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Intense atomic and molecular beams via neon buffer-gas cooling

David Patterson, Julia Rasmussen and John M Doyle¹

Physics Department, Harvard University, Cambridge, MA, USA

E-mail: doyle@physics.harvard.edu

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Abstract. We realize a continuous, intense, cold molecular and atomic beam source based on buffer-gas cooling. Hot vapor (up to 600 K) from an oven is mixed with cold (15 K) neon buffer gas, and then emitted into a high-flux beam. The novel use of cold neon as a buffer gas produces a forward velocity distribution and low-energy tail that is comparable to much colder helium-based sources. We expect this source to be trivially generalizable to a very wide range of atomic and molecular species with significant vapor pressure below 1000 K. The source has properties that make it a good starting point for laser cooling of molecules or atoms, cold collision studies, trapping, or nonlinear optics in buffer-gas-cooled atomic or molecular gases. A continuous guided beam of cold deuterated ammonia with a flux of 3×10^{11} ND₃ molecules s⁻¹ and a continuous free-space beam of cold potassium with a flux of 1×10^{16} K atoms s⁻¹ are realized.

¹ Author to whom any correspondence should be addressed.

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1. Introduction

Cold, slow, laser-cooled atomic sources of atoms have been laboratory standards for over a decade but only recently have cold, slow sources of molecules been demonstrated [1]. This is largely because the complex level structure of molecules makes laser cooling, the workhorse of atom cooling, very difficult for the vast majority of molecules. (Laser cooling of molecules has yet to be demonstrated, although a recent proposal indicates a path towards this with some molecules [2].) Techniques to produce cold, low-velocity samples of molecules are buffer-gas cooling [3], slow molecule filtering from a warm, effusive source [4]–[6], mechanical slowing [7], deceleration of molecules from a seeded supersonic source using time-varying electric fields [1, 8, 9], photoassociation of cold and ultracold atoms [10], and the formation of molecules from ultracold atoms using Feshbach resonances [11]. In general, the fluxes achieved with these molecular sources have been far lower than those achieved in cold atom sources.

1.1. Buffer-gas cooling and direct injection of atoms and molecules

Cryogenic helium buffer-gas cooling has proven to be a versatile tool for producing cold, dense stationary gases and high-flux beams of cold atoms and molecules [3, 12]. The species of interest is injected into a cell containing cold helium gas where it thermalizes, producing a cold mixture with the helium. Until recently, the primary methods of injection have been laser ablation, *in situ* electric discharge and direct introduction of hot molecular beams. Ablation and discharges deposit far more energy into the buffer gas than is necessary for cooling the injected species. For example, in laser ablation of a typical metal, a laser pulse with an energy of ≈ 10 mJ will result in $\approx 10^{12}$ buffer-gas-cooled atoms. Assuming a heat of vaporization of 1000 K per atom, this represents an efficiency of 10^{-4} [13]. Even in the best cases, low efficiencies limit the available flux of cold atoms or molecules, and generally requires experiments to be run in a pulsed mode.

