

Taming molecular beams; towards a gas-phase molecular laboratory on a chip

Samuel A. Meek, Gabriele Santambrogio, Horst Conrad, and Gerard Meijer

Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, D-14195
Berlin, Germany

e-mail: meijer@fhi-berlin.mpg.de

Abstract: The manipulation of gas-phase molecules with electric and magnetic fields above a chip is an emerging field of research. Miniaturization of the electric and magnetic field structures allows for the creation of large field gradients and tight traps above the chip. Present-day microelectronics technology enables the integration of complicated tools and devices on a compact surface area. The molecules can be positioned extremely accurately and reproducibly above the chip where they can be held isolated from their environment and where there is excellent access to them. It is expected that several of the gas-phase molecular beam experiments that are currently being done in machines that are up to several meters in length can in the future be performed on a surface area of a few cm² and that many new experiments will become possible.

In this progress report, we start with a description of the state-of-the-art and the objectives of the manipulation of molecular beams with electric and magnetic fields, using “conventional” (macroscopic) set-ups. Then we will discuss the objectives for a molecular laboratory on a chip and the operation principle of a decelerator on a chip is briefly explained. This decelerator is the essential tool that makes the loading of molecules directly from a molecular beam onto the chip possible, and that enables novel detection schemes for molecules on the chip to be implemented. Some possible future molecular physics experiments are discussed at the end.

1. Manipulation of molecules with external fields; state-of-the-art and objectives

Atomic and molecular beams have played central roles in many experiments in physics and chemistry – from seminal tests of fundamental aspects of quantum mechanics to molecular reaction dynamics – and have found a wide range of applications [1]. Nowadays, sophisticated laser-based detection methods exist to selectively detect molecules in specific quantum states. In the early days, quantum-state selectivity in the detection process was achieved by implementing inhomogeneous magnetic and/or electric field sections that influenced the trajectories of the particles to the detector. This was the approach used by Otto Stern and Walther Gerlach in 1922 [2], and the key concept of their experiment, *i.e.* the sorting of quantum states via space quantization, has been extensively used ever since. The original experimental geometries were devised to create strong magnetic or electric field gradients on the beam axis to efficiently deflect particles [3]. Later, both magnetic and electric field geometries were designed to focus particles in selected quantum states onto the detector. An electrostatic quadrupole focuser was used to couple a beam of ammonia molecules into a microwave cavity. The resulting inverted population distribution led to the invention of the maser by Gordon, Zeiger and Townes in 1954-1955 [4]. By using several multipole focusers in succession, with interaction regions with electromagnetic radiation in between, versatile set-ups to unravel the quantum structure of atoms and molecules were developed. In scattering experiments, multipole focusers in combination with electrostatic orientation fields were exploited to study steric effects. Variants of these methods were implemented in many laboratories, and have yielded a wealth of detailed information on stable molecules, radicals and molecular complexes, thereby contributing enormously to our present understanding of intra- and inter-molecular forces.

The manipulation of beams of atoms and molecules with electric and magnetic fields is thus about as old as the field of atomic and molecular beams itself, and it actually has been crucial for the success of the latter field. However, this manipulation exclusively involved the transverse motion of the molecules. In 1999, we experimentally demonstrated that appropriately designed arrays of electric fields in a so-called Stark decelerator can also be used to influence and control the longitudinal (forward) velocity of the molecules in a beam, for example, to decelerate a beam of neutral polar molecules [5]. Since then a variety of decelerators have been designed and built, and have made a whole variety of new experiments possible. For instance, when the molecules are decelerated to a standstill, they can be loaded and confined in traps. This enables the observation of molecules in complete isolation from their environment for times up to several seconds, and enables the investigation of molecular properties and their interactions in unprecedented detail.

