Cold Molecules

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During the last years there has been a rapidly growing interest in the field of cold molecules.[1–3] This has obviously been inspired by the spectacular successes in the closely related field of cold atoms, which have recently been recognized by the award of the 2001 Nobel Prize in Physics to Cornell, Ketterle, and Wieman. But molecules have much more to offer than simply extending the experiments already performed with atoms to more complex species.

KEYWORDS:

Bose – Einstein condensates · clusters · cooling · low temperature physics · polar molecules

There are several fundamental physics studies that are currently being pursued, for which molecules are used as test-beds. In the search for the electric dipole moment of the electron, for instance, specific molecules like YbF and PbO are considered ideal candidates;[4, 5] the valence electron in these molecules is exposed to (internal) electric fields that are orders of magnitude larger than those obtainable otherwise. The measurement of the temporal variation of the fine-structure constant[6] is another ambitious goal, which might be achieved by accurately measuring the ratio of the transition frequencies of various types of optical transitions in molecules. To study the effect of the weak interaction, chiral molecules are used, as the weak interaction is expected to lead to differences in molecular transition frequencies for different enantiomers, namely for molecules that are each others' mirror-image.[7] For all these types of measurements it would be a major advantage if they could be performed on cold molecules, molecules that are almost motionless. This would lengthen the interaction time and thereby greatly increase the precision in the various experiments.

In a sample of cold molecules, the relative velocities of the molecules are, by definition, very small but the absolute velocity of the ensemble of molecules can have any arbitrary value. Beams of "velocity compressed" molecules with a tunable absolute velocity, that is, with a tunable de Broglie wavelength, offer fascinating perspectives for molecular optics[8] as well as for molecular collision studies and for reactive scattering experiments. In molecular collisions, huge scattering resonances have been predicted at low collision energies.[9] These resonances can occur, for instance, when colliding molecules begin to rotate, leaving them with insufficient translational kinetic energy to overcome their van der Waals attraction. These resonances can be very narrow, and measuring the width and position of these resonances as a function of collision energy would provide a new experimental characterization of the potential energy surface of the interacting molecules very close to the dissociation limit. Similarly, the formation of long-lived, transiently bound molecular complexes might enable reactions to occur via tunneling through reaction barriers, thus opening up novel routes for low-temperature chemistry.

Production of cold molecular gases in the quantum degenerate regime obviously remains an important goal as well. When a molecular Bose – Einstein condensate will be formed, there is the possibility to study its behavior for molecules in selected vibrational and (end-over-end) rotational states. One should be well aware of the complexity that might arise in these systems, in particular when different nuclei with nonzero nuclear spin are present in the molecule. Arguably the most interesting aspect of molecules compared to atoms is that many molecules do have a permanent electric dipole moment. The anisotropic interaction between the dipoles might on the one hand complicate matters but, on the other, it will give rise to an extremely rich and new physics in the cold dipolar gases.[10] It has been pointed out that the stability diagram of a trapped gas of bosonic molecules interacting dominantly through dipole – dipole forces is strongly influenced by the trapping geometry.[11] This offers new possibilities for controlling and engineering macroscopic quantum states. It has also been predicted that cold dipolar gases of fermions are excellent candidates for achieving the superfluid transition.[12] Another challenging prospect is that trapped polar molecules might be used for quantum computation; in this design, the qubits would be the electric dipole moments of ultracold diatomic molecules, oriented parallel or antiparallel to an external electric field, with coupling between bits through the electric dipole – dipole interaction.[13]

Production of Cold Molecules

For the production of samples of cold molecules, the method of laser cooling, that has been so crucial for the success in the field