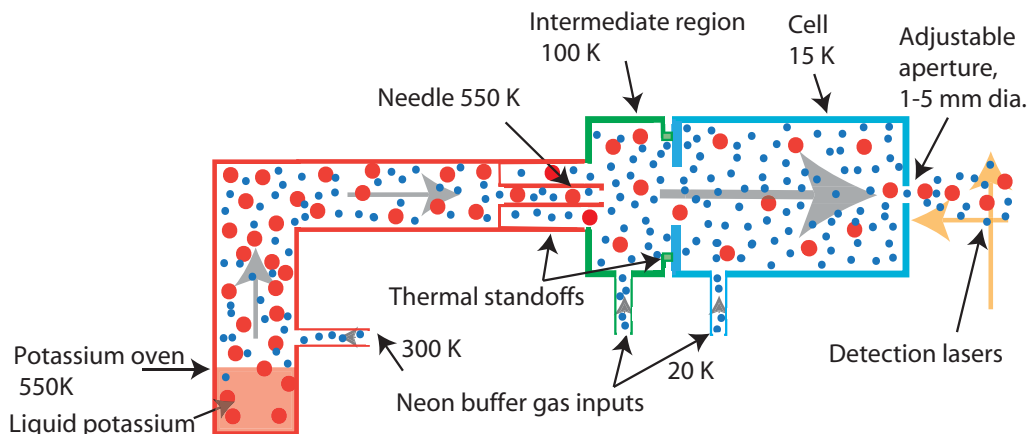


Figure 1. Overview of apparatus. An oven supplies a hot (550 K) mixture of potassium and neon that flows through an intermediate region (100 K) to a cold tube ‘cell’ (15 K). The oven, intermediate plate, and cells are separated by thermal standoffs to reduce heat loads. A cold beam of potassium and neon exits an adjustable aperture on the far side of the cell where its characteristics are measured by absorption spectroscopy.

Directly injecting a hot molecular beam is very energy efficient, but so far has been limited to peak input fluxes of 5×10^{13} molecules s^{-1} in 10 ms pulses [14].

Using helium buffer-gas cooling, we previously [12] produced a continuous beam of cold molecular oxygen, $^{16}\text{O}_2$ guided 30 cm from a cryogenic cell to a room temperature UHV chamber. In that work, oxygen gas was flowed into the cold ($T \approx 1.5$ K) helium buffer gas through a $T \approx 60$ K capillary line. The oxygen cooled and then exited through a 4 mm diameter aperture. The resultant beam was used to load a magnetic guide at high molecular flux. In this work, we present substantial developments of this technique and demonstrate them by continuous production of large fluxes of cold potassium atoms and deuterated ammonia molecules. We employ a much hotter capillary ($T = 600$ K) and use, in addition to helium, a neon buffer gas. We show that the use of neon as a buffer gas leads to beams with more slow atoms or molecules than helium-based beams, even though the neon gas is at a significantly higher temperature (due to vapor pressure constraints). This new variant of buffer-gas beam sources represents a major departure from previous approaches and can be trivially generalized to provide a continuous, high-flux source of essentially any species that has a significant vapor pressure below 1000 K.

2. Apparatus

Our potassium beam apparatus is centered around a cold tube (the ‘cell’), $T = 6\text{--}15$ K, into which we flow target atoms or molecules and inert gas, as depicted in figure 1. This cell is the coldest part of the apparatus and is 1 cm in diameter and 8 cm long. A hot (up to 600 K) gas mixture of neon and potassium flows into one end of the tube. The mixture cools to the temperature of the tube walls as it flows down the tube and out into vacuum, realizing the beam.

In more detail, a hot (up to 600 K) copper tube is attached via a 2 cm long stainless steel tube to a copper plate (about 100 K), which in turn is anchored to the first stage (40 K) of

a closed cycle pulse-tube refrigerator [15]. This plate absorbs the bulk of the radiative and conductive heat load from the hot input tube, as well as possibly precooling the gas mixture. The tube continues via a 1 cm long, 1 cm diameter, thermally isolating section of epoxy/fiberglass composite and enters the cold cell through a 6 mm aperture in a copper plate. The cold cell is thermally connected to the second stage (6 K) of the pulse tube refrigerator and has an adjustable aperture (typically 3 mm diameter) at the far end. Total heat loads on the cryorefrigerator from the apparatus are about 40 W at the first stage and 1 W at the second stage, which coincides both with the cooling power of the pulse tube and with typical heat loads in a nitrogen/helium liquid cryogen cryostat. The total distance between the end of the hot copper tube and the entrance to the cell is 1.5 cm. Our current maximum running temperature of 600 K is limited by this 40 W heat load on the first stage of the cryocooler, and could be significantly increased by better thermal isolation, or the construction of an additional intermediate cooling plate at 300 K.