The Stark decelerator for neutral polar molecules is conceptually similar to a linear accelerator for charged particles. Whereas in charged particle accelerators the force that is exerted on a particle depends on its charge and the electric field strength, in a Stark decelerator, the *dipole* (charge times separation) in a polar molecule is acted upon by *electric field gradients*. This quantum-state specific force is typically eight orders of magnitude smaller than the forces that are used in charged particle accelerators, but nevertheless suffices to achieve complete control over the motion of polar molecules using techniques akin to those used to control charged particles. A Stark decelerator allows one to decelerate (or to accelerate) part of the molecular beam to any desired velocity, while keeping the selected part of the beam together as a compact packet. After exiting the decelerator, these decelerated packets of molecules can be investigated while in free flight in the beam machine, or they can be confined in a variety of traps. Trapping of state-selected ND₃ molecules in a quadrupole electrostatic trap [6], in an AC electric trap [7] (equivalent to a Paul trap for ions), in an electrostatic storage ring [8] and in a molecular synchrotron [9] has been demonstrated. Other molecules like, for instance, OH radicals have also been trapped in pure electrostatic [10] and magnetostatic [11] traps as well as in traps using a combination of magnetic and electric fields [12].

Inspired in part by the manipulation of polar molecules with electric fields, a magnetic analogue of the Stark decelerator has been developed. Deceleration based on the magnetic interaction allows the manipulation of a wide range of atoms and molecules to which the Stark deceleration technique cannot be applied. The Zeeman deceleration technique (not to be confused with the optical Zeeman-slowing method that has been used extensively for alkali atoms) has first been experimentally demonstrated by the deceleration of ground-state H and D atoms [13,14]. It has also been applied to bring metastable Ne atoms and ground-state oxygen molecules to low velocities [15].

The new level of control over the motion and orientation of molecules like ammonia and the hydroxyl radical that has been obtained offers a multitude of new possibilities for precision studies of molecular properties and interactions. During the last few years, these new possibilities have been demonstrated in (i) beam collision studies, in (ii) spectroscopic studies, and in (iii) lifetime measurements on molecules that are confined in traps.

(i): The dynamics of collisions between molecules is highly sensitive to the relative velocity of the colliding species, in particular at low velocities where the kinetic energy of the molecules is comparable to the rotational energy level splitting in the collision complex. At these energies, translational energy can be transferred into rotational energy, effectively binding the molecules transiently together. Long living excitations of the collision complex show up as sharp resonances in the collision energy dependence of the scattering cross sections [16-18]. The measurement of these resonances provides stringent tests of the intermolecular potential energy surfaces. Molecular beams with a tuneable velocity are well suited to investigate this, as we have demonstrated by scattering a Stark decelerated beam of OH radicals with a conventional beam of Xe atoms [19]. By varying the collision energy over the energetic thresholds for different scattering channels, the threshold behaviour of the inelastic cross sections could be accurately measured. Scattering of magnetically trapped OH radicals with incoming beams has been used for this purpose as well [11].

(ii): Ultimately, the precision in any spectroscopic measurement is limited by the interaction time of the particle to be investigated with the radiation field. In conventional molecular beam experiments this interaction time is typically a few hundred microseconds. The ability to produce slow intense molecular beams enhances significantly the obtainable interaction time and hence resolution. The improved resolution can potentially be used for stringent tests of fundamental physics theories. For instance, polar molecules are being used to test violation of time-reversal symmetry (in particular, YbF [20], PbO [21] or PbF[22]), in the search for a difference in transition frequency between chiral molecules that are each other's mirror image [23], and for testing a possible time-variation of the proton-to-electron mass ratio [24,25]. Proof-of-principle high-resolution microwave spectroscopy experiments using Stark decelerated beams of $^{15}\text{ND}_3$ [26] and OH [27] have been performed.

(iii): Trapped molecules can be investigated for several seconds. This long interaction time is often of limited use to perform high-resolution spectroscopy, as the molecules interact with the trapping field [28]. However, it allows one, for instance, to accurately measure lifetimes of long-lived metastable states. We have used electrostatic trapping of molecules after Stark deceleration to measure the lifetime of the first vibrationally excited state of OH ($X^2\Pi$, $v''=1$), thereby benchmarking the Einstein A-coefficients in the important Meinel system of OH [29]. The same experimental approach has been applied to accurately measure the lifetimes of CO molecules in the electronically excited metastable $a^3\Pi$ state [30]. An alternative method to load molecules in a trap is by injecting a fast beam of molecules into a cryogenic cell filled with He gas. Thermalizing collisions with the He atoms make the molecules translationally and rotationally sufficiently cold that they can then be trapped in a quadrupole magnetic trap that is superimposed on the cryogenic cell. Trapping of CaH molecules via this so-called buffer-gas cooling method has been reported as early as in 1998 by John Doyle and co-workers [31], and, more recently, they have used this approach to accurately measure the lifetime of the vibrationally excited level of the imidogen radical, NH ($X^3\Sigma$, $v''=1$) [32].

