess whether increasing $P_{in}$ or the centrifugal factor or both will indeed enable us to reach our benchmark, $T \approx 0.3$ K.

Even our current rudimentary device may find useful applications. Aside from trapping, slowing, and cooling molecules, it also offers marked advantages for many techniques using external fields to deflect or focus molecules.[21] For all such techniques, the force exerted on the molecule is inversely

proportional to its translational kinetic energy. The most probable kinetic energy[22] of our current best $O_2$ beam is only $E_{mp}/k_B = 20$ K, and for our benchmark only 0.45 K. In particular, supplying molecules with such low kinetic energy would greatly foster deceleration and trapping by devices using electric field gradients.[10,11] Likewise, for experiments involving molecular diffraction,[4] the rotating source can provide beams with sizable deBroglie wavelengths. For our best $O_2$ beam, $\lambda_{mp}$ is 1.4 Å; for our benchmark it is 9 Å.

For initial trials, we chose $O_2$ and $CH_3F$ chiefly because these are particularly well suited for current magnetic[23] or electrostatic[24] traps or storage rings.[25] As is evident in Figure 3, slowing and cooling such light gases is facilitated by seeding them in a heavy diluent species such as Xe, although this comes at some cost of intensity. In principle, however, the rotating source can be most effective in slowing heavy molecules, either as pure beams or seeded in light diluents. This requires higher rotational speeds, but the centrifugal effect of eq 5 is then much enhanced.

When spun in the opposite direction, the rotor augments rather than cancels the flow velocity, so it enables the beam velocity, at a fixed source temperature, to be scanned over a wide range. In runs using both slowing and speeding modes with Kr beams, we found that the centrifugal effect operates equally well in both modes (lowering $T$ about 5-fold as $v_{rot}$ went from zero to $\pm$311 m/s). The rotating supersonic source provides a much narrower velocity spread than the swatting technique for molecular acceleration,[13] which relied on thermal evaporation from the rotor tip.

This whirling variant of a supersonic beam, with improvements in prospect, offers a versatile and relatively simple means to enhance a host of experiments dependent on molecular velocities. It is thus apt to recall that the very first measurements testing the Maxwell−Boltzmann distribution were made with an apparatus mounted within a rotating cylinder, devised by Otto Stern 80 years ago.[26]

**Acknowledgment.** We have enjoyed discussions about this project with Daniel Katz and Peter Siska. Thanks are also due to architect Frank Gehry; his awesome Guggenheim Bilbao Museum in Bilbao, sheathed in titanium, aroused in the senior author thoughts of high-speed rotation as a route to slow molecules. We are grateful for support received from the Research Corporation, the National Science Foundation, and the Milton Fund of Harvard University.

## References and Notes

(1) Chu, S. *Rev. Mod. Phys.* **1998**, *70*, 685; Cohen-Tannoudji, C. N. *ibid.* **1998**, *70*, 707; Phillips, W. D. *ibid.* **1998**, *70*, 721.

(2) Fried, D. G.; Killian, T. C.; Willmann, L.; Candhuis, D.; Moss, S. C.; Kleppner, D.; Greytak, T. J. *Phys. Rev. Lett.* **1998**, *81*, 3811, and work cited therein.

(3) Hau, L. V.; Harris, S. E.; Dutton, Z.; Behroozi, C. H. *Nature* **1999**, *397*, 594.

(4) Wieman, C. E.; Pritchard, D. E.; Wineland, D. J. *Rev. Mod. Phys.* **1999**, *71*, S263.

(5) Forrey, R. C.; Balakrishnan, N.; Dalgarno, A.; Hafferty, M. R.; Heller, E. J. *Phys. Rev. Lett.* **1999**, *82*, 2657.

(6) Herschbach, D. R. In *Chemical Research−2000 and Beyond: Challenges and Visions*; Barkan, P., Ed.; Oxford University Press: New York, 1998; p 113; *Rev. Mod. Phys.* **1999**, *71*, S411.

(7) Bahns, J. T.; Stwalley, W. C.; Gould, P. L. *J. Chem. Phys.* **1996**, *104*, 9689.