Buffer gas can be introduced—in combination or separately—at the oven (600 K), the first copper plate (100 K) or the cell (15 K). The temperature of the gas mixture at the exit of the cell is measured to be ≈ 15 K regardless of where the gas is introduced (see section 3 below); the flux of cold potassium is maximized when the buffer gas flow through the oven dominates over the flow into the plate or cell, likely because this flow transports the comparatively low-pressure potassium (1 Torr at 600 K) down the long hot copper tube and cooling occurs directly within the cell. Our potassium flux is limited by the vapor pressure of potassium at our current maximum running temperature of 600 K, suggesting that fluxes could be substantially higher if we were using a species with a higher vapor pressure. The oven and transport tube could be run at higher temperature with straightforward engineering improvements to the thermal isolation. Further details of the apparatus, including the design for our high-speed charcoal cryopumps, are described in [12].

3. Thermalization and loss to the cell walls

The key physical process that leads to production of cold potassium is the flow and thermalization of the initially hot potassium/buffer-gas mixture within the cell. In order to realize a cold, high-flux beam, the experimental conditions generally need to satisfy two competing conditions. The gas mixture must thermalize with the cold walls, and the mixture must exit the cell (a tube) before much of the potassium diffuses to the walls, where it has a high probability of permanently sticking.

It is not clear *a priori* that it is possible to achieve thermalization without significant atom loss. Assuming a tube of radius r , a neon density of n_{Ne} , a gas temperature T , and hard sphere elastic cross sections of $\sigma_{\text{Ne-Ne}}$ and $\sigma_{\text{K-Ne}}$, respectively, the characteristic time τ for the gas to thermalize with the walls is

$$\tau = r^2 n_{\text{Ne}} \sigma_{\text{Ne-Ne}} (m_{\text{Ne}}/2k_{\text{B}}T)^{1/2}.$$

Assuming that $n_{\text{Ne}} \gg n_{\text{K}}$, the diffusive loss of potassium to the walls will occur on a timescale τ_{loss} :

$$\tau_{\text{loss}} = r^2 n_{\text{Ne}} \sigma_{\text{K-Ne}} (m_{\text{Ne}}/2k_{\text{B}}T)^{1/2}.$$

These expressions imply that in order to thermalize with only order unity loss in atom flux, we need $\sigma_{\text{Ne-Ne}} \lesssim \sigma_{\text{K-Ne}}$. Although we did not make accurate density measurements in

our cell under actual running conditions due to the high (>100) optical density, we have evidence that loss to the cell walls is minimal; we have been unable to find a regime where the temperature of the beam differs significantly from the measured temperature of the cell walls. We therefore speculate that $\sigma_{\text{Ne-Ne}} < \sigma_{\text{K-Ne}}$, but note that this conclusion and the sensitivity of the overall cooling process to the ratio of these cross sections remains open to both theoretical and experimental investigation.

4. Neon as a buffer gas

In order to achieve the lowest possible temperature, the overwhelming majority of buffer-gas cooling experiments to date have used helium as the buffer gas. With a sufficiently high vapor pressure at temperatures as low as 300 mK (for ^3He), compared to a lower limit of ≈ 12 K for neon, helium is the natural choice when low temperature is the only figure of merit.

Both magnetic and electrostatic traps have depths of at most a few K. It is therefore natural to maximize the flux of molecules with a total energy less than a given energy $E_{\text{trappable}} \approx 1$ K, assumed here to be less than $T_{\text{cell}} \approx 4\text{--}20$ K. Somewhat counterintuitively, by this metric a hydrodynamic [12] beam using 15 K neon as the buffer gas can substantially outperform a similar beam using 4 K helium.