The experimental tools described above that have been developed during the last decade to control the internal and external degrees of freedom of neutral molecules have caused a renaissance of the field of molecular beams; experiments that were unthinkable ten years ago seem routine by now. At the same time one has to realize, however, that the level of control that one has nowadays over polar molecules is still quite a bit inferior to that for atoms. Inspiration for methods to further improve the control over molecules can be obtained from the field of ultracold atoms. An important development in the latter field has been the manipulation of atoms above a chip using magnetic fields produced by current carrying wires [33]. Such atom chips have revolutionized the field of quantum degenerate gases, allowing a great experimental simplification in creating degenerate quantum matter [34]. The atom chips have been employed for matter-wave interferometry and in inertial and gravitational field sensing [35] and are sufficiently robust that they can be used, for instance, in free-fall experiments [36].

2.1. Trapping molecules on a chip; methodology and objectives

Many of the advances that have been foreseen for microscopic magnetic traps for neutral atoms [37] and for “integrated atom optics” on a chip [38] about a decade ago have become reality by now. The decisive advantage of the versatility of the lithographic wire structures can also be exploited for molecules, adding a new dimension to this field. The ability of a molecule to rotate and to vibrate allows for the coupling to photons over an enormous range of frequencies. Coupling at microwave frequencies, in particular, provides a convenient interface between quantum optic and solid-state technologies [39] and might enable, for instance, to implement proposed schemes of quantum computation that use polar molecules as qubits [40]. The use of miniaturized traps brings quantum-degeneracy for samples of polar molecules closer. A tight trap permits fast adiabatic changes of the confining potential and via compression of a cloud of trapped molecules increased thermalization rates and shortened times for forced evaporative cooling can be achieved. It will be interesting to study the lifetime of the molecules in the miniaturized traps, close to the surface. Apart from the intrinsic molecular lifetime and trap losses due to nonadiabatic (‘spin-flip’) transitions, the reduction of the trapping time due to heating of the

molecules by interaction with the surface can be studied as a function of temperature and distance from the surface. The developments in the control of neutral molecules with electric and magnetic fields together with the advances demonstrated by the manipulation of atoms on a chip, makes us believe that a wide variety of molecular physics experiments can – and will – in the future be performed on a chip.

There are two major obstacles that make experiments with molecules on a chip less straightforward than with atoms. First, the methods to load molecules onto the chip are less matured than for atoms. It is more difficult to produce dense samples of quantum-state selected cold molecules than it is for atoms. As discussed above, several experimental schemes that have been demonstrated to produce samples of trapped, cold molecules start with a molecular beam. The molecules in the beam are decelerated by the use of electric, magnetic or optical fields, and are subsequently trapped [41], but it is non-trivial to transport these samples onto the chip. Recently, spectacular progress has been made in the production of samples of ultracold heteronuclear alkali dimers in their ro-vibrational ground state level [42,43], starting from samples of ultracold atoms. It is anticipated that these samples of polar molecules can be loaded from the optical trap in which they are held now into electric field traps above a chip in the not too distant future.

A second obstacle for the experiments with molecules on a chip is that detection methods for molecules are (much) less sensitive than those for atoms. As molecules in general lack a closed two-level system, efficient detection using absorption or laser induced fluorescence is not possible; whereas a single alkali atom can scatter up to 10^7 photons per second, a molecule normally ends up in a different quantum-state – and turns dark – after scattering only a few photons. In vacuum, molecules can normally be most sensitively detected using ionization based detection schemes, but these schemes are difficult to implement close to the chip. Recent results in detecting atoms directly above a chip using laser-induced fluorescence [44] or photo-ionization [45] indicate that on-chip detection of molecules might nevertheless become possible in the future. Detection of molecules on a chip using micro-cavities, like is done for atoms [46,47], might also be an option, although even there, molecules have the disadvantage that a single ro-vibrational transition in an electronic band is generally weaker than an electronic transition for atoms, and that for many molecules the electronic transitions are in the more difficult to access near-UV region of the spectrum.

