A continuous source of translationally cold dipolar molecules

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The past years have seen an explosion of activity in the field of cold atomic gases [1]. It is interesting and desirable to extend these investigations to molecules, which have a complex internal structure and can as a consequence possess a permanent electric dipole moment. Trapping cold polar molecules will lead to new physics due to the long range and anisotropy of the dipole-dipole interaction. Slow molecules for precision measurements or interferometry are further motivations behind the ongoing efforts. However, the complexity and density of energy levels in the rotational and vibrational manifolds largely precludes the effective use of laser cooling techniques [2]. Therefore, a number of different approaches has been considered for cooling and trapping molecules. Buffer-gas cooling in a cryogenic environment is one possibility, but requires a rather complex setup [3]. Another method is photoassociation, but this is limited to simple molecules with laser-cooled precursor atoms [4]. A novel technique uses deceleration by the Stark effect, where packages of polar molecules are decelerated with time-varying electric fields [5, 6, 7]. Other, mostly mechanical methods have also been proposed but remain to be demonstrated [8, 9].

It is, however, not necessary to produce slow molecules, as they are present in any thermal gas, even at room temperature. Slow molecules only need to be filtered out. For this reason, already in the 1950s it was attempted to select the slowest atoms from a hot beam using gravity [11]. These attempts failed, mostly because the slow particles were kicked away by the fast ones. Much later, it was demonstrated that slow lithium atoms can be efficiently guided out of a hot beam with strong permanent magnets, providing a robust and cheap source of slow atoms [11], e.g. for Bose-Einstein condensation experiments. In the same spirit, an efficient and simple filtering technique could play an important role towards the production of a cold molecular gas.

In this Letter we describe an experiment in which the Stark interaction of polar molecules with an inhomogeneous, electrostatic field is exploited to efficiently select and guide slow molecules out of a room-temperature reservoir into ultrahigh vacuum. Whether a dipolar molecule is weak-field seeking and trapped by an electric field minimum, or strong-field seeking and expelled, depends on whether the average orientation of the rotating molecular dipole is antiparallel or parallel to the local electric field, respectively. Weak-field seeking molecules are trapped if their kinetic energy is less than the Stark-potential barrier for them. Thus a static quadrupolar potential forms a two-dimensional trap for molecules with small transverse velocity components with respect to the quadrupole axis. In a linear guide, transverse and longitudinal components of the velocity are completely decoupled. The longitudinal velocity of the molecules is limited by guiding them around a bend in the quadrupole, downstream from the reservoir. The centripetal force due to the electric field gradient guides only the slowest molecules around the bend. The fast molecules escape the guide and are pumped away.

Important parameters for characterizing such a continuous source of slow molecules are the velocity distribution and the flux of the molecules. For a given molecular state, we have a maximum transverse ($v_{l\text{max}}$) and longitudinal ($v_{t\text{max}}$) velocity that will be guided. Under the assumptions that the Stark shift of the molecule is linear with electric field, respectively. Weak-field seeking molecules are trapped if their kinetic energy is less than the Stark-potential barrier for them. Thus a static quadrupolar potential forms a two-dimensional trap for molecules with small transverse velocity components with respect to the quadrupole axis. In a linear guide, transverse and longitudinal components of the velocity are completely decoupled. The longitudinal velocity of the molecules is limited by guiding them around a bend in the quadrupole, downstream from the reservoir. The centripetal force due to the electric field gradient guides only the slowest molecules around the bend. The fast molecules escape the guide and are pumped away. For this reason, already in the 1950s it was attempted to select the slowest atoms from a hot beam using gravity [11]. These attempts failed, mostly because the slow particles were kicked away by the fast ones. Much later, it was demonstrated that slow lithium atoms can be efficiently guided out of a hot beam with strong permanent magnets, providing a robust and cheap source of slow atoms [11], e.g. for Bose-Einstein condensation experiments. In the same spirit, an efficient and simple filtering technique could play an important role towards the production of a cold molecular gas.

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hydro (H$_2$CO), which has a relatively large dipole moment of 2.34 Debye and is one of the best studied 4-atomic asymmetric-top molecules. At room temperature (300 K) almost all the H$_2$CO molecules will be in the vibrational ground state. A large number of rotational states consistent with the Boltzmann distribution at 300 K are populated. The experiment itself (Fig. 1(a)) consists of the bent quadrupolar guide starting at a 0.5 mm diameter ceramic nozzle of 11 mm length and ending in a UHV detection chamber, passing through a vacuum chamber where the filtering is performed. The guide has a length of 18 cm and is made of 1 mm diameter stainless steel rods, with a 1 mm gap between neighboring rods. The rods are built around the ceramic nozzle. Typical operation pressures in the nozzle, which injects the molecules directly into the quadrupole, are below 0.05 mbar in order to maintain molecular flow conditions. Most of the molecules are not guided and escape into the vacuum chamber, where a typical operational pressure of a few times $10^{-7}$ mbar is maintained by a 500 l/s turbo-molecular pump. The 90° bent section of the guide is about 60 mm downstream from the nozzle and its radius of curvature is 13.5 mm. After the bend, the guide passes through a 5 cm long and 4 mm narrow tube for differential pumping before entering the detection chamber, where a 561/s turbo pump maintains a pressure of $\approx 2 \times 10^{-9}$ mbar. The guide abruptly ends 25 mm before the ionization volume of a quadrupole mass spectrometer (QMS) [Pfeiffer vacuum, Prisma QMA 200]. Apart from measuring the direct flux, the QMS monitors the residual gas in the detection chamber. Standard pressure gauges are used to monitor the pressure in the UHV chamber.