Consider a beam consisting of a small fraction of potassium ‘impurities’ of mass m_{K} mixed with a much larger flow of a noble gas of mass m . The mixture is assumed to thermalize to a temperature T before it exits the nozzle. Depending on the number of collisions within and just outside the nozzle, such a beam can operate in one of three regimes:

1. *Effusive*, characterized by $N < 1$ collision. In such beams, only the atoms in the cell which happen to diffuse through the small nozzle aperture are emitted into the beam. These beams have a forward velocity of order $\bar{v} \approx (2k_{\text{B}}T/m_{\text{K}})^{1/2}$ and relatively low flux.
2. *Hydrodynamic*, characterized by $1 < N \lesssim 100$ collisions; the flow through the nozzle is fluid dynamic, but isothermal. These beams typically have a large flux enhancement compared to effusive beams due to hydrodynamics within the cell, as described in [12]. Hydrodynamic beams have a modestly boosted mean forward velocity of $\bar{v} \approx (2k_{\text{B}}T/m)^{1/2}$.
3. *Fully supersonic*, characterized by $N > 1000$ collisions in and outside the nozzle. In this regime, there is significant adiabatic cooling as the gas expands into the vacuum; the forward velocity distribution is sharply peaked around a velocity $\bar{v} \gtrsim 2 \times (2k_{\text{B}}T/m)^{1/2}$.

Figure 2 shows the forward energy spectrum to be expected in each of these regimes.

In order to take advantage of large flux enhancements, we run our beams in the hydrodynamic regime described above (blue curve in figure 2). Figure 3 shows the simulated forward velocity distribution for such a source using 4.2 K helium, 15 K neon and 40 K argon as the buffer-gas. It is clear that despite the higher temperature required for a neon or argon beam, there are more cold, low-energy atoms in a neon-cooled beam than in a helium-cooled beam.

Using neon instead of helium as the buffer gas carries several important technical advantages as well. Most notably, neon has a negligible vapor pressure at our cryostat’s base temperature of 5 K, meaning that any cold surface becomes a high-speed cryopump. The vacuum outside the cell can be maintained at a high level (1×10^{-6} Torr in the main chamber and $< 10^{-8}$ Torr in a differentially cryopumped ‘experiment chamber’) even with a high flow of

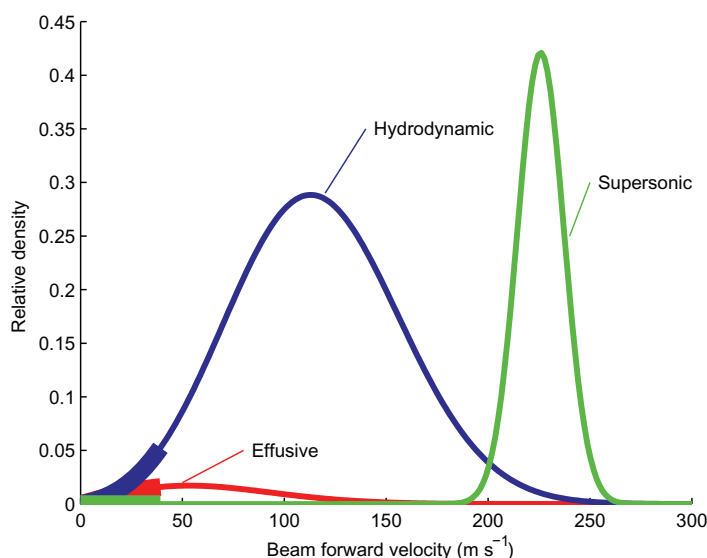


Figure 2. Simulated forward velocity distributions for buffer gas beams in various regimes. As a mixture of potassium and neon buffer gas emerges from the nozzle, a typical potassium atom collides with neon atoms as it moves away from the nozzle aperture. Qualitatively, effusive beams suffer zero collisions, ‘hydrodynamic’ beams a few collisions, and supersonic beams many collisions, remaining fluid dynamic for many aperture diameters. The beams demonstrated in this paper are hydrodynamic, but generally not supersonic (blue curve).

order 10^{19} neon atoms s^{-1} through the cell. In future experiments, simple shutters could be used to rapidly close off this differentially pumped chamber, which we believe will rapidly achieve an extremely high vacuum. The higher temperature means the cell can tolerate substantially higher heat loads. The principle disadvantage of neon is that the higher temperature leads to less rotational cooling in molecules than that available from 4 K helium.