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of cold atoms, is not particularly well suited. Laser cooling relies on the possibility of repeatedly inducing an absorption–emission cycle in an atom, thus leading to an efficient transfer of momentum from the photons in the laser beam to the atom. With laser beams in the proper configuration, atoms can be slowed and cooled. The more complex energy-level structures of molecules compared to atoms makes it nearly impossible to find a (sufficiently) closed two-level system in molecules for efficient laser cooling. This complication is not due to the rotation of the molecule; transitions between different rotational states are governed by strict selection rules, and for a variety of molecules optical transitions can be found that are electronically and rotationally closed. The problem is in the vibrations of the molecule; transitions between different vibrational states are governed by Franck–Condon factors, that is, the overlap of the corresponding vibrational wavefunctions. After spontaneous emission the molecules therefore are generally distributed over a large number of vibrational states. Only in favorable cases, namely when the sum of all off-resonant Franck–Condon factors is sufficiently small, could laser cooling be applied to molecules. Although not yet demonstrated, it appears that the NH radical might be a suitable molecule for laser cooling.[14]

There are currently three different methods that have produced cold molecules that have subsequently been trapped. The first, and up to now most widely applied, method starts from samples of cold atoms as, for instance, obtained in a "standard" magneto-optical trap (MOT). Through the process of photoassociation, two atoms are bound together.[15] The thus-produced molecules are translationally as cold as the atoms from which they are produced. The atoms are bound together at a rather large internuclear distance, favoring the formation of molecules in highly vibrationally excited states just below the dissociation limit. The molecules are produced in low angular momentum states and often in electronically excited, metastable triplet states. Using a variety of laser schemes, the internal state distribution can be manipulated, and the molecules can be transferred into their rotational–vibrational and electronic ground state. Photoassociation has up to now mainly been used to produce homonuclear alkali dimers, although heteronuclear diatomics can be produced as well. Trapping of the cold molecules can be performed in a far off-resonance quasielectrostatic trap, like in the focus of a CO_2 laser beam.[16] More recently, approximately 2×10^5 Cs_2 molecules at 50 µK, produced via photoassociation, have been trapped in the inhomogeneous magnetic field that was used to produce the Cs MOT.[17]

In the second method, molecules are injected in a cold He buffer gas and allowed to equilibrate with the buffer gas after multiple collisions. When ^3He is used as buffer gas, the temperature can be as low as 250 mK while still maintaining a sufficient buffer gas density for efficient cooling. At these temperatures, molecules will be in their electronic and vibrational ground states

and only a very limited number of rotational states will be populated. Molecules in so-called low-field seeking states can be trapped in a magnetic field minimum near the center of the buffer gas cell, which is created by two magnet coils in anti-Helmholtz configuration. This cooling method is generally applicable, and relatively large numbers of paramagnetic molecules can be trapped this way. In the initial experiments, approximately 10^8 CaH molecules, produced via laser ablation of a solid precursor placed inside the buffer gas cell, were magnetically trapped at 400 mK.[18]

In the third method, developed in our laboratory and detailed here, a pulsed beam of dipolar molecules is decelerated by letting it pass through an array of time-varying inhomogeneous electric fields.[19] The bunches of slow molecules that exit this so-called Stark decelerator are subsequently stored in an electrostatic storage ring[20] or trapped in an electrostatic quadrupole trap.[21] In particular, we succeeded in electrostatic trapping of ground state ammonia molecules residing in a single rotational–vibrational level at densities of (better than) 10^7 molecules per cubic centimeter and at temperatures of around 25 mK. In Figure 1, a schematic overview of the experimental set-up used for these experiments is shown, together with a photograph of the 35 cm long Stark decelerator with the attached quadrupole trap.
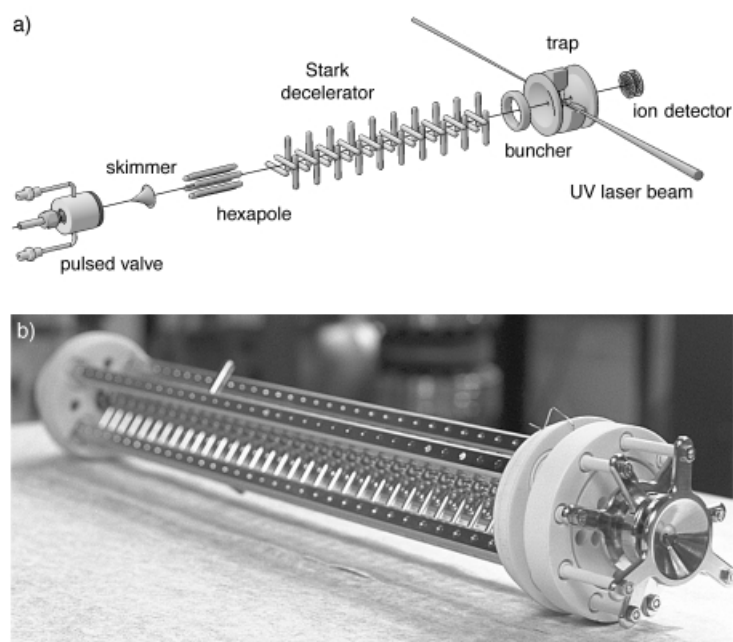