(8) Nikolov, A. N.; Eyler, E. E.; Wang, X. T.; Li, J.; Wang, H.; Stwalley, W. C.; Gould, P. L. *Phys. Rev. Lett.* **1999**, *82*, 703, and papers cited therein.

(9) Weinstein, J. D.; deCarvalho, R.; Guillet, T.; Friedrich, B.; Doyle, J. M. *Nature* **1998**, *395*, 148.

(10) Bethlem, H. L.; Berden, G.; Meijer, G. *Phys. Rev. Lett.* **1999**, *83*, 1558.

(11) Maddi, J. A.; Dinneen, T. P.; Gould, H. *Phys. Rev. A*, **1999**, *60*, 3882.

(12) Friedrich, B. *Phys. Rev. A*, accepted.

(13) Accelerating molecular beams by a high-speed rotor was pioneered 50 years ago by Philip Moon; see Moon, P. B. *Proc. R. Soc. London A* **1978**, *360*, 303, and work cited therein. Moon considered effusion from a hollow rotor (J. S. Simon, private communication) but opted instead to produce fast molecules by swatting.

(14) For many applications it would be preferable to produce a roughly unidirectional beam rather than a 360° spray. This may be feasible by use of a fast piezoelectrically activated valve, pulsed synchronously with the rotor. For stationary sources, such a value operating at kHz frequencies has been devised by D. Gerlich, Institut für Physik, Technische Universität, Chemnitz (private communication).

(15) The rotor described here has operated without mishap for several months in over 100 runs; the breaking limit, computed from its mass distribution, is 615 m/s. Earlier versions, of symmetrical double-bladed form, promptly broke apart when spun. At $v_{rot} = 350$ m/s, the force/mass ratio at the rotor tip is more than $10^5$ times that due to gravity.

(16) Elettromeccanica, Giordano Colombo, sri, Milan.

(17) Miller, D. R. In *Atomic and Molecular Beam Methods*, Vol. I; Scoles, G., Ed.; Oxford University Press: New York, 1988; p 14.

(18) Beams, J. W. *Rev. Mod. Phys.* **1938**, *10*, 245.

(19) Although in our slowing mode $u_{lab}$ cannot be reduced to zero because of swatting, most of this can be avoided by aiming the nozzle to direct the beam centerline slightly below the plane in which the rotor arm whirls. Even a small downward velocity component ($\sim$5 m/s) allows molecules to escape swatting and can serve to carry them to a collection device, such as a storage ring.

(20) Gentry, R. W. In *Atomic and Molecular Beam Methods*, Vol. I; Scoles, G., Ed.; Oxford University Press: New York, 1988; p 54. See also Beck, S. M.; Liverman, M. G.; Monts, D. L.; Smalley, R. F. *J. Chem. Phys.* **1979**, *70*, 232.

(21) Seideman, T. *J. Chem. Phys.* **1997**, *107*, 10420, and work cited therein.

(22) With appropriate Jacobian factors included, the kinetic energy distribution equivalent to eq 1 has the form $E^{1/2} \exp[-(E - 2E_u^{1/2}E^{1/2} + E_u^2)/T]$ with $E_u = 1/2mu^2$ and $T$ from eq 3. The corresponding distribution of deBroglie wavelengths is $\lambda^{-4} \exp[-\lambda_o^2(\lambda^{-1} - \lambda_u^{-1})^2]$, with $\lambda_o = h/(m\Delta v)$ and $\lambda_u = h/(mu)$.

(23) Friedrich, B.; deCarvalho, R.; Kim, J.; Patterson, D.; Weinstein, J. D.; Doyle, J. M. *J. Chem. Soc., Faraday Trans.* **1998**, *94*, 1783.

(24) Jongma, R. T.; von Helden, G.; Berden, G.; Meijer, G. *Chem. Phys. Lett.* **1997**, *170*, 304.

(25) Katz, D. P. *J. Chem. Phys.* **1997**, *107*, 8491.

(26) Stern, O. *Z. Physik* **1920**, *2*, 29; **1920**, *3*, 417. See Ramsey, N. F. *Molecular Beams*; Oxford University Press: New York, 1956; p 21.