5. Potassium results

A cold, high-flux beam of potassium was characterized to study this new beam system. The density, transverse velocity distribution and longitudinal velocity distribution are inferred from the amplitude and Doppler broadening of the absorption spectra. Two lasers (potassium D2 line, 767 nm) are used to characterize the atomic beam. The first, used to measure the beam flux and transverse velocity distribution, passes through the center of the combined neon/potassium beam 2 cm away from the nozzle.

The second laser propagates parallel to and slightly below the atomic beam, intersecting the edge of the beam. This laser is used to measure the forward velocity distribution of the beam. Figure 4 shows typical measured velocity distributions.

Under typical running conditions (potassium oven and injection needle heated to 580 K, the cell held at 17 K, 110 sccm of neon buffer-gas flowing), we measure an optical density of about five in the transverse laser path, and a mean forward velocity of 130 m s^{-1} , in excellent agreement with a simple simulation of a hydrodynamic beam at 17 K. These measurements

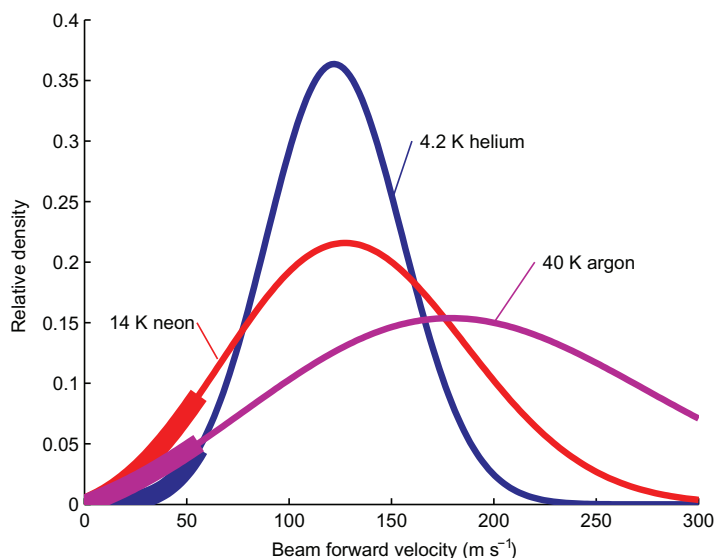


Figure 3. Theoretical energy spectrum for hydrodynamic beams using 4.2 K helium, 14 K neon, or 40 K argon as a buffer gas. The portion of the distribution with an energy of less than 4 K is shown in bold. Neon is clearly an attractive choice for experiments interested in maximizing the flux of very cold atoms or molecules.

imply a density of about 8×10^{11} potassium cm^{-3} with an atomic beam diameter of about 1 cm and a total beam flux of 1×10^{16} atoms s^{-1} . The beam divergence is measured to be about 0.7 sr, consistent with our model of beams in this hydrodynamic regime.

6. Applications

6.1. Demonstrated: loading an electrostatic beamguide

It is natural for a variety of experiments to want a cold beam of molecules that has been separated from the helium or neon buffer gas. As an example application of this source, we have demonstrated loading a cold beam of ND_3 into a curved electrostatic guide; the output of such a guide consists of pure, state-selected ND_3 molecules. (We note that after our ND_3 experimental work was completed we learned of similar results presented in [16].) In addition, the guide acts as a filter that passes only molecules moving slowly enough to be guided around the bend. Such a guide can be used to bring the beam into a differentially pumped region, either cryogenic (this work) or at room temperature [12, 16].