Figure 1. a) Schematic view of the experimental set-up used for trapping neutral ammonia molecules. A pulsed beam of ammonia molecules with a mean velocity of 275 m s^{-1} passes through a skimmer. Ammonia molecules in a selected quantum state are focused by an electrostatic hexapole lens on the entrance of the Stark decelerator. In the decelerator, a fraction of the ammonia molecules is decelerated and exits the decelerator as a compact package of about 1–2 mm length, moving with an absolute velocity of around 15 m s^{-1} and with a velocity spread of several m s^{-1}. The slow beam of ammonia molecules is focused and bunched inside an electrostatic quadrupole trap, which is abruptly switched on once the molecules have come to a near complete standstill at the center of this trap. In order to detect the ammonia molecules, a pulsed laser is focused inside the trap. The resulting ions are extracted and counted using an ion detector. b) Photograph of the 35 cm long Stark decelerator with the attached quadrupole trap.

Molecular Beam Deceleration and Trapping

The Molecular Beam

In our experimental method, full advantage is taken of the process of adiabatic cooling in a pulsed gas expansion. When a dilute mixture of the molecule of interest with a rare gas is expanded from a high-pressure region into vacuum, efficient cooling of all internal degrees of freedom of the molecule occurs through multiple collisions in the expansion region. Pulsed molecular beams produced this way have rotational temperatures of a few degrees Kelvin and can have densities of 10^{12} molecules per cubic centimeter per quantum state. Such beams have been used extensively to greatly simplify molecular spectroscopy and to produce weakly bonded molecular complexes.[22] The relative velocity spread in the beam is typically on the order of 10%, corresponding to a translational temperature in the moving frame of the molecular beam of approximately 1 K. To be applicable for molecular trapping, one needs to transfer the high phase-space densities present in the moving frame of the molecular beam to the laboratory frame. The typical absolute velocities in a molecular beam are in the 250–2000 m s^{-1} range, mainly determined by the mass and initial temperature of the carrier gas. As the absolute velocity in the molecular beam is the velocity with respect to the source, one possibility to produce slow beams in the laboratory frame is to move the source backwards at the appropriate velocity. Although this method has been demonstrated to work in principle,[23] collisions in the source region of the molecular beam severely limit its practical use. In our experiments, we use a "standard" pulsed expansion of a mixture of 1% of the molecules of interest (CO, ND_3) in Xe, and we cool the pulsed valve as far as possible without liquifying the gases. In this way, a pulsed molecular beam with an absolute velocity of around 275 m s^{-1} and with a width corresponding to a translational temperature of 0.5 K is produced. This is about as slow and as cold a molecular beam as can be made with conventional means and serves as a good starting point for deceleration and trapping experiments.

