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An AC electric trap for ground-state molecules

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We here report on the realization of an electrodynamic trap, capable of trapping neutral atoms and molecules in both low-field and high-field seeking states. Confinement in three dimensions is achieved by switching between two electric field configurations that have a saddle-point at the center of the trap, i.e., by alternating a focusing and a defocusing force in each direction. AC trapping of ¹⁵ND₃ molecules is experimentally demonstrated, and the stability of the trap is studied as a function of the switching frequency. A 1 mK sample of ¹⁵ND₃ molecules in the high-field seeking component of the $|J, K\rangle = |1, 1\rangle$ level, the ground-state of para-ammonia, is trapped in a volume of about 1 mm³.

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During the last years, trapping of molecules using optical fields [1], magnetic fields [2], and electric fields [3] has been experimentally demonstrated. In the largest and deepest traps thus far realized, made of static inhomogeneous magnetic and/or electric fields, paramagnetic or polar molecules in low-field seeking quantum states can be confined [4]. There is a special interest in polar molecules, as they are promising for a variety of fundamental physics studies and applications [5, 6]. For most of these investigations, however, trapping of molecules in high-field seeking states is required. For instance, only when polar molecules are trapped in their absolute ground-state, which is high-field seeking for any molecule, increasing the phase-space density via evaporative cooling is expected to be possible; the strong dipole-dipole interaction between polar molecules is predicted to lead to high trap loss rates due to inelastic collisions for molecules in low-field seeking states [7], thereby hampering the evaporative cooling process. Furthermore, when biomolecules or heavy diatomic molecules, e.g., those that are important for measurements on the electric dipole moment of the electron [8, 9], are to be trapped, a trap for molecules in high-field seeking states is the only viable option. Due to their small rotational energy level spacings, these molecules are purely high-field seeking already in relatively small electric or magnetic fields. In principle, optical trapping can be used for molecules in high-field seeking states [1, 10]. In this Letter we experimentally demonstrate an AC electric trap, a new trap for molecules in high-field seeking states with a depth and a volume that is considerably larger than obtainable with an optical trap.

Creating a maximum of the static electric field in free space in three dimensions, which would allow trapping of molecules in high-field seeking states, is fundamentally impossible [11]. In two dimensions such a field maximum can be created. In a cylindrically symmetric geometry, for instance, a static electric field can be made with a maximum in the radial direction and a minimum in the

axial direction, or vice versa. Switching between these two saddle-point configurations results in a field that is alternately exerting a focusing and a defocusing force in each direction for molecules in high-field (and low-field) seeking states. Confinement in three dimensions can thus be obtained.

In our experiment we create the two required saddle-point electric field configurations either by adding a hexapole field to, or by subtracting a hexapole field from, a dipole field, as suggested by Peik [12]. As switching between the two configurations involves changing the hexapole field only, the electric field in the center of the trap doesn't change, which has, for instance, the advantage that Majorana transitions are prevented. The magnitude of the force on a molecule, which is proportional to the distance from the center of the trap, is the same in the two electric field configurations, but the direction of the force is opposite. At any fixed position, the force on a molecule will average out over time. However, a molecule does not stay at a fixed position but will move towards the center of the trap under the influence of a focusing force. It will then be closer to the center, where the force is smaller, when the defocusing force is applied. This defocusing force will move the molecule further away from the center again, bringing it in a region of a larger force when the focusing force is applied. On average, molecules will thus be further away from the center of the trap when the focusing force is applied than they are while being defocused, resulting in a net focusing force. This is shown in Fig. 1, where three trajectories of molecules along the z-direction are displayed during several switching periods. The molecules perform a fast micromotion at the switching frequency superimposed on a slower 'secular' motion. The grey area encompasses all stable trajectories, and shows the size of the package of molecules along the z-direction in the trap as a function of time. This size is indeed seen to be largest (smallest) when focusing (defocusing).