We have recently demonstrated that one can trap molecules on a chip using direct loading from a pulsed supersonic beam. This makes the approach applicable to any polar molecule that one can produce with sufficient density in a pulsed beam. Upon arrival above the chip, the molecules are confined in miniaturized electric field traps that move along with the molecular beam at a velocity of several hundred meters per second [48,49]. A whole array of these moving traps is subsequently brought to a complete standstill over a distance of only a few centimetres. After a certain holding time, *e.g.* after the experiments with the trapped molecules on the chip are over, the molecules are accelerated off the chip again. In this way, sensitive detection schemes that have been developed over the years for molecules in free flight in a molecular beam can be used. This is a new loading and detection methodology for molecules on a chip that is applicable to a wide variety of species [50].

2.2. Trapping molecules on a chip; the present status

The operation principle of the Stark decelerator on a chip – the main tool that is used to bring the molecules onto the chip as well as to bring them to the detector afterwards – has been explained in detail elsewhere [49], and only a brief description will be given here. Its operation relies on the superposition of electric fields created by the electrodes on the chip. When two dipolar fields with different length scales and opposite directions are superimposed, a minimum of the electric field strength is created. This minimum is located at the point where the long-range dipole that dominates far from the surface is cancelled by the short-range dipole that dominates close to the surface. Such a minimum of the electric field strength presents a trap for polar molecules that are in a low field seeking quantum state. When an array of electrodes is used, an array of electric field minima can be created above the surface. The Stark

decelerator on a chip that we have used thus far [48-50] and that is shown in Figure 1 consists of an extended array of equidistant parallel electrodes with a length of 4 mm arranged on a flat support. The electrodes have a width of 10 μm with a centre-to-centre separation of neighbouring electrodes of 40 μm . This structure is periodically extended over about 50 mm. Each electrode is electrically connected to the electrodes that are (multiples of) six positions further. By applying the appropriate potentials, tubular minima of electric field strength with a diameter of about 20 μm are generated every 120 μm . It is noted that the two ends of the 4 mm long tubular minima are closed in the present design by the fringe fields near the ends of the electrodes. These electric field minima can be continuously moved over the full 50 mm length at a constant height of about 25 μm . The temporal variation that is required for each of the six potentials to keep the electric field minima at a constant height while moving at a constant velocity turns out to be harmonic. Three of the potentials can always be positive, the other three always negative, and within each polarity set the potentials need to be phase-shifted by 120° . Time variation of the potentials with a fixed frequency ν , which is in the MHz regime for our experiments, results in a movement of the minima at a speed given by $120 [\mu\text{m}] \cdot \nu [\text{MHz}]$ [48]. The operation principle of this “supersonic conveyor belt” is similar to that of a magnetic conveyor belt for atoms [51].

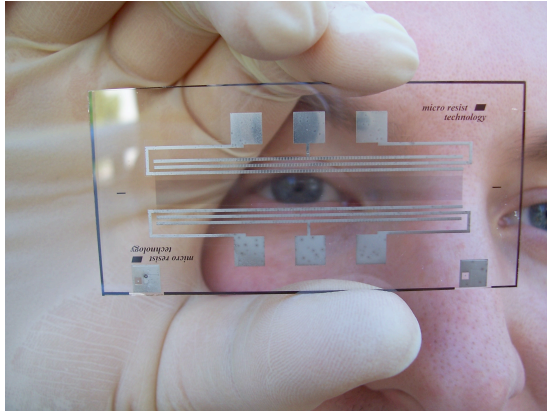


Figure 1: Photograph of the molecule chip, held by one of us (S.A.M.). The array of 1254, 10 μm diameter and 4 mm long electrodes extends over a total length of 50 mm. The electrodes run vertically, and are connected to either one of the six macroscopic pads to which the sinusoidal voltages are applied. (Photograph by Dr. Cynthia E. Heiner, FHI).