The Stark Decelerator

To focus and decelerate this molecular beam we exploit the interaction of dipolar molecules with inhomogeneous electric fields. If a molecule is in a quantum state that experiences an increase in Stark energy with increasing electric field, this molecule will be decelerated while moving from a zero into a high electric field. When moving out of the high electric field again, the molecule will be accelerated, and regain its original kinetic energy in zero electric field. This has often been used in molecular beam experiments to spatially manipulate molecules in these so-called "low-field seeking" states, in particular to control the off-axis velocity component of the molecules. In an electrostatic hexapole, for instance, the electric field close to the axis is cylindrically symmetric and quadratically increases with the distance from the symmetry axis. Molecules in quantum states that experience a linear positive Stark shift thus feel a force directed towards the symmetry axis, increasing linearly with the distance from the axis. For a beam of molecules directed along the symmetry axis of the hexapole, the hexapole thus acts as a positive lens. Although the spatial manipulation of molecules with inhomogeneous electric fields has been used before, it had not been equally well appreciated that time-varying inhomogeneous electric fields can be used to manipulate the molecules in velocity as well. However, it is evident that when the electric field is abruptly switched off once the molecules have moved into it, the molecules will not regain their original kinetic energy; part of the kinetic energy of the molecules is then transferred to the power supply that creates the time-varying electric field.

The amount by which the kinetic energy of a molecule can be reduced by letting it move into a single electric field section, that is rapidly switched off once the molecule is in the maximum field, is rather small however. It depends strongly on the molecule and on the quantum state and is determined by the maximum positive Stark shift that the molecule experiences in experimentally realizable electric fields. For various small molecules, positive Stark shifts of typically 1 cm^{-1} can be obtained in electric fields up to 100 kV cm^{-1}. This value should be compared to the kinetic energy of the molecules in our molecular beam, which is on the order of 100 cm^{-1}. Therefore, to decelerate the molecular beam to low velocity, this process has to be repeated and an array of electric field sections is required. In the prototype Stark decelerator that we constructed for this, and which is schematically shown in Figure 2, we have used an
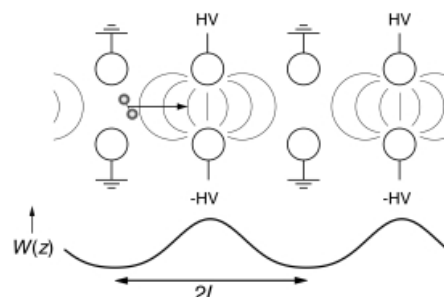

Figure 2. *Scheme of the Stark decelerator, together with the Stark energy W of a molecule as a function of position z along the molecular beam axis. In our prototype set-up there are 64 electric field stages separated by a distance L = 5.5 mm. Each stage consists of two parallel cylindrical metal rods of 3 mm diameter centered 5 mm apart. One of the rods is connected to a positive and the other to a negative switchable high-voltage (HV) power supply. Typically, the decelerator is operated at voltages of ± 10 kV, leading to a maximum electric field on the molecular beam axis of 90 kV cm^{-1}. The maximum Stark shift for ND_3 molecules in the particular low-field seeking quantum state that is selected in these experiments is then 1.1 cm^{-1}. Close to the electrodes, the electric field is 120 kV cm^{-1}, leading to a focusing of the molecules in low-field seeking states towards the molecular beam axis.*

array of 64 equidistant electric field stages, alternately positioned horizontally and vertically. The electric fields in the various sections are switched on and off synchronously with the pulsed beam of molecules that passes through it. While passing through the Stark decelerator, the molecules in low-field seeking states stay confined near the symmetry axis of the decelerator, as there the electric field is at a minimum.

Also in the forward direction, namely along the molecular beam axis, a bunch of molecules can be kept together throughout the decelerator when the electric fields are switched at the appropriate times. To see this, consider a molecule moving with exactly the average velocity along the molecular beam axis and entering a region of high electric field, as indicated in Figure 2. If we switch off the field when this particular molecule is somewhere "uphill" but not yet in the maximum field, this molecule will be decelerated, though less efficiently than could be. Molecules that are faster, those molecules that are ahead, will be decelerated more, while slower molecules, those that are lagging behind, will be decelerated less or even be accelerated. This process can be described as the trapping of molecules in a potential well that travels with a velocity determined by the timing sequence of the array of electric fields,[19] and is fully equivalent to the concept of phase stability in charged particle linear accelerators (LINACs).