To interpret experimental results, a Monte Carlo simulation was performed. The electric field in a 90°-bent quadrupole with short straight sections on either side was calculated numerically in 3 dimensions. The input distribution of the molecules injected into the guide in the simulation is random and consistent with the thermal speed distribution. Its angle distribution is that of a 3 mm deep field-free nozzle [2]. This is a good approximation to the experimental situation, where a 11 mm long nozzle in the presence of an electric field results in the transverse acceleration of molecules, giving a wall-to-wall collision-free path length of approximately 3 mm for the guidable molecules. For each molecular species, the Stark shifts of the relevant states were calculated by direct numerical diagonalization of the Stark Hamiltonian [3]. In the relevant electric field range (0-100 kV/cm), the majority of thermally populated rotational states of H$_2$CO and ND$_3$ show linear Stark shifts. These were gathered in a small number (20) of groups with similar Stark shifts. The trajectories under the influence of the electrostatic forces were calculated by a Runge-Kutta method. The simulation confirmed the expected quadratic dependence of the flux on the electric field, see Fig. 2(d). The simulation also yields the overall transmission of the guide, the velocity distribution inside the guide, the output divergence and velocity distribution of the beam. Finally, the simulation shows that the guide enriches the gas with states having a large Stark shift.

![FIG. 1: (a) Schematic of the experimental setup, and (b) a contour plot of the quadrupolar electric field in a plane perpendicular to the electrode rods.](image)

For detecting H$_2$CO molecules the quadrupole mass filter is set at mass 29, the strongest peak in the H$_2$CO mass spectrum. The influence of other gases at this mass is negligible. The channeltron-amplified QMS ion current can be tapped directly for transient measurements. Switching on and off the quadrupolar field resulted in a modulated QMS signal, see Fig. 2(a) and (b). It was checked that the electric disturbance from switching the

![FIG. 2: (a) The high voltage applied to the guide electrodes and (b) the molecular flux as a function of time. Note the delay and the relatively slow rise of the signal, corresponding to a gradual build-up of molecular flux, compared to the sudden fall due to the loss of molecules when the field is switched off. The line is a 20-point running average. (c) The flux as a function of applied electrode voltage. The symbols denote the measured height of the step function in (b). (d) The number of trajectories intersecting with the detector in a simulation where $10^6$ particles were injected into the guide, corresponding to a 1% fraction of the full input distribution. The lines in (c) and (d) are quadratic fits.](image)
high voltage did not significantly influence the signal apart from a short spike of sub micro-second duration. At high background pressures of polar molecules in the filter chamber and without injection of polar molecules into the guide, the ion signal decreases slightly when the HV is turned on. This effect is attributed to a decrease of conductance of the differential pumping section for polar molecules in the presence of electric fields. The effect is absent for non-polar molecules. As a further test that the signal represents the direct flux of the guide, a mechanical shutter was installed that can prevent the direct flux from reaching the QMS. It is designed so that it only prevents the guided molecules from entering the QMS. With the shutter blocking the direct flux into the QMS, the modulation of the QMS signal was a factor of \( \approx 5 \) smaller. With the above tests we conclude that the observed changes in the QMS signal are due to real changes in the density of the guided gas.

![Figure 3: The longitudinal velocity distribution](image)

FIG. 3: The longitudinal velocity distribution (data points with statistical error bars) derived from data obtained with an electrode voltage of \( \pm 5 \) kV. The smooth curve is a (normalized) fit to the data of the functional form \( (2\alpha v^3) \exp[-v^2/\alpha^2] \), with \( \alpha = 54 \) m/s, the stepped curve is the Monte-Carlo results for 5 kV. The negative values at high velocities are due to statistical noise in the data at short times.