Whether the trap is stable or not depends on the (ra-

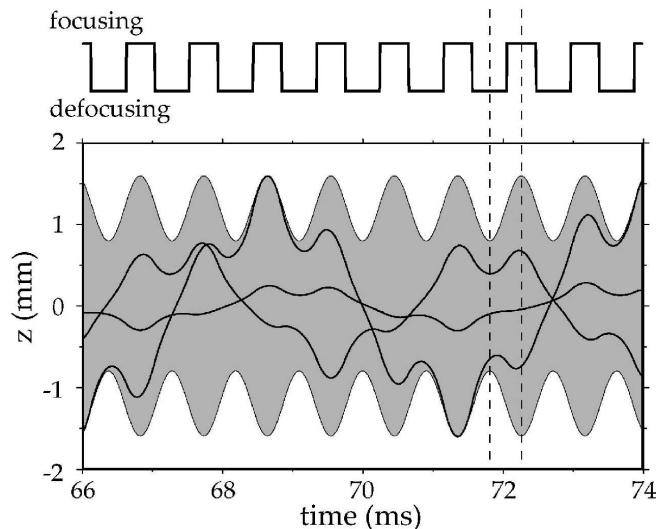


FIG. 1: Trajectories of molecules along the z -direction of the AC trap as a function of trapping time, for a switching frequency of 1100 Hz. The block-wave above the graph indicates whether the electric field configuration is focusing or defocusing along \hat{z} . The grey area shows the size of a package of molecules, which is seen to be largest when the electric field configuration is focusing. The two dotted lines are at 79 and 79.5 periods of switching, when the measurements on the size of the trapped sample shown in Fig. 4 are performed.

dial) frequency Ω with which the two electric field configurations are switched. This is most conveniently discussed when it is assumed that the electric hexapole field is modulated with a sinusoidal function, rather than with a square wave. The qualitative behavior of the trap as a function of Ω will be the same, but for a sinusoidally varying field the trajectories of the molecules are solutions of the well-known Mathieu equation [12, 13]:

$$\frac{d^2 z}{d\tau^2} + (a - 2q_z \cos 2\tau)z = 0, \quad (1)$$

with

$$a = 0; \quad q_z = \frac{12\mu_{eff}U_3}{m\Omega^2 z_0^3}; \quad \tau = \frac{\Omega t}{2}, \quad (2)$$

where μ_{eff} is the effective dipole moment of the molecule, U_3 is the amplitude of the oscillating hexapole term, m is the mass of the molecule, and z_0 is the characteristic radius of the trap. When $a = 0$, solutions of the Mathieu equation are stable along the z -direction when $|q_z| < 0.907$. If the voltages on the trap are kept constant, the value of $|q_z|$ can be changed by changing Ω . At low frequencies, solutions to the Mathieu equation are unstable and the amplitude of the molecular motion will increase exponentially in time. Above some cut-off frequency, the trap will abruptly become stable. When the frequency is increased further, the molecules have less time to move in between switching times. Their amplitudes during focusing and defocusing approach each

other and the net force on the molecules averages out more. The trap will thus be deepest for frequencies just above the cut-off frequency and will become less deep for higher frequencies, whereas no trapping at all is possible for frequencies below the cut-off frequency. The same arguments and equations apply for the x - and y -direction, with $q_x = q_y = -q_z/2$.

The operation principle of this AC electric trap for neutral polar molecules is similar to that of a Paul trap for charged particles [14]. Using magnetic fields, AC trapping has already been demonstrated for cesium atoms [15] and several proposals for an AC electric trap for atoms have been put forward [12, 16, 17]. In two dimensions the same principle has been used to focus neutral molecules in high-field seeking states in an alternate gradient (AG) decelerator [19, 20] and in an AC electric guide [18]. It is noted that in an AG focuser constant voltages can be applied; the molecules will experience an alternating field due to their forward velocity.

In the experimental setup shown in Fig. 2 a mixture of 5% $^{15}\text{ND}_3$ seeded in xenon expands from a pulsed valve at a 10 Hz repetition rate. In the expansion region, about 60% of the ammonia molecules cool to the $|J, K\rangle = |1, 1\rangle$ level, the ground-state of para-ammonia. After passing through a skimmer, the beam is coupled into a Stark decelerator by a hexapole. This part of the set-up and the operation principle of the decelerator have been described in detail elsewhere [21, 22]. Upon exiting the decelerator with a forward velocity of around 15 m/s, $^{15}\text{ND}_3$ molecules in the low-field seeking component of the $|J, K\rangle = |1, 1\rangle$ level are transversally and longitudinally focused into the AC trap by a second hexapole and a buncher [23], respectively. A cross-section of the AC electric trap together with the voltages applied to the electrodes for the two different electric field configurations is shown in Fig. 2 [25]. The trap has a hexapole geometry consisting of two ring electrodes with an inner diameter of 10 mm and two end caps, with a closest separation of 9.1 mm. Between the two ring electrodes there is a 2.9 mm gap through which the UV detection laser is coupled in. There is a 2 mm diameter hole in the entrance end cap for coupling the $^{15}\text{ND}_3$ molecules into the trap and a 2 mm diameter hole in the exit end cap for extracting the laser-produced molecular ions. When the package of slow ammonia molecules enters the trap, voltages are applied to the trap electrodes such that molecules are brought to a standstill near the center of the trap. At that time a 20 μs duration pulse of 1.43 GHz radiation can be coupled in to induce the transition from the low-field seeking to the high-field seeking hyperfine levels in the $|J, K\rangle = |1, 1\rangle$ state of $^{15}\text{ND}_3$ [22]. Under optimum conditions, about 20% of the ammonia molecules are pumped to high-field seeking levels.