The process in our Stark decelerator, the LINAC for neutral molecules, can be viewed as slicing a bunch of molecules with a narrow velocity distribution, corresponding to a translational temperature of a few millikelvin, out of the original beam and decelerating these to arbitrarily low absolute velocities; acceleration of the beam is of course possible as well. In this process not only the phase-space density (defined as the number of molecules per unit volume per unit momentum space) but also the shape in phase space remains the same, and one can thus efficiently transfer the high phase-space densities from the moving frame of the molecular beam to the laboratory frame.[19]

Electrostatic Trapping of Molecules

Once molecules are decelerated to sufficiently low velocities, they can be trapped in a variety of ways. By bending a hexapole in the shape of a torus, a storage ring for neutral dipolar molecules can be formed. A molecular beam can be tangentially injected in this storage ring, passing between the rods of the hexapole, which is abruptly switched on once the molecules have entered it. To

confine molecules to stable orbits in this storage ring, the inhomogeneous electric field inside the ring has to be sufficiently large to provide the required centripetal force. The diameter of the storage ring needed thus scales with the square of the forward velocity of the molecules, and having a slow molecular beam for injection in the ring is a good asset for testing the performance of such a device. In Figure 3 a scheme of the experimental set-up used for demonstrating a prototype molecular storage ring is shown. The measurements shown in the same Figure demonstrate that bunches of about 10^6 state-selected ND_3 molecules with a forward velocity of 89 m s^{-1} make
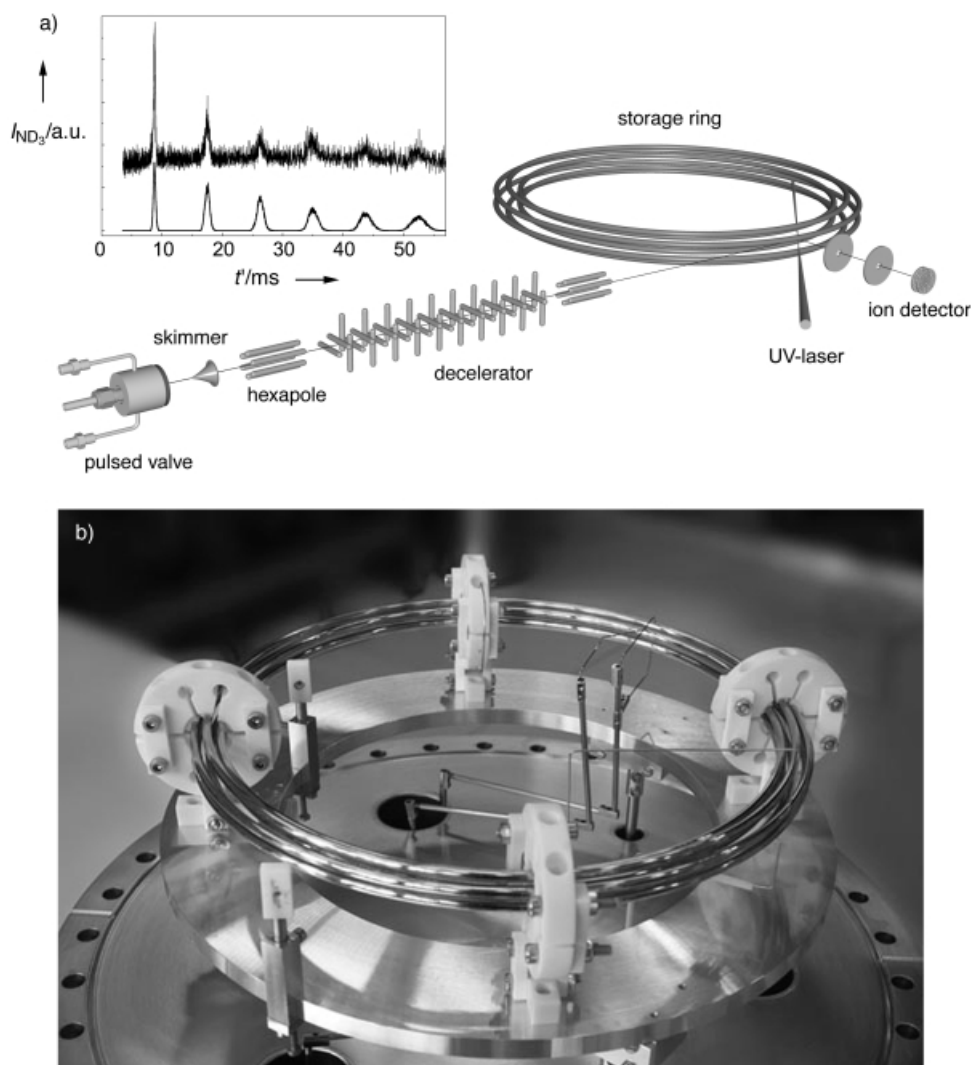