The proof-of-principle experiments have been performed with CO molecules in the low field seeking levels of the metastable $a^3\Pi_1$ ($v'=0$, $J'=1$) state. For these molecules the electric field minima correspond to traps with a depth of about 35 mK when a constant frequency ν is applied. When the frequency ν is changed linearly in time, i.e., when a constant acceleration is applied, the diameter and the depth of the traps decrease but three-dimensional confinement is maintained up to an acceleration of about $1.5 \cdot 10^6 \text{ m/s}^2$. Along the long axis of the trap, molecules can be lost due to nonadiabatic transitions to non-trappable degenerate states. In magnetic traps for atoms on a chip this hole at the centre of the trap is commonly plugged by adding a homogeneous magnetic field. An offset magnetic field, directed parallel to the long axis of the trap, could also be added in the present setup. For molecules in electric traps, however, there often exists the unique alternative solution to simply select an isotopologue with a favourable hyperfine level structure such that there is no degeneracy between trappable and non-trappable states in zero electric field [52]. The most abundant carbon monoxide isotopologue, $^{12}\text{C}^{16}\text{O}$, has no hyperfine structure and the low field seeking $M\Omega = -1$ level of the $a^3\Pi_1$ ($v'=0$, $J'=1$) state is degenerate with the $M = 0$ level in zero electric field, making this species susceptible to nonadiabatic transitions. In $^{13}\text{C}^{16}\text{O}$, however, the coupling of the nuclear spin of the ^{13}C nucleus with the orbital angular momentum results in a lifting of this degeneracy. The low field seeking levels never come closer to the non-trappable level than about 50 MHz in any electric field for ^{13}CO , effectively preventing non-adiabatic transitions to occur. Using ^{13}CO , we have demonstrated that we can bring a whole array of electric field minima containing polar molecules – directly loaded from a molecular beam – to a complete standstill on the chip, and that we can subsequently accelerate all the molecules off the chip again for sensitive detection [50]. In these experiments, the density of molecules in the moving traps on

the chip was about 10^7 cm^{-3} . By placing the chip closer to the source this density can be increased by two to three orders of magnitude, which is important for the future experiments discussed below.

3. Molecular physics experiments on a chip; future plans

An advantage of using metastable CO in the proof-of-principle experiments has been that these molecules can be readily detected upon impacting a gold surface, as then Auger electrons are released. To make the detection scheme more general – and presumably even more sensitive – we will use resonance enhanced multi-photon ionization (REMPI) with a pulsed laser in the future. When the laser is collimated to a 1 mm diameter beam and fires at the time that the molecules that are ejected off the chip are space-focused in the laser beam, the molecules from all traps on the chip will be detected simultaneously. Metastable CO can be detected via one-colour (1+1)-REMPI on the $b^3\Sigma^+ \leftarrow a^3\Pi$ transition around 283 nm. When the laser is focused (or restricted) to a 20 μm diameter beam and is positioned closely behind the chip, molecules coming from the individual traps on the chip can be detected one after the other. Once the ionization detection scheme has been tested on metastable CO and the position and timing of the laser have been optimized, experiments with ground-state molecules, in particular (various isotopologues of) ammonia and the hydroxyl radical will be pursued.

For metastable CO molecules there exists an alternative method to detect them while they are still on the chip; every metastable molecule will emit one photon in the 206-240 nm spectral region that can in principle be detected. When a UV-transparent objective is used together with a gateable image-intensified CCD camera system, two-dimensional images of the spatial distribution of the CO molecules in stationary traps on the chip can be recorded with a sub-micrometer spatial resolution. For molecules like OH, planar laser-induced fluorescence imaging can be pursued using the same experimental setup. For these experiments it is important to realize that the experiment is very reproducible and can be run at a high repetition rate, such that sufficient measurement statistics is obtained.

