Radboud Repository

Radboud University Nijmegen

PDF hosted at the Radboud Repository of the Radboud University Nijmegen

The following full text is a preprint version which may differ from the publisher's version.

For additional information about this publication click this link.
http://hdl.handle.net/2066/99118

Please be advised that this information was generated on 2017-12-06 and may be subject to change.

Nonadiabatic transitions in electrostatically trapped ammonia molecules

Moritz Kirste, Boris G. Sartakov[1], Melanie Schnell[*], and Gerard Meijer

Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, D-14195 Berlin, Germany
[1]*General Physics Institute RAS, Vavilov Street 38, 119991 Moscow, Russia*
(Dated: June 18, 2009)

Nonadiabatic transitions are known to be major loss channels for atoms in magnetic traps, but have thus far not been experimentally reported upon for trapped molecules. We have observed and quantified losses due to nonadiabatic transitions for three isotopologues of ammonia in electrostatic traps, by comparing the trapping times in traps with a zero and a non-zero electric field at the center. Nonadiabatic transitions are seen to dominate the overall loss rate even for the present samples that are at relatively high temperatures of 30 mK. It is anticipated that losses due to nonadiabatic transitions in electric fields are omnipresent in ongoing experiments on cold molecules.

The recent development of a large variety of methods and devices for the manipulation and trapping of neutral polar molecules offers new opportunities for molecular physics experiments [1]. Decelerated beams and trapped samples of polar molecules can be used to study intrinsic molecular properties, such as energy level splittings [2, 3] and lifetimes of metastable states [4, 5], with unprecedented precision. Once the densities of the trapped molecules become high enough and their temperatures become low enough, the inter-molecular interactions are anticipated to enable interesting new studies and applications [6, 7]. For all these studies it is not only of importance to increase the phase-space density of the trapped molecules but also to increase the time during which the molecules can stay confined in the trap, i.e., to reduce the trap loss processes. Neutral polar molecules in low-field seeking states are routinely trapped in magnetostatic or electrostatic traps by exploiting the Zeeman or Stark effect, respectively [8, 9]. These traps typically exhibit zero field at the trap center. Within a certain area around the trap center, the trapped molecules can undergo nonadiabatic transitions, widely also referred to as spin flip or Majorana transitions, from a trapped state into a state in which the molecules are no longer trapped. In atomic physics, nonadiabatic transitions in quadrupole magnetic traps seriously hindered the generation of the first Bose-Einstein condensates, as these spin flips made it impossible to reach the required ultracold regime [10, 11]. The trap losses associated with the presence of the zero field at the trap center were eliminated by implementing the TOP (time-averaged, orbiting potential) trap on the one hand [12] and by keeping the atoms away from the trap center with an optical plug on the other hand [13]. Generally, trap loss due to nonadiabatic transitions can be completely suppressed by creating a non-zero field minimum in the trap center. Already in 1962, Ioffe introduced a special variation of a magnetostatic trap with a field offset in the center for nuclear physics experiments [14]. Pritchard suggested in 1983 to use such a trap for the confinement of neutral atoms [15]. This type of magnetostatic trap is now widely known as the Ioffe-Pritchard (IP) trap.

For trapped polar molecules, losses due to nonadiabatic transitions have not been experimentally reported upon yet. Nevertheless, several possible geometries for an electrostatic analogue of an IP-type trap have been suggested to prevent these possible losses, such as a chain-linked trap [16] or a six-wire trap [17]. More recently, a trap with an electric field offset in the center has been demonstrated for Rydberg atoms [18]. Theoretical studies on the loss of molecules from purely electrostatic traps [19] or, more generally, from magneto-electrostatic traps [20] due to nonadiabatic transitions have also recently appeared. In both studies it was concluded that – although this can be different for any particular molecule – the loss rate is negligible at the milli-Kelvin temperatures that are currently achieved in electrostatic traps.