Figure 3. a) Schematic view of the experimental set-up used for demonstrating the prototype storage ring for neutral molecules. Bunches of ammonia molecules, that exit the Stark decelerator with a forward velocity of 89 m s^{-1} and with a velocity spread corresponding to a translational temperature of 10 mK, are tangentially injected into the electrostatic storage ring and pass through the 2 mm wide "slit" between the 4 mm diameter hexapole rods. The density of ammonia molecules at the point at which they are coupled into the 25 cm diameter storage ring is measured using a UV-laser based ionization detection scheme. The measurements demonstrate that bunches containing approximately a million ammonia molecules can be detected for up to six round trips in the storage ring (t' = 0 ms corresponds to the time at which the molecules are injected into the storage ring). The detected package of molecules is seen to gradually broaden in time and to decrease in peak intensity I_{ND_3}, mainly due to the tangential velocity spread (dispersion), as confirmed by the result of trajectory calculations shown underneath the measurements. b) Photograph of the 25 cm diameter hexapole torus, the prototype storage ring for neutral polar molecules.

up to six round trips in the 80 cm circumference storage ring at translational temperatures of approximately 10 mK.[20]

A sectional version of the storage ring will allow field-free interaction regions and various out-coupling regions. In such a ring, bunches of cold molecules can be made to interact repeatedly, at well defined times and at distinct locations, with electromagnetic fields and/or particles. This yields unique opportunities for high-resolution molecular spectroscopy as well as for a large variety of collision studies. By incorporating a rebunching element in the ring, the gradual spreading of the package that is observed can be avoided and the stored molecules can be confined to the same region in phase space for a time-period limited only by collisions with the background gas. Different molecules can be stored simultaneously; slowly over-taking or counterpropagating bunches can be stored and used for collision studies. The excellent optical access to the storage ring makes laser detection schemes feasible to accurately determine where the molecules are in the ring at each given moment in time. If this is done in a nonintrusive manner, this information can be used to actively correct the trajectories of individual molecules towards the ideal, circular orbit, thus increasing the phase-space density of the stored molecules (stochastic cooling).

In the storage ring, the molecules still have a forward velocity of around 100 m s^{-1}. If the molecules are decelerated to lower velocities, they can be trapped in a quadrupole electrostatic trap. Typical trap depths that can be achieved are on the order of (a fraction of) 1 K, implying that molecular velocities below approximately 20 m s^{-1} are required for trapping. Moreover, if a molecular beam with a low absolute velocity and with a certain velocity spread is injected in the trap, one would like to first get rid off (most of the) forward velocity and the final temperature of the trapped species be solely determined by the width of the velocity distribution. This can be done by applying voltages to the trap electrodes asymmetrically, such that the molecules have to climb one last potential hill on their way into the trap. By properly choosing the velocity of the molecules entering the trap, molecules can be made to come to a standstill near the center of the trap. If we would not change anything, the molecules would subsequently be reflected back out of the trap again. At this time, however, the voltages on the trap electrodes are switched to a symmetrical configuration, creating a potential well from which the molecules cannot escape.