Once the loading and detection schemes for the molecules on the chip are optimized, a detailed study of the lifetime of the molecules in the stationary traps on the chip can be undertaken. The intrinsic molecular lifetime of both $^{13}\text{C}^{16}\text{O}$ and $^{13}\text{C}^{18}\text{O}$ in the $v'=0$ and $v'=1$ level of the $a^3\Pi_1$ state will be determined from the measurement of the signal on either the Auger detector or the ionization detector (whatever works best) as a function of the time that the molecules have been held in the stationary traps on the chip. These lifetimes are around 2.5 ms only, and will not be influenced by blackbody radiation or other trap-loss mechanisms. Comparing these lifetimes with the previously measured value for $^{12}\text{C}^{16}\text{O}$ [30] will shed light on the isotope effect in the coupling between the $a^3\Pi$ and the $^1\Pi$ electronically excited states in CO. This measurement will also demonstrate the dramatic reduction in the size of the required experimental setup between the measurements in 2007 and the new ones. After these measurements we will study the trapping time of ground-state OH radicals in the miniaturized electrostatic traps on the chip, using ionization detection after ejecting the molecules from the chip. To avoid nonadiabatic transitions, we will apply an offset magnetic field directed perpendicularly to the electric fields. The trapping time will be measured as a function of the distance to the surface and as a function of the temperature to study surface-induced heating of the polar molecules [53]. The temperature of the molecules trapped on the chip will be determined via microwave-UV double-resonance depletion experiments. For all the molecules mentioned thus far, transitions to non-trapped states (either to the other Λ -doublet component, to the other inversion component, or to another rotational state) can be induced using radiation in the GHz region. From the spectral shape of the microwave transition one can infer the spatial distribution of the molecules in the traps, and thereby their temperature. Depending on the outcome of these studies, we might decide to perform similar studies on other molecules trapped on a chip (NH ($a^1\Delta$), NH_3/ND_3) as well.

When the loading and detection methodology has been optimized and the lifetimes of the molecules understood, experiments can be performed with two different molecular species that are trapped simultaneously on the chip. For this, a chip could be designed that contains, for instance, two

independent decelerators that are displaced sideways, *i.e.* along the direction of the long axis of the tubular electric field minima, by a few mm. This chip can be loaded simultaneously from two opposite sides with different species. Arrays of electric field traps containing the molecules coming from either side can be brought to a standstill at predefined positions on the chip. There, an array of miniaturized electrostatic guides can be switched on, basically connecting the stationary tubular traps from one decelerator with the other. Additional offset electric or magnetic fields can be used to push the molecules to one end of the elongated trap on a millisecond time-scale, after which the electrostatic guides are switched off again. After the molecules are held for a certain time in the array of miniaturized reaction vessels on the chip, they can be accelerated and ejected off the chip again and space-focused in the detection region. Inelastic collisions and reactions can be monitored *via* the depletion of the signal as a function of interaction time.

An interesting advantage of the Stark decelerator on a chip is that the molecules that are confined in the tubular electric field minima are never exposed to fields that are higher than a few kV/cm. In these fields, several rotationally excited quantum states of heavy polar molecules are low field seeking, and can therefore be decelerated as well. In particular, the first rotationally excited $N''=1$ level of YbF ($X^2\Sigma^+$), that still has a significant population in a cooled molecular beam, has a component that is low field seeking up to 18 kV/cm, where the Stark shift has reached $+0.15\text{ cm}^{-1}$ [54]. It should be possible to remove about 50% of the kinetic energy from this heavy molecule above the 5 cm long chip when it enters with a velocity of 300 m/s. This offers an interesting alternative to AG deceleration schemes that have been applied to this species [55], even though the absolute number of molecules that can be decelerated above a chip might not yield sufficient statistics for subsequent precision experiments.

References:

