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## Preparation of an ultra-cold sample of ammonia molecules for precision measurements

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## ABSTRACT

We present experiments in which an ultra-cold sample of ammonia molecules is released from an electrostatic trap and recaptured after a variable time. It is shown that, by performing adiabatic cooling before releasing the molecules and adiabatic re-compression after they are recaptured, we are able to observe molecules even after more than 10 ms of free expansion. A coherent measurement performed during this time will have a statistical uncertainty that decreases approximately as the inverse of the square root of the expansion time. This offers interesting prospects for high-resolution spectroscopy and precision tests of fundamental physics theories.

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After eliminating more mundane broadening effects, the resolution of a spectroscopic measurement is ultimately limited by the time an atom or molecule interacts with the radiation field. In typical molecular beam experiments, this interaction time is limited to around 1 ms, resulting in a measured linewidth on the order of a kilohertz. The development of cooling techniques for atoms has led to a dramatic increase in the interaction times and hence the attained accuracy. Currently, atom and ion clocks reach accuracies on the order of  $10^{-18}$ , allowing for tests of fundamental physics theories such as relativity [1], quantum electrodynamics [2] and the time-invariance of the fine-structure constant [3] at an unprecedented level. It is anticipated that cooling techniques for molecules will lead to a similar increase in accuracy, allowing for more precise tests of time-reversal symmetry [4,5] and the time-invariance of the proton-to-electron mass ratio [6], observation of parity violation [7] and weak interactions in chiral molecules [8]. Unfortunately, the cooling techniques for molecules demonstrated so far, such as Stark and Zeeman deceleration, buffer gas cooling, photo-association and laser cooling, have a poor efficiency (for recent review papers, see Refs. [9–11]). Therefore, although in proof-of-principle experiments prolonged interaction times have been achieved [12,13], this was accomplished at the expense of a large decrease in signal. Hence, the Allan deviation – a measure of the statistical uncertainty of the experiment – was increased

rather than decreased by using slow molecules. But the statistical uncertainty is not all that matters, usually the accuracy of experiments is limited by systematic effects rather than by the statistical uncertainty. As most systematic effects are proportional to the measured line-width [14], an experiment with a large Allan deviation but a small line-width is in some cases preferable over an experiment with a smaller Allan deviation but a large line-width. If the Allan deviation is too large, however, the required measurement times become unrealistically long.

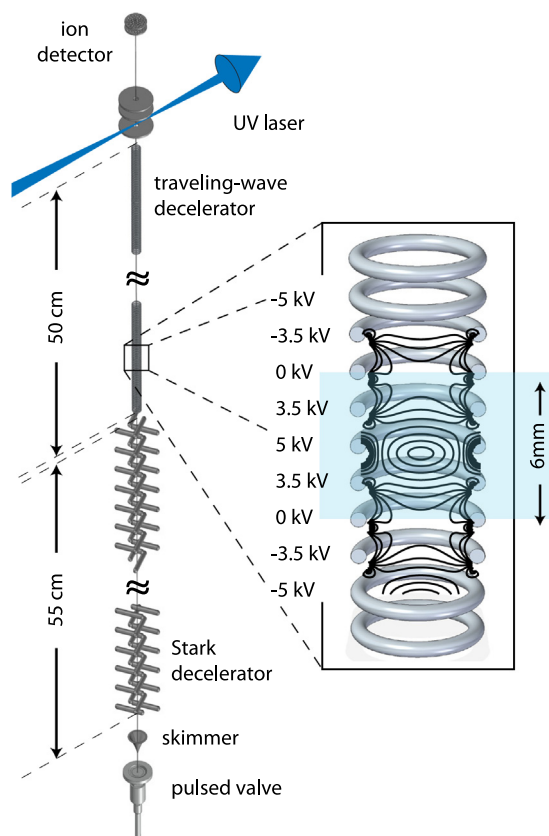
Recently, our group has decelerated and trapped ammonia molecules using a series of rings to which oscillating voltages are applied [15,16]. The advantage of such a traveling-wave decelerator [17–19] is that molecules are confined in a genuine 3D trap throughout the deceleration process, which avoids the losses at low velocity that plague conventional Stark decelerators. The necessary voltages for the decelerator are generated by amplifying the output of an arbitrary wave generator using fast high voltage (HV) amplifiers, giving us great control over the trapping potential. In this paper, we present measurements in which we adiabatically cool  $\text{ND}_3$  molecules to sub-mK temperatures, release them by switching off the trap voltages completely, recapture them after a variable expansion time, re-compress and finally detect them. We discuss how this method could be used for obtaining high-resolution spectra.