In Figures 4a and 4b the electrode configuration used to load and trap a slow beam of state-selected ND_3 molecules is shown.[21] Upon exiting the decelerator, the molecules pass through a ring electrode that is used to focus and bunch, namely to focus in the forward direction, the package of molecules at the center of the electrostatic quadrupole trap. The density of the ND_3 molecules at the center of the trap is measured using resonant laser ionization followed by mass-selective detection of the ions. The oscillations in the ND_3 density, as observed soon after switching on the trapping field, reflect the "slushing" of the package of molecules inside the (anharmonic) trap. The absolute number density of the trapped ND_3 molecules is better than 10^7 molecules per cubic centimeter while their temperature is measured to be around 25 mK. The package of trapped
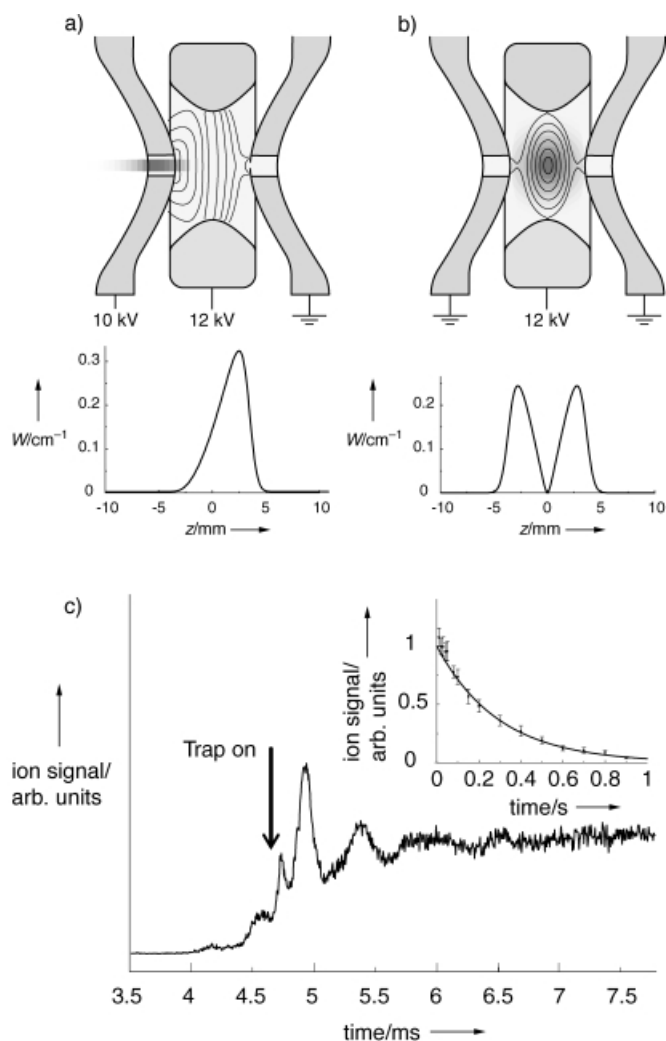

Figure 4. Electric field configuration used for a) loading and b) trapping neutral polar molecules. In both cases, a cut through the trap is shown and the applied voltages indicated. The solid curves inside the trap are lines of equal electric field. The potential energy along the molecular beam axis for ammonia molecules in the selected low-field seeking state is shown for these two cases as well. c) The density of ammonia molecules recorded at the center of the trap is shown as a function of time. The time at which the voltages are switched from the loading to the trapping geometry is indicated with an arrow. After the oscillations in the ammonia density at the center of the trap have faded away, a steady signal of trapped ammonia molecules, corresponding to a density of at least 10^7 molecules per cubic centimeter, remains. Inset: The density of ammonia molecules on a longer timescale and an exponential fit to the data with an exponential decay time of 0.3 s.

molecules has an oblate shape, about 1 mm diameter and 0.5 mm thick. The trapping time is presently limited by collisions with background gas to a fraction of a second, and is expected to lengthen considerably in a better vacuum. The deceleration and trapping scheme that we have used relies on the magnitude of the Stark shift in a given quantum state relative to the kinetic energy of the molecules. It is therefore possible to start with a beam containing different molecules and to simultaneously trap these; we have recently demonstrated simultaneous trapping of bosonic $^{14}ND_3$ and fermionic $^{15}ND_3$ at the above-mentioned densities and temperatures.[24]