- [1] G. Scoles, (ed.) *Atomic and Molecular Beam Methods*, Vol. 1 & 2, (Oxford Univ. Press, New York, 1988 & 1992).
- [2] W. Gerlach and O. Stern, *Z. für Phys.* **9**, 349-352, (1922).
- [3] I.I. Rabi, S. Millman, P. Kusch, and J.R. Zacharias, *Phys. Rev.* **55**, 526-535, (1939).
- [4] J.P. Gordon, H.J. Zeiger, and C.H. Townes, *Phys. Rev.* **95**, 282-284, (1954); *ibid* **99**, 1264-1274, (1955).
- [5] H.L. Bethlem, G. Berden, and G. Meijer, *Phys. Rev. Lett.* **83**, 1558-1561, (1999).
- [6] H.L. Bethlem, G. Berden, F.M.H. Crompvoets, R.T. Jongma, A.J.A. van Roij, and G. Meijer, *Nature* **406**, 491-494, (2000).
- [7] J. van Veldhoven, H.L. Bethlem, and G. Meijer, *Phys. Rev. Lett.* **94**, 083001, (2005).
- [8] F.M.H. Crompvoets, H.L. Bethlem, R.T. Jongma, and G. Meijer, *Nature* **411**, 174-176, (2001).
- [9] C.E. Heiner, D. Carty, G. Meijer, and H.L. Bethlem, *Nature Physics* **3**, 115-118, (2007).
- [10] S.Y.T. van de Meerakker, P.H.M. Smeets, N. Vanhaecke, R.T. Jongma, and G. Meijer, *Phys. Rev. Lett.* **94**, 023004, (2005).
- [11] B.C. Sawyer, B.K. Stuhl, D. Wang, M. Yeo, and J. Ye, *Phys. Rev. Lett.* **101**, 203203, (2008).
- [12] B.C. Sawyer, B.L. Lev, E.R. Hudson, B.K. Stuhl, M. Lara, J.L. Bohn, and J. Ye, *Phys. Rev. Lett.* **98**, 253002, (2007).
- [13] N. Vanhaecke, U. Meier, M. Andrist, B.H. Meier, and F. Merkt, *Phys. Rev. A* **75**, 031402, (2007).
- [14] S.D. Hogan, D. Sprecher, M. Andrist, N. Vanhaecke, and F. Merkt, *Phys. Rev. A* **76**, 023412, (2007).
- [15] E. Narevicius, A. Libson, C.G. Parthey, I. Chavez, J. Narevicius, U. Even, and M.G. Raizen, *Phys. Rev. Lett.* **100**, 093003, (2008); *ibid A* **77**, 051401(R), (2008).
- [16] N. Balakrishnan and A. Dalgarno, *J. Chem. Phys.* **113**, 621-627, (2000).
- [17] E. Bodo and F.A. Gianturco, *Int. Rev. Phys. Chem.* **25**, 313-351, (2006).
- [18] P. S. Zuchowski and J.M. Hutson, *Phys. Rev. A* **79**, 062708, (2009).
- [19] J.J. Gilijamse, S. Hoekstra, S.Y.T. van de Meerakker, G.C. Groenenboom, and G. Meijer, *Science* **313**, 1617-1620, (2006).
- [20] J.J. Hudson, B.E. Sauer, M.R. Tarbutt, and E.A. Hinds, *Phys. Rev. Lett.* **89**, 023003, (2002).
- [21] D. Kawall, F. Bay, S. Bickman, Y. Jiang, and D. DeMille, *Phys. Rev. Lett.* **92**, 133007 (2004).
- [22] N.E. Shafer-Ray, *Phys. Rev. A* **73**, 034102 (2006).