In this Letter, we experimentally study trap losses due to nonadiabatic transitions in electrostatically trapped ammonia molecules. We quantify this trap loss mechanism from measurements of the trapping times in an IP-type electrostatic trap with either a zero field or an offset field at the center, under otherwise identical conditions. Moreover, we measured the trapping times for three isotopologues of ammonia, namely $^{14}NH_3$, $^{14}ND_3$ and $^{15}ND_3$. These molecules have subtle differences in their energy level structure, resulting in different probabilities for nonadiabatic transitions. Contrary to the theoretical expectations, the results demonstrate the enormous importance of trap losses due to nonadiabatic transitions even at the present 30 mK temperatures.

Figure 1(a) shows the energy of the $|J, K\rangle = |1, 1\rangle$ level of NH_3 and ND_3 in electric fields up to 150 kV/cm. The upper (lower) inversion doublet components of this level are seen to split into a low-field (high-field) seeking set of hyperfine levels labeled MK=-1 (MK=+1) and into a set of hyperfine levels labeled MK=0 that are basically not shifted in the fields. The zero-field inversion splitting of $^{14}NH_3$ is with W_{inv}=23.7 GHz considerably larger than the W_{inv}=1.59 GHz of $^{14}ND_3$. At low electric fields, the Stark shift of the MK=-1 set of levels is proportional to E^2/W_{inv}, and is therefore considerably less for $^{14}NH_3$ than for $^{14}ND_3$ in a given electric field with magnitude E. In Figure 1(b) the Stark shift of the upper inversion doublet component of $^{14}NH_3$ is shown for low electric fields on an expanded scale, such that the individual hyperfine

levels can be recognized. The quadrupole interaction of the ^{14}N nucleus causes the observed main splitting in three groups of levels, labeled with the quantum number F_1 [21]. The nuclear spins of the hydrogen atoms cause small additional splittings. In the electric field, the overall splitting behavior into (almost) unshifted levels belonging to MK=0 and levels that shift quadratically with E, belonging to MK=-1, is recognized. The individual hyperfine levels have a definite angular momentum projection quantum number M_F, where $\vec{F}$ is the total angular momentum. As the electric field scales linearly with the distance from the center in a quadrupole electrostatic trap, a distance scale (x, in μm) appropriate for the trap used in our experiment is indicated on top of the Figure as well. In the lower two panels of Figure 1 the behavior of the hyperfine levels of ^{14}ND$_3$ and ^{15}ND$_3$ in electric fields is shown [22]. Note that the electric field scale, and thereby the distance scale, is now a factor four smaller than for ^{14}NH$_3$. For ^{14}ND$_3$ and ^{15}ND$_3$, the magnitude of the inversion splitting is quite similar (1.59 GHz and 1.43 GHz, respectively) but their hyperfine structure is rather different. As the deuterium atoms have a nuclear spin of one, the hyperfine structure in the deuterated isotopologues of ammonia is generally quite complicated. In ^{14}ND$_3$, the hyperfine levels again split in three groups of levels due to the quadrupole coupling of the ^{14}N nucleus [22]. The ^{15}N nucleus has no quadrupole moment, resulting in less (11, instead of 16 for ^{14}ND$_3$) hyperfine levels, but these are closer spaced and without a clear sub-structure for ^{15}ND$_3$ [2].