Future Prospects

Given the densities and temperatures that can be achieved in a state-of-the-art pulsed molecular beam, we are confident that it will be possible to get a total number of 10^7 molecules at a density of 10^{10} molecules cm^{-3} and at a temperature below 10 mK in the trap; a straightforward scaling up of the present molecular beam machine, Stark decelerator, and trap will suffice for this. Further improvement might be obtained from optimizing the timing sequence of the various electric fields, through using feedback control and genetic learning algorithms. It should be realized that these number densities are expected to be obtainable in a single loading cycle. Accumulation of molecules from successive deceleration cycles in the trap would appear to be the ideal method for further increasing the number density. However, increasing the phase-space density is not possible when only conservative forces are applied (Liouville's theorem); simply opening the trap to allow molecules from the next deceleration cycle to enter cannot be performed without losing or heating the molecules that are already stored. For the NH radical there is an experimental scheme to circumvent this fundamental obstacle. The essence of this scheme is that the molecules are decelerated and brought to a near standstill by the Stark decelerator while residing in a long-lived electronically excited state. By applying an intense laser field, the spontaneous decay of these radicals to their electronic ground state can then be strongly enhanced. In their electronic ground state the NH radicals can be trapped in inhomogeneous magnetic fields. The laser-induced spontaneous decay thus provides a unidirectional path to load the magnetic trap, in which the molecules can then be accumulated.[14] This might be applicable to other molecules as well using IR transitions. For most other molecules, increasing the phase-space density will only be possible when there is dissipation. The feasibility of evaporative cooling and of laser cooling on rotational–vibrational transitions needs to be investigated for various molecules. Another possible route might be in sympathetically cooling molecules in a rubidium magnetic trap, especially given the recent success in producing a potassium Bose–Einstein condensates via sympathetic cooling of potassium with rubidium.[25] Whenever any kind of cooling of the trapped molecules works, reloading of the trap can be used to further increase the number of trapped molecules.

It would be a major advantage if molecules in "high-field" seeking states could be decelerated and trapped as well. The rotational ground state of any molecule is always lowered in energy by an external perturbation and is therefore high-field seeking. Evaporative cooling is much more likely to be successful on molecules in their lowest energy quantum state, and many new molecules, in particular those considered useful for the electric dipole moment measurements, would become available for these studies. It might appear to be straightforward to apply the above methods to molecules in high-field seeking states by simply reversing the situation and letting the molecules fly out of, instead of into, the region of high electric field for instance. However, Maxwell's equations do not allow for a maximum of the electric field in free space, and therefore transverse stability cannot be maintained easily; molecules in high-field seeking

states have the tendency to crash into the electrodes, where the electric fields are the highest. The same situation is encountered in charged particle accelerators where this problem has been resolved by using alternate gradient (AG) focusing. This method came out of the realization that a long-established fact of geometrical optics was applicable to ion optics as well: For a pair of lenses that have equal focal lengths but with one lens converging and the other diverging, the total focal length is always positive. This same principle can be applied to polar molecules when using electrostatic dipole lenses. These lenses focus the molecular beam in one direction but simultaneously defocus the beam in the orthogonal direction. By alternating the orientation of these lenses, an electric field geometry with a focusing effect in both dimensions can be created. By switching these lenses on and off at the appropriate times, AG focusing and deceleration of a molecular beam can be achieved simultaneously, as we have recently successfully demonstrated.[26] In particular cases, an alternative method to produce a beam of slow molecules in high-field seeking states might be to first decelerate and focus the molecules while in a low-field seeking state and to then induce a transition (microwave, radiofrequency) to a high-field seeking state. The latter method also allows for the preparation of the molecules in selected, single hyperfine levels. In either way, once the molecules are sufficiently slow they can be trapped in an electrodynamic trap or in a ring geometry, for instance around a wire or in a (AG) storage ring.

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