When the AC electric trap is switched on, the voltages on the trap are alternated at a frequency Ω between the two configurations shown in Fig. 2. The configurations on the left-hand side and on the right-hand side in the figure focus high-field (low-field) seeking molecules in the axial

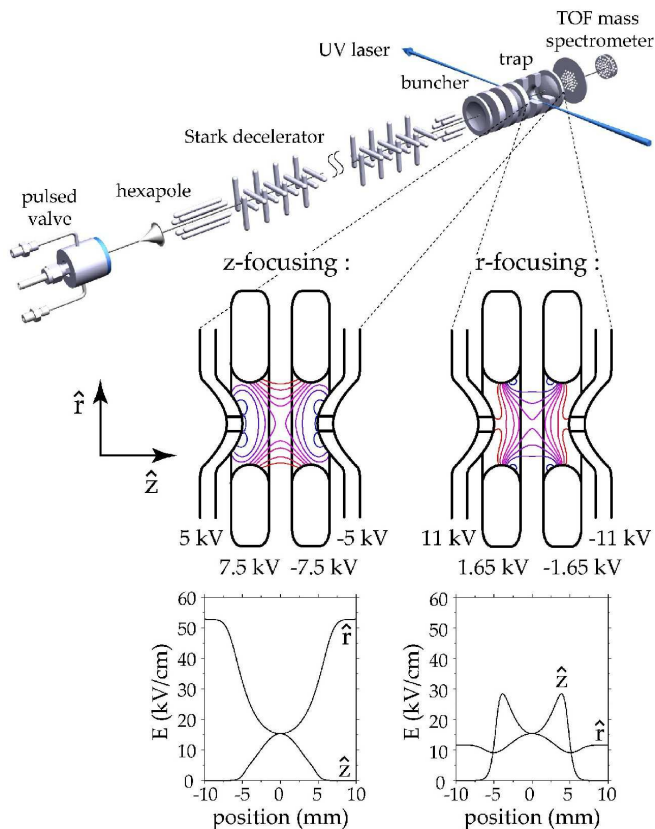


FIG. 2: Experimental setup. A molecular beam of $^{15}\text{ND}_3$ molecules in the low-field seeking component of the $|J, K\rangle = |1, 1\rangle$ level is decelerated and brought to a standstill in the AC electric trap. Before turning on the trap a transition from low-field seeking to high-field seeking levels can be made by applying a microwave pulse. Cross-sections of the trap are shown with lines of equal electric field strength (contours are shown at 5, 10, 12.5, 15, 17.5, 20, 25 and 30 kV/cm, increasing from blue to red (on-line only)) for both electric field configurations, together with plots of the electric field strength along the r- and the z-direction.

(radial) and the radial (axial) direction, respectively. The resulting trapping potential is about twice as deep in the axial direction as it is in the radial direction. Therefore, increasing the trap depth in the r-direction at the cost of decreasing the trap depth in the z-direction will improve the overall trap performance. This can experimentally be done by adjusting the duty cycle, i.e., the fraction of each switching period during which the electric field configuration is applied that is denoted as the z-focusing configuration in Fig. 2. In the measurements the duty cycle is 45% when trapping high-field seekers and 55% when trapping low-field seekers. The trap is switched on with the r-focusing (z-focusing) configuration when trapping high-field (low-field) seekers, and the first switching to the other configuration is performed after a quarter period.

After a certain trapping time, the trap is switched off, and the molecules are detected using pulsed UV-laser ion-

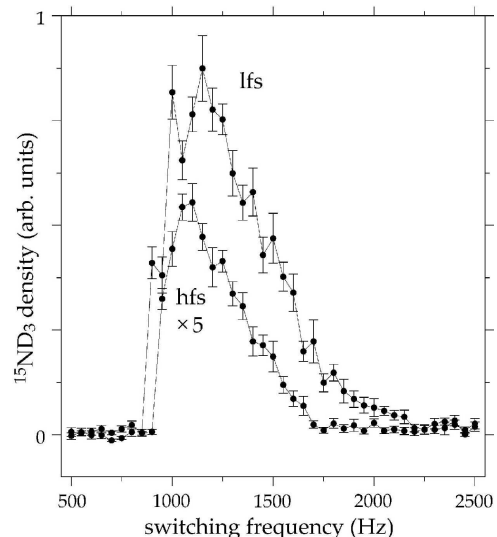


FIG. 3: Density of $^{15}\text{ND}_3$ molecules in low-field seeking (lfs) and high-field seeking (hfs) levels of the $|J, K\rangle = |1, 1\rangle$ state at the center of the trap as a function of the switching frequency, after the trap has been on for 72 ms. The signal of the high-field seekers is scaled up by a factor of 5.