The apparatus for our guided ND_3 work is simpler than for our high-flux potassium beam. Specifically, the ND_3 is fed directly into the cell and the capillary temperature is a lower $T = 280$ K. Cold ND_3 molecules exit the cell and after a short gap of 30 mm, enter an electrostatic guide. ND_3 molecules exiting the guide were detected via 2 + 1 REMPI using a 6 mJ pulsed laser at 317 nm. The guide consists of six 1 mm diameter stainless steel rods with a center–center spacing of 2 mm. The guide entrance is positioned 30 mm from the exit aperture of the cell, slightly beyond the expected position of the last collision between a typical ND_3 molecule and buffer gas atom. The position of this last collision is velocity dependent, with slow

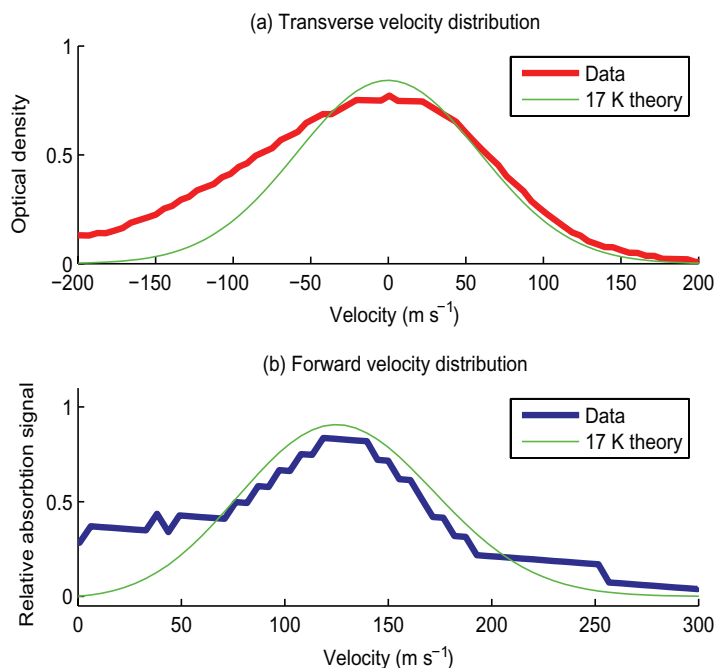


Figure 4. Measured transverse (a) and forward (b) velocity distribution of a high-flux beam of potassium. The green curves represent the Doppler broadened spectrum expected from the velocity distribution of a hydrodynamic beam with $T_{\text{gas}} = 17$ K. The amplitude is the only free parameter in these fits. The left side of each plot is somewhat distorted by the nearby $F = 0$ hyperfine line. These data represent a beam with a potassium flux of 1×10^{15} atoms s^{-1} ; we have observed fluxes as high as 1×10^{16} atoms s^{-1} , but they are not shown here because the high optical density (≈ 8) makes their interpretation less straightforward.

molecules suffering more collisions. We therefore speculate that the dominant loss mechanism for the very slowest molecules in the guided velocity distribution is collisions with buffer-gas atoms in the first few cm of the guide. A schematic of the guide and ion detection assembly is shown in figure 5.

The guide is constructed from alternating rods held at 5 kV and ground, producing a linear hexapole field. The guide rods were held in place with Ultem brackets; the entire assembly was mounted to the 6 K cold plate. No detectable ($< 0.1 \mu\text{A}$) leakage current flowed along the surface of the Ultem brackets despite the fact that few precautions were taken to avoid such currents. We speculate that such surface currents are suppressed at low temperatures. The guide is 70 cm long and describes a complete loop with a bend radius of 8 cm. The first 30 cm of the guide can be switched on or off independently from the rest of the guide.

In a typical experimental run, the first 30 cm of the guide (the ‘gate’) is initially turned off and the remainder of the guide is charged. The first section is then turned on, and, after a variable delay, the pulsed REMPI laser is fired, ionizing ammonia molecules a few cm away from the guide exit. These ions are then detected by a multi-channel plate.