## 1. Experimental setup

Fig. 1 shows a schematic view of our vertical molecular beam machine. A beam of ammonia molecules seeded in xenon is

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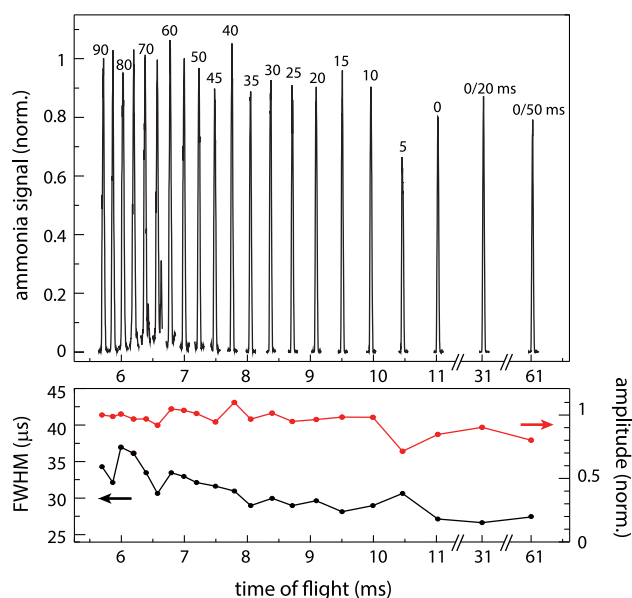
**Fig. 1.** Schematic view of the vertical molecular beam machine. A beam of molecules is decelerated and trapped using a combination of a Stark decelerator and a traveling-wave decelerator. The inset shows the electric field magnitude inside the traveling-wave decelerator (in steps of 2.5 kV/cm) with the voltages as indicated.

released into vacuum using a pulsed valve. This beam is decelerated from around 330 m/s to 90 m/s using a Stark decelerator consisting of 101 pairs of electrodes to which voltages of +10 and -10 kV are applied. The voltages are abruptly (<100 ns) switched using four independent HV switches (Behlke HTS 151-03-GSM) that are triggered by a programmable delay generator. The traveling-wave decelerator, which is mounted 24 mm above the last electrode pair of the first decelerator, consists of a series of 336 rings, each of which is attached to one of eight stainless steel rods to which voltages of up to  $\pm 5$  kV are applied. At any moment in time, the voltages applied to successive ring electrodes follow a sinusoidal pattern in  $z$ , where  $z$  is the position along the beam axis. These voltages create minima of electric field every 6 mm, which act as 3D traps for weak-field seeking molecules. By modulating the voltages sinusoidally in time, the traps are moved along the decelerator. A modulation frequency that is constant in time results in a trap that moves with a constant positive velocity along the molecular beam axis. A constant acceleration or deceleration can be achieved by applying a linear chirp to the frequency [18,19]. The necessary voltages are generated by amplifying the output of an arbitrary wave generator (Wuntronic DA8150) using eight fast HV amplifiers (Trek 5/80). The  $\text{ND}_3$  molecules are state-selectively ionized 20 mm above the traveling-wave decelerator using a focused UV laser beam and counted by an ion detector. More details of the setup are given in Quintero-Pérez et al. [15] and Jansen et al. [16]. Compared to the setup used in our previous experiments, we have made a number of changes: (i) the general valve has been replaced by a Jordan valve; (ii) the distance between the decelerator and the detection region has been

decreased from 40 mm to 20 mm; (iii) the suspension of the traveling-wave decelerator has been altered such that the detection region is better pumped; and (iv) the alignment of the ring electrodes with respect to each other as well as the alignment of the complete traveling-wave decelerator with respect to the beam line has been improved.

## 2. Experimental results

The upper panel of Fig. 2 shows the density of  $\text{ND}_3$  molecules above the decelerator as a function of time when different waveforms are applied to the ring decelerator. In all cases, a packet of molecules is decelerated to 90 m/s using the conventional Stark decelerator and injected into the traveling-wave decelerator at  $t = 0$ . The time-of-flight (TOF) trace labeled as '90' is recorded when a sinusoidal voltage with an amplitude of 5 kV and a constant frequency of 7.5 kHz is applied to the traveling-wave decelerator. In this case, molecules at a velocity of 90 m/s are guided through the decelerator and arrive at the detection zone after 5.5 ms. The TOF traces that are labeled as '85'-'0' are recorded when the frequency is first linearly decreased to a certain value and subsequently increased to its original value. With these waveforms, molecules are decelerated to the indicated value (in m/s) before being accelerated back to their original velocity. Consequently, the molecules will spend a longer time in the decelerator and will arrive at the detection zone at later times. The TOF trace that is labeled as '0' corresponds to the situation in which molecules come to a complete standstill before being accelerated back to 90 m/s and detected. As expected, the time the molecules spend in the decelerator is now approximately 2 times longer than when the molecules are guided. In these experiments the acceleration is changed from 0  $\text{m/s}^2$ , for guiding, to 17,000  $\text{m/s}^2$ , for deceleration to 0 m/s. Once the molecules come to a standstill, we can hold them for arbitrarily long times by keeping the voltages constant. The profiles that are labeled as '0/20' and '0/50' correspond to the situations in which molecules are trapped for 20 and 50 ms, respectively.



**Fig. 2.** Measured time-of-flight (TOF) profiles when  $\text{ND}_3$  molecules are guided at 90 m/s, decelerated to 85–0 m/s and trapped for 20 and 50 ms before being accelerated back to 90 m/s and detected (upper graph). The lower graph shows the full-width at half-maximum (FWHM) (black curve, left-hand side axis) and the amplitudes (red curve, right-hand side axis) of Gaussian fits to the TOF profiles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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