- [23] C. Daussy, T. Marrel, A. Amy-Klein, C.T. Nguyen, Ch. J. Bordé, and Ch. Chardonnet, *Phys. Rev. Lett.* **83**, 1554-1557, (1999).
- [24] E. Reinhold, R. Buning, U. Hollenstein, A. Ivanchik, P. Petitjean, and W. Ubachs, *Phys. Rev. Lett.* **96**, 151101, (2006).
- [25] A. Shelkovnikov, R.J. Butcher, Ch. Chardonnet, and A. Amy-Klein, *Phys. Rev. Lett.* **100**, 150801, (2008).
- [26] J. van Veldhoven, J. Küpper, H.L. Bethlem, B. Sartakov, A.J.A. van Roij, and G. Meijer, *Eur. Phys. J. D* **31**, 337-349, (2004).
- [27] E.R. Hudson, H.J. Lewandowski, B.C. Sawyer, and J. Ye, *Phys. Rev. Lett.* **96**, 143004, (2006).
- [28] M.R. Tarbutt, J.J. Hudson, B.E. Sauer, and E.A. Hinds, *Faraday Discuss.* **142**, 37-56, (2009).
- [29] S.Y.T. van de Meerakker, N. Vanhaecke, M.P.J. van der Loo, G.C. Groenenboom, and G. Meijer, *Phys. Rev. Lett.* **95**, 013003, (2005).
- [30] J.J. Gilijamse, S. Hoekstra, S.A. Meek, M. Metsälä, S.Y.T. van de Meerakker, G. Meijer, and G.C. Groenenboom, *J. Chem. Phys.* **127**, 221102, (2007).
- [31] J.D. Weinstein, R. deCarvalho, T. Guillet, B. Friedrich, and J.M. Doyle, *Nature* **395**, 148-150, (1998).
- [32] W.C. Campbell, G.C. Groenenboom, H.-I. Lu, E. Tsikata, and J.M. Doyle, *Phys. Rev. Lett.* **100**, 083003, (2008).
- [33] J. Fortágh and C. Zimmermann, *Rev. Mod. Phys.* **79**, 235-289, (2007), and references therein.
- [34] W. Hänsel, P. Hommelhoff, T.W. Hänsch, and J. Reichel, *Nature* **413**, 498-501, (2001).
- [35] T. Schumm, S. Hofferberth, L.M. Andersson, S. Wildermuth, S. Groth, I. Bar-Joseph, J. Schmiedmayer, and P. Krüger, *Nature Physics* **1**, 57-62, (2005).
- [36] W. Ertmer and E.M. Rasel for the QUANTUS-Team, *Nucl. Phys. B (Proc. Suppl.)* **166**, 307-309, (2007).
- [37] J.D. Weinstein and K.G. Libbrecht, *Phys. Rev. A* **52**, 4004-4009, (1995).
- [38] E.A. Hinds and I.G. Hughes, *J. Phys. D: Appl. Phys.* **32**, R119-R146, (1999).
- [39] A. André, D. DeMille, J.M. Doyle, M.D. Lukin, S.E. Maxwell, P. Rabl, R.J. Schoelkopf, and P. Zoller, *Nature Physics* **2**, 636-642, (2006).
- [40] D. DeMille, *Phys. Rev. Lett.* **88**, 067901, (2002).
- [41] S.Y.T. van de Meerakker, H.L. Bethlem, and G. Meijer, *Nature Physics* **4**, 595-602, (2008).
- [42] J. Deiglmayr, A. Grochola, M. Repp, K. Mörtlbauer, C. Glück, J. Lange, O. Dulieu, R. Wester, and M. Weidemüller, *Phys. Rev. Lett.* **101**, 133004, (2008).
- [43] K.K. Ni, S. Ospelkaus, M.H.G. de Miranda, A. Pe'er, B. Neyenhuis, J.J. Zirbel, S. Kotochigova, P.S. Julienne, D.S. Jin, and J. Ye, *Science* **322**, 231-235, (2008).
- [44] M. Wilzbach, D. Heine, S. Groth, X. Liu, B. Hessmo, J. Schmiedmayer, *Opt. Lett.* **34**, 259-261, (2009).
- [45] A. Stibor, S. Kraft, T. Campey, D. Komma, A. Günther, J. Fortágh, C.J. Vale, H. Rubinsztein-Dunlop, and C. Zimmermann, *Phys. Rev. A* **76**, 033614, (2007).
- [46] M. Trupke, J. Goldwin, B. Darquié, G. Dutier, S. Eriksson, J. Ashmore, and E.A. Hinds, *Phys. Rev. Lett.* **99**, 063601, (2007).
- [47] Y. Colombe, T. Steinmetz, G. Dubois, F. Linke, D. Hunger, and J. Reichel, *Nature* **450**, 272-276, (2007).
- [48] S. A. Meek, H.L. Bethlem, H. Conrad, and G. Meijer, *Phys. Rev. Lett.* **100**, 153003, (2008).
- [49] S.A. Meek, H. Conrad, and G. Meijer, *New J. Phys.* **11**, 055024, (2009).
- [50] S.A. Meek, H. Conrad, and G. Meijer, *Science* **324**, 1699, (2009).
- [51] W. Hansel, J. Reichel, P. Hommelhoff, and T.W. Hänsch, *Phys. Rev. Lett.* **86**, 608-611, (2001).
- [52] M. Kirste, B.G. Sartakov, M. Schnell, and G. Meijer, *Phys. Rev. A* **79**, 051401(R) (2009).
- [53] S.Y. Buhmann, M.R. Tarbutt, S. Scheel, and E.A. Hinds, *Phys. Rev. A* **78**, 052901, (2008).
- [54] M.R. Tarbutt, J.J. Hudson, B.E. Sauer, and E.A. Hinds, in: *Cold Molecules: Theory, Experiment, Applications* (eds.: R.V. Krems, W.C. Stwalley, B. Friedrich) p. 555-596, CRC Press (2009).
- [55] M.R. Tarbutt, H.L. Bethlem, J.J. Hudson, V.L. Ryabov, V.A. Ryzhov, B.E. Sauer, G. Meijer, and E.A. Hinds, *Phys. Rev. Lett.* **92**, 173002, (2004).