Polar molecules are oriented by the local electric fields inside the trap, and they normally remain in the same quantum state and adiabatically follow the field while moving through the trap. The adiabatic eigenstates can be quantized with respect to the axis along the electric field vector $\vec{E}$ and can thus be assigned the quantum number M_F. Nonadiabatic transitions can occur when the molecules can not follow the rapid change of the direction of the electric field when they pass with velocity v at a close distance x to the region of zero electric field at the trap center. When this rate of change, given by v/x, is larger than the energy difference between levels in the molecule, nonadiabatic transitions can occur. As $\vec{v}$ is in general not oriented along $\vec{E}$, nonadiabatic transitions with ΔM_F=0 as well as with $\Delta M_F = \pm 1$ will be possible. In the case of ammonia, nonadiabatic transitions from the MK=-1 to the MK=0 set of hyperfine levels will cause trap loss. The frequency of these transitions at a distance x from the trap center is proportional to x^2/W_{inv}. From this it is seen that nonadiabatic transitions can occur when the molecules come closer to the trap center than a typical distance $b \propto (W_{inv}v)^{1/3}$. In Figure 1(b)-(d) curves of v/x are indicated (in MHz) for two different velocities (v=1, 10 m/s), relative to the highest frequency MK=0 hyperfine level. The crossing point of these curves with each of the MK=-1 hyperfine levels gives the corresponding distance b for nonadiabatic losses from that particular hyperfine level. Obviously, when there is a large non-zero field at the center of the trap, nonadiabatic transitions from the MK=-1 to the MK=0 levels will no longer be possible. Nonadiabatic transitions within the MK=-1 set of hyperfine
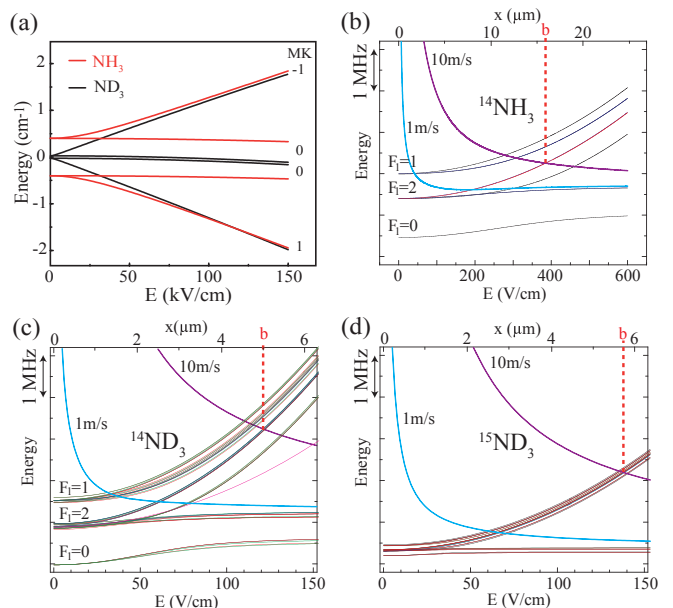

FIG. 1: (Color online) Energy of the $|J,K\rangle = |1,1\rangle$ level of ^{14}NH$_3$ (dashed line) and ^{14}ND$_3$ (straight line) in fields up to 150 kV/cm (a). In panel (b), (c) and (d) the Stark shift of the upper inversion doublet hyperfine components in low electric fields is shown on expanded scales for ^{14}NH$_3$, ^{14}ND$_3$ and ^{15}ND$_3$, respectively. For the calculation of the electric field dependence of the hyperfine components, see Ref. [22]. The v/x curves indicate the rate of change of the direction of the electric field for a molecule with velocity v (1 and 10 m/s) passing the trap center at a distance x (upper axis).

levels can still occur to some extent, although also these will be largely suppressed as only the magnitude of the electric field, but no longer the direction, changes rapidly when the molecules pass through the center of the trap.

In the experiment (*vide infra*), we detect molecules in all low-field seeking MK=-1 hyperfine levels simultanuously. It is evident from inspection of the Stark curves shown in Figure 1 that each of these levels will have a different nonadiabatic transition rate. Therefore, when the trap loss is dominated by nonadiabatic transitions, a multi-exponential decay is expected to be observed. The rate for nonadiabatic transitions depends on how often the molecule passes through the volume with cross-section b^2 near the center of the trap. This rate can be approximated by $f_{osc}(b/b_0)^2$, where f_{osc} is the oscillation frequency of the molecules in the trap and b_0 is the mean value of the impact parameter of the trajectories of the molecules. With a typical value for f_{osc} of 1-2 kHz and with b_0 on the order of 0.2 mm, a value of b around 5 μm will lead to a loss rate due to nonadiabatic transitions on the order of 1 Hz.