The velocity distribution in the guide, shown in figure 6, is inferred by varying the delay between opening the gate and detection. We can only present a conservative lower bound on the guided molecule flux as absolute density measurements are notoriously unreliable in MPI

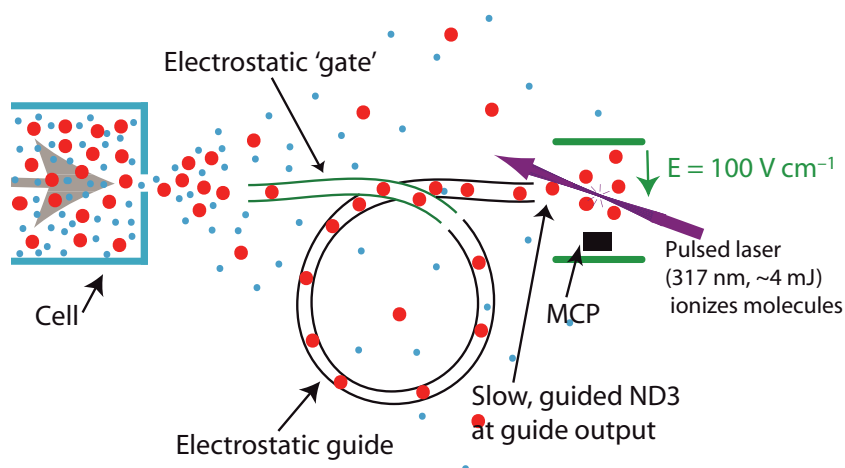


Figure 5. Apparatus overview for guiding and detecting ND₃ molecules. As in [4], the bent guide passes only slow moving low field seeking molecules.

experiments; our quoted flux is based on very conservative comparisons with similar, calibrated experiments on ND₃ done in other groups [17]. With our most sharply bent guide (figure 6(b)), we find a lower bound of 3×10^8 guided molecules s⁻¹ with an average energy corresponding to a temperature of about 4 K.

The broad linewidth (≈ 5 GHz) of the pulsed laser used for the 2 + 1 REMPI spectroscopy makes direct measurement of the beam velocity via Doppler spectroscopy impossible. The longitudinal velocity distribution is measured via the gate/time-of-flight method described above, while the transverse velocity distribution is limited by the guide depth of ≈ 600 mK. In addition, the rotational temperature of both the guided and unguided beams was found to be less than 20 K in all cases, and in excellent agreement with rotational ND₃ spectra taken in supersonic beams [17].

The ND₃ beam was initially demonstrated using both helium and neon as a buffer gas. Figure 6(a) shows the measured velocity distribution of a guided beam of ND₃ using helium and neon as buffer gas under otherwise similar conditions. The guide used in this work has a significantly larger (70 cm) radius of curvature, leading to a much higher velocity cutoff in the beam guide output velocity distribution. Significantly more slow molecules are produced in the beam using neon buffer gas.

6.2. Next step I: collisional studies

There is substantial interest in studying both elastic and inelastic collisions of molecules at low energies [1]. The guided output of the deuterated ammonia beam described above represents a pure, rotationally cold, state selected, slow molecular beam. In addition, a particular velocity class can be selected by controlled switching of the guide. Such a beam could be crossed with a similar beam, a laser cooled atomic beam, a trapped sample, or a cold thermal beam of hydrogen, helium, or neon, providing a ‘lab bench’ to study cold collisions with energies of a few cm⁻¹. Such a system would provide an attractive set of ‘knobs’ to the experimenter—collision energy, incoming rotational state and external fields can all be controlled. Even a small fraction of the beam being scattered inelastically into high-field seeking states would be detectable, since the output of the beam guide consists of very pure low-field seeking states.