ization followed by mass-selective detection of the parent ions. The (2+1) Resonance Enhanced Multi Photon Ionization (REMPI) scheme that is used selectively ionizes the $^{15}\text{ND}_3$ molecules in the upper or lower component of the $|J, K\rangle = |1, 1\rangle$ inversion doublet, containing the low-field seeking or high-field seeking levels, respectively.

In Fig. 3 the density of $^{15}\text{ND}_3$ molecules at the center of the trap is shown as a function of the switching frequency for molecules in low-field seeking and high-field seeking states. The signal for the high-field seekers is scaled up by a factor of five, to correct for the 20% conversion efficiency in the microwave pumping process. To arrive at the relative densities, a correction has also been made for the quantum-state specific ionization efficiency. The measurements are made after the AC trap has been on for 72 ms, enabling data-acquisition at the 10 Hz repetition rate of the experiment. The trapping lifetime is actually expected to be several tenths of seconds, limited by the residual background pressure. The measurements agree with the qualitative description of the dependence of the stability of the trap on Ω as described above. If the cut-off frequency is calculated using Eq. 2 with $\mu_{eff} = 0.75$ Debye, $z_0 \approx 4.5$ mm, and $U_3 = 3$ kV, a frequency of around 890 Hz is found. The numerically calculated cut-off frequency for $^{15}\text{ND}_3$ in this trap is around 980 Hz, in good agreement with the measurements. Note that the cut-off frequency for the high-field seekers is slightly higher than for the low-field seekers due to an about 4% difference in μ_{eff} in the electric field at the center of the trap. With the present settings, the highest density of trapped molecules is observed at a switching frequency of 1100 Hz. Due to the anharmonicity of the trap it is expected that the trap works better

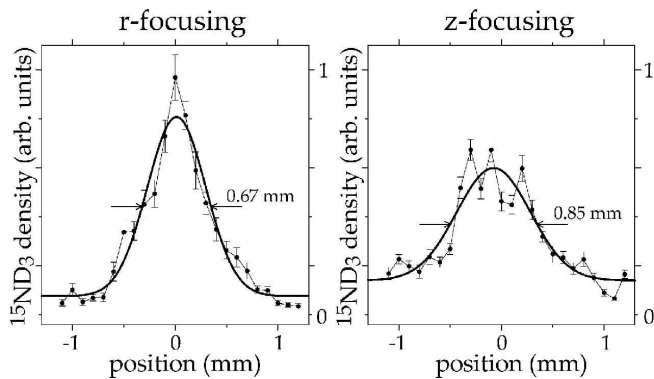


FIG. 4: Spatial distribution along \hat{z} of $^{15}\text{ND}_3$ molecules in high-field seeking states after trapping at 1100 Hz for 79 (left) and 79.5 (right) switching periods. A Gaussian distribution is fitted to the measurements and the width (FWHM) of this distribution is given.

for molecules in low-field than in high-field seeking states, in agreement with the observations.

In Fig. 4 measurements of the spatial distribution along the z -direction are shown for $^{15}\text{ND}_3$ molecules in high-field seeking states. The measurements are performed after trapping for about 72 ms at a switching frequency of 1100 Hz. The data shown on the left are taken after 79 periods of switching, i.e., in the middle of r -focusing. The data shown on the right are taken half a period later, i.e., in the middle of z -focusing. It is clear from these measurements that the package of molecules in the z -

direction is indeed smaller during r -focusing (denoted as defocusing in Fig. 1) than it is during z -focusing (denoted as focusing in Fig. 1). From the observed spatial distributions, a temperature of the trapped sample of around 1 mK is deduced.

In the AC electric trap demonstrated here, neutral atoms and molecules, both in low-field and in high-field seeking states, can be trapped. For molecules in high-field seeking states only optical traps, which have a typical trap depth below 1 mK and a typical volume of 10^{-5} cm^3 [10], had been demonstrated thus far; a considerably deeper and larger microwave trap has recently been proposed [24]. We have demonstrated here, using $^{15}\text{ND}_3$ as a prototypical polar molecule, an AC electric trap with a trap depth of about 5 mK and a trapping volume of about 20 mm^3 . This relatively deep and large trap for ground-state molecules holds great promise for future applications.

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