The experimental setup, together with an expanded view of the IP-type electrostatic trap, is schematically shown in Figure 2(a). A detailed description of the molecular beam machine, and in particular of the deceleration of a beam of ammonia molecules, is given elsewhere [23]. Decelerated pack-
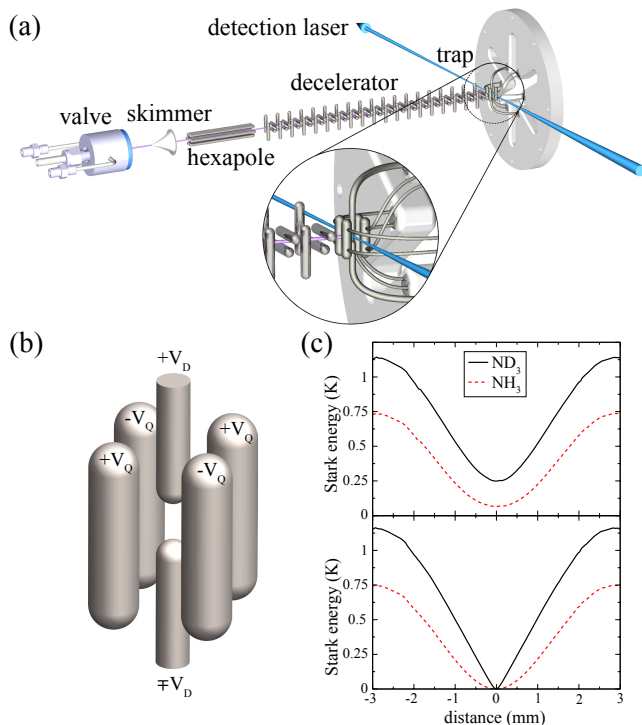
FIG. 2: Scheme of the experimental setup, with an expanded view of the IP-type electrostatic trap. The voltages applied to the six cylindrical electrodes are shown (b), together with the resulting potential energies for NH_3 (dashed line) and ND_3 (straight line) along a line in the horizontal symmetry plane for a non-zero (top) and a zero (bottom) electric field at the trap center (c).

FIG. 3: (Color online) (a) Lifetime measurements for $^{14}ND_3$ in the two trap configurations, both on a linear and on a logarithmic scale (inset). In the inset, straight lines are included to guide the eye. (b) Lifetime measurements for $^{15}ND_3$ with original ($V_{D/Q}$) and reduced ($V_{D/Q}/4$) trap voltages on a logarithmic scale. The measurements have been performed with intervals of 20 ms and with 16 averages per timepoint. The non-zero field in the trap center is 16 kV/cm for all measurements shown.

ets of either $^{14}NH_3$, $^{14}ND_3$, or $^{15}ND_3$ molecules leave the decelerator in the low-field seeking MK=-1 hyperfine levels of the $|J, K\rangle = |1, 1\rangle$ level with a mean velocity of around 20 m/s, and with a full width at half maximum (FWHM) velocity spread of about 10 m/s. The decelerated packet contains appproximately 10^5-10^6 molecules and has a spatial extent of about 2 mm along the molecular beam axis and is about 2x2 mm^2 in the transverse direction at the exit of the decelerator [23]. The molecules are decelerated to a mean velocity around zero m/s upon entering the trap, which is centered 8 mm behind the decelerator. After a certain trapping time, the trap is turned off and the molecules are state-selectively detected using a laser based ionization detection scheme.

The trap consists of six cylindrical electrodes, shown more clearly in Figure 2(b). To the four outer electrodes (3 mm diameter) positive and negative voltages ($\pm V_Q$) are applied such as to generate a quadrupole field. The two electrodes (2 mm diameter) centered on the symmetry axis of the trap enable to create an additional dipole field. When voltages of opposite polarity ($\pm V_D$) are applied to these electrodes, a non-zero electric field is generated at the center of the trap; by applying the same polarity ($+V_D$) to the center electrodes, a standard quadrupole trap with a zero field at the center is obtained. In Figure 2(c) the potential energy for the MK=-1 levels of NH_3
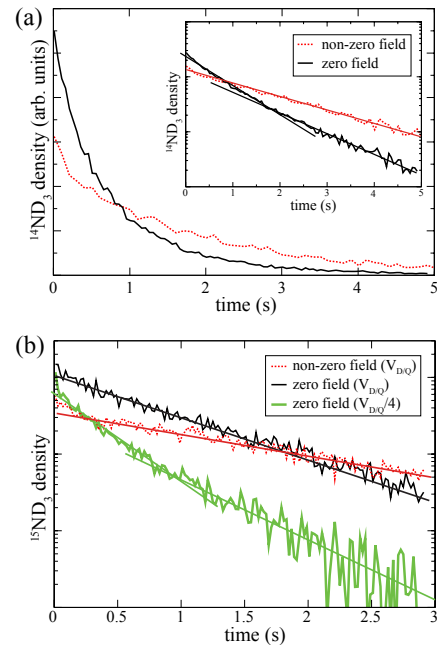
and ND_3 in the trap is shown along a line in the horizontal symmetry plane, both for a zero (bottom) and a non-zero field (top) at the trap center. In the latter case, the trap depth is actually limited by the occurence of four saddle points of the electric fields in between the electrodes. For V_Q=10 kV and V_D=3 kV, the offset electric field at the center is 16 kV/cm and the trap depth is about 290 mK for ND_3 and 130 mK for NH_3. With no electric field at the center, the trap is considerably deeper, about 1.2 K for ND_3 and 750 mK for NH_3.