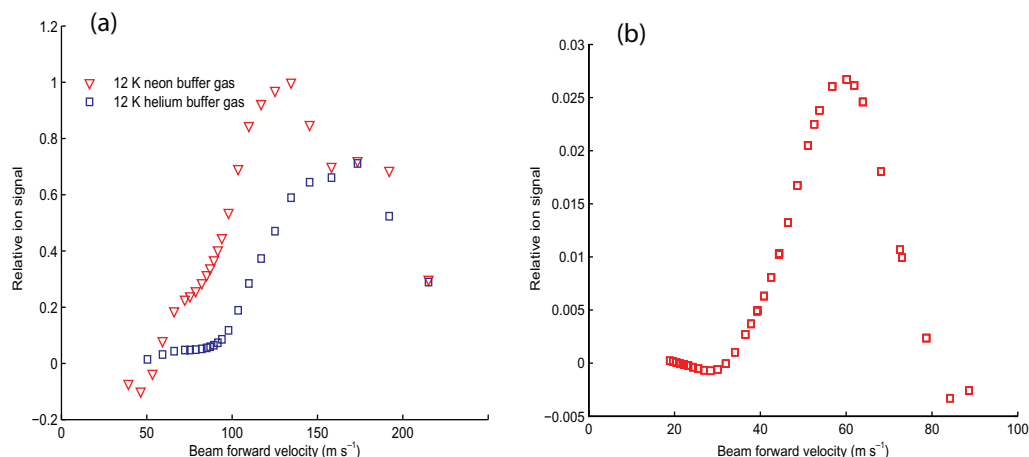


Figure 6. (a) Measured forward velocity distribution of a guided ND₃ beam using 12 K helium (blue square) and 12 K neon (red triangle) as a buffer gas. The beam using neon contains substantially more low-velocity molecules. Molecules moving faster than $\approx 150 \text{ m s}^{-1}$ are unlikely to be guided by this bent electrostatic guide (bend radius $r = 70 \text{ cm}$), although the exact fraction guided depends on the transverse energy of the molecules. The red data (neon) represents a guided beam of about $2 \times 10^{10} \text{ ND}_3 \text{ molecules s}^{-1}$ at a mean energy of $\approx 20 \text{ K}$. A background count rate has been subtracted. (b) Velocity distribution at the output of a 70 cm long electrostatic guide with a bend radius of 8 cm. The output of this guide is completely separated from the neon buffer gas, and represents a pure, state-selected, low energy beam. This data represents $3 \times 10^8 \text{ ND}_3 \text{ molecules s}^{-1}$, with a mean energy of about 4 K.

6.3. Next step II: general loading of traps

Low velocities and high fluxes make this source an attractive first step for loading electrostatic or magnetic traps. In order to load atoms or molecules into a conservative potential, energy must be removed from the particles while they are within the trap volume. Demonstrated methods to load traps from beams include using light forces [18, 19], switching electric [17] or magnetic [20] fields and buffer-gas loading [14]. Chandler *et al* [21] have proposed loading traps via collisions with another beam, and it should also be possible to optically pump slow moving atoms or molecules directly into trapped states [22]. Recent proposals [2] suggest molecules as well as atoms could be laser cooled from a source like ours, and all other methods listed here are trivially generalizable to molecules. All of these methods would benefit from starting with our bright, cold, slow moving source.

6.4. Next step III: nonlinear optics in buffer-gas cells

There has been recent interest [23, 24] in studying nonlinear optical effects in cold buffer-gas cells. There is theoretical [23] and experimental [25] evidence that decoherence times between ground-state magnetic sublevels in alkali atoms are very long. To date, only cells loaded via laser ablation, and in one case light-induced desorption [26], have been used for this work. Although substantial optical densities have been achieved, such experiments tend to be plagued

by anomalous losses at high buffer-gas pressures, limiting lifetimes to < 100 ms. Ablation is a violent and difficult to control process which is known to produce both clusters [27] and momentary dramatic heating [13] in buffer-gas cells. We believe that continuously loading cold vapor cells directly from an oven will not only provide higher density samples, but also do so controllably in a clean environment. Possible demonstrations include improved EIT [24], guiding light [28] and high-resolution magnetometry [29]. We have already achieved in the experiments described above continuous in-cell potassium optical densities much greater than 100.

7. Conclusion

We have demonstrated an extremely high-flux and moderately cold atomic beam of potassium using a neon buffer gas. We believe this source can be trivially generalized to any atomic and many molecular species that can be produced in a gas at temperatures up to 600 K and perhaps as high as 1000 K. The demonstrated cold flux of potassium represents a new benchmark for cold atom fluxes. In addition, an ND_3 from such a source has been coupled into an electrostatic beam guide. A broad range of possible applications with such guided molecules includes collision studies, trap loading and nonlinear optics studies.

Acknowledgments

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