As a first measurement, the guiding signal was recorded as a function of the applied voltage. The result, shown in Fig. 2(c), shows the expected quadratic dependence. To further characterize the source, the rising slope of the signal, coming from the time-of-flight, was analyzed in more detail. By using fast switches, the turn-on time of the high voltage is less than 1 \( \mu \)s, much less than all other relevant time scales in the experiment. With the high voltage off, the density of slow molecules that could be guided decays rapidly with distance from the nozzle. Therefore, it can be assumed that the molecules arriving at the detector must have entered the guide after the high voltage has been switched on. After a delay of a few ms, which depends on the applied voltage, the fastest guided molecules arrive at the QMS and the signal starts rising until it levels off after \( \approx 10 \) ms. From the delay and the rising slope, the longitudinal velocity distribution can be derived by differentiation. The analysis incorporates the following additional inputs: 1) the Monte Carlo simulation was used to estimate the velocity-dependent probability of hitting the detector after emerging from the guide. This probability was used to perform a velocity-dependent correction, which accounts for the acceleration of the molecules when leaving the guide and the fact that longitudinally slow molecules are more likely to miss the detector, because they spread out over a larger solid angle; 2) making the reasonable assumption that the ionizing probability of the QMS depends on the molecular velocity and does not saturate at the lowest measured velocities, a velocity-dependent ionization probability was calculated. The resulting velocity distribution obtained for an electrode voltage of \( \pm 5 \) kV is shown in Fig. 2. Subtracting the signal with the shutter closed had only a small effect on the velocity distribution, and was omitted.

Note that for a single molecular state the velocity distribution should show a relatively sharp velocity cut-off. But given a mixture of states with different Stark shifts, the cut-off is smeared out. Surprisingly, this leads to a velocity distribution which can reasonably well be described by a 1-dimensional thermal distribution. For example, the smooth solid curve in Fig. 3 represents a fit to the data with a temperature of 5.4 K. The experimentally determined velocity distribution is slightly narrower than the one from the Monte-Carlo simulation, and the mean velocity is shifted to a smaller value. This deviation is probably caused by experimental imperfections such as surface roughness of the electrodes, deviations from the design geometry, etc. The transverse velocity distribution of the guided molecules could not be measured directly, however, simulations indicate a distribution characterized by a temperature of 0.5 K inside the guide for \( \pm 5 \) kV electrode potential.

The absolute flux was measured by calibrating the ion current from the QMS with the background signal caused by non-guided hot H\(_2\)CO molecules. The partial pressure of the hot H\(_2\)CO was measured with the QMS and related to an absolute pressure via a Penning pressure gauge [Balzers, compact full range gauge PKR260], taking into account the relative ionization probabilities from Ref. [14]. An ionization gauge [Varian UHV-24] confirmed the Penning gauge readings to within 50%. The ionization volume is modelled by a (3 mm\(^3\))\(^3\) box, as indicated by the QMS supplier. Unfortunately, the exact dimensions of the ionization volume are hard to verify for our conditions. Therefore, we estimate a systematic error in the absolute flux of the order of a factor of 2. The flux that was calibrated in this way is plotted in Fig. 2 as a function of the absolute nozzle pressure. The measured H\(_2\)CO content in the nozzle is approximately 12%. In Fig. 3, the nozzle pressure was deliberately increased to a point where the molecular flow approximation is no longer valid. As can be seen in the figure, for low nozzle
pressures the flux increases linearly, but starts deviating from the linear dependence above 0.05 mbar, reaches a maximum of nearly $10^9$ s$^{-1}$, with peak density inside the guide of $\approx 10^8$ cm$^{-3}$, around 0.15 mbar before actually reducing with pressure. This behavior is caused by collisions. When increasing the pressure in the nozzle, the beam characteristics change from an effusive, thermal velocity distribution to a distribution which is peaked to a higher velocity. Moreover, slow molecules are kicked away by the fast ones in the guide and by background molecules. Therefore, it is important to keep the pressure in the nozzle sufficiently low.

FIG. 4: The absolute measured flux through our guide as a function of nozzle pressure for an electrode voltage of $\pm 5$ kV. The symbols are data points, the curve a guide to the eye. For low pressures the gas in the nozzle is in the molecular flow regime and the flux is linear in the nozzle pressure. Above $\approx 0.02$ mbar the mean free path for thermal molecules becomes less than the nozzle length and collisions start playing a role. As a consequence, the increase of the flux slows down and eventually the absolute guided flux is reduced.

From the Monte-Carlo simulation, the measured H$_2$CO concentration and the theoretical flow impedance of the nozzle, the expected flux can be determined at a nozzle pressure of 0.025 mbar and an electrode voltage of $\pm 5$ kV. The result is $3.5 \times 10^9$ s$^{-1}$, a factor of 10 larger than observed in the experiment. The discrepancy might be due to background collisions and, more likely, a non-perfect nozzle and a non-perfect alignment of the detector and the electrodes.

To demonstrate the general character of the source, also a slow beam of deuterated ammonia (ND$_3$) was produced, with similar results to those obtained for H$_2$CO. The deuterated species was used because of a null QMS background signal from contaminants at the mass peaks of interest and because its Stark shift is larger than that of NH$_3$.

In conclusion, we have experimentally demonstrated a continuous, high-flux source of translationally cold molecules at temperatures of a few kelvin. The molecules are conveniently delivered “on spot”, e.g. into an ultrahigh vacuum, where they can be stored in an electrostatic trap or an electrostatic ring. This simple and versatile method is generally applicable to all molecules with a reasonably large dipole moment. By reducing the radius of curvature of the bend, slower output beams can easily be achieved. In principle, the source can also be used as a novel kind of gas chromatograph that selects polar molecules from a non-polar buffer gas, or for enrichment of a gas with strong low-field-seeking states.

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