Figure 3(a) shows measurements of the density of $^{14}ND_3$ molecules in the electrostatic trap as a function of the time during which the trap has been switched on, both for zero and non-zero field at the center. Although the number density in the zero field trap is originally higher due to a better trap loading, it is also seen to decay significantly faster. In both cases, the trap is considerably deeper than the temperature of the trapped ensemble of molecules; the latter has been determined from expansion measurements to be around 30 mK. The molecules can therefore only leave the trap via collisions, via optical pumping due to blackbody radiation or via nonadiabatic transitions. Losses due to collisions can either originate from (in)elastic collisions of the trapped molecules with background gas or from cold collisions amongst the trapped molecules. At the present densities in the trap (10^7-10^8 cm^{-3}), collisions of the trapped molecules with each

other can be neglected. We have experimentally verified this by changing the number density of trapped molecules over one order of magnitude, without observing any change in the trapping time. The $^{14}ND_3$ molecules in the low-field seeking MK=-1 hyperfine levels can be optically pumped to excited ro-vibrational levels due to blackbody radiation from the room-temperature environment, leading to a calculated finite lifetime of about 7 s [24]. The purely single exponential decay of the number of $^{14}ND_3$ molecules in the trap with the non-zero field has a 1/e time constant of 1.9 s. This decay time results from the combination of optical pumping due to blackbody radiation and (in)elastic collisions with background gas (background pressure 10^{-8} mbar). By simply changing the polarity on one of the center electrodes of the trap, under otherwise identical conditions, a much faster and multi-exponential decay is observed (Figure 3(a)); the additional trap loss observed in the trap with zero electric field at the center is solely due to nonadiabatic transitions. From these measurements, it appears that the nonadiabatic transitions for $^{14}ND_3$ can be approximated with only two time constants. Referring to Figure 1(c), the fast decay can be attributed to nonadiabatic transitions within the F_1=2 set of hyperfine levels, whereas the slow decay can be attributed to nonadiabatic transitions from the F_1=1 to the F_1=2 set of hyperfine levels. When the nonadiabatic transitions are modelled in this way (with the pre-factors for the two exponentials determined by the number of M_F levels), time constants of about 0.25 s and 2.0 s, respectively, are extracted for these processes. The errors on these 1/e time constants are less than 20 ms.

We have performed a similar series of experiments for $^{14}NH_3$ (data not shown); this is actually the first time that this ammonia isotopologue has been trapped at all. In the zero field trap, a bi-exponential decay is also observed for $^{14}NH_3$, which can be explained in the same way as for $^{14}ND_3$. After correction for the overall decay as measured for $^{14}NH_3$ in the trap with the offset field, the time constants for the fast and the slow process are determined as 0.13 s and 3.5 s. The faster initial decay can be explained by the larger value of b whereas the slower decay from the F_1=1 to the F_1=2 set of hyperfine levels than for $^{14}ND_3$ can be rationalized by the slightly larger energetic separation and the reduced number of levels of $^{14}NH_3$.

In Figure 3(b), the density of $^{15}ND_3$ molecules in the trap is shown as a function of time. In this case, the decay in the zero field trap is seen to be almost single exponential, and the 1/e time constant due to the nonadiabatic transitions is 1.2 s. A more pure single exponential behaviour can indeed be expected as all low-field seeking hyperfine levels behave very similar in the electric field (see Figure 1(d)). When all voltages on the trap (and thus also the electric field offset in the non-zero field trap) are reduced by a factor of four, the lifetime in the trap with the offset electric field stays the same (data not shown). In the zero field trap, however, the decay is not only faster due to the larger value of b but it is also multi-exponential as the grouping of the hyperfine levels is now relatively more important.

As evidenced by the experimental results presented in this work, nonadiabatic transitions can be a dominant loss channel in electrostatically trapped molecules, even at temperatures of tens of milli-Kelvins. These trap losses can be effectively prevented by using an electrostatic trap with a non-zero electric field at the center, or, for instance, by orbiting the molecules off-center in a storage ring [25]. The importance of trap losses due to nonadiabatic transitions depends on the detailed energy level structure of the molecule under consideration. Contrary to the situation in magnetic fields, there is not necessarily a degeneracy of low-field and high-field seeking levels for molecules at zero electric field. As a consequence, molecules in levels that are exclusively low-field seeking, like $^{14}NH_3$ and $^{14}ND_3$ molecules in the F_1=1 hyperfine manifold of the $|J, K\rangle = |1, 1\rangle$ level (see Figure 1(b,c)), can be rather immune to nonadiabatic transitions. For some applications it might not be desirable to introduce additional offset fields. In that case it might be possible to select another isotopologue with a slightly different hyperfine structure to effectively suppress the losses due to nonadiabatic transitions.

*corresponding author: schnell@fhi-berlin.mpg.de

[1] S. Y. T. van de Meerakker, H. L. Bethlem, and G. Meijer, Nature Phys. **4**, 595 (2008).
[2] J. van Veldhoven, et al., Eur. Phys. J. D **31**, 337 (2004).
[3] E. R. Hudson, et al., Phys. Rev. Lett. **96**, 143004 (2006).
[4] S. Y. T. van de Meerakker, et al., Phys. Rev. Lett. **95**, 013003 (2005).
[5] W. C. Campbell, et al., Phys. Rev. Lett. **100**, 083003 (2008).
[6] R. V. Krems, Int. Rev. Phys. Chem. **24**, 99 (2005).
[7] A. André, et al., Nature Phys. **2**, 636 (2006).
[8] J. D. Weinstein, et al., Nature **395**, 148 (1998).
[9] H. L. Bethlem, et al., Nature **406**, 491 (2000).
[10] W. Ketterle, Rev. Mod. Phys. **74**, 1131 (2002).
[11] E. A. Cornell and C. E. Wieman, Rev. Mod. Phys. **74**, 875 (2002).
[12] W. Petrich, et al., Phys. Rev. Lett. **74**, 3352 (1995).
[13] K. B. Davis, et al., Phys. Rev. Lett. **75**, 3969 (1995).
[14] Y. V. Gott, M. S. Ioffe, and V. G. Telkovsky, Nuclear fusion **3**, 1045 (1962).
[15] D. E. Pritchard, Phys. Rev. Lett. **51**, 1336 (1983).
[16] N. E. Shafer-Ray, et al., Phys. Rev. A **67**, 045401 (2003).
[17] G. Xu, Ph.D. thesis, The University of Texas at Austin (2001).
[18] S. Hogan and F. Merkt, Phys. Rev. Lett. **100**, 043001 (2008).
[19] M. Kajita, Eur. Phys. J. D **38**, 315 (2006).
[20] M. Lara, B. L. Lev, and J. L. Bohn, Phys. Rev. A **78**, 033433 (2008).
[21] S. G. Kukolich, Phys. Rev. **156**, 83 (1967).
[22] J. van Veldhoven, et al., Phys. Rev. A **66**, 32501 (2002).
[23] H. L. Bethlem, et al., Phys. Rev. A **65**, 053416 (2002).
[24] S. Hoekstra, et al., Phys. Rev. Lett. **98**, 133001 (2007).
[25] F. M. H. Crompvoets, et al., Nature **411**, 174 (2001).