Experiments with Stark decelerated and trapped OH radicals

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Abstract.   In this paper, we review recent developments in our efforts to get complete control over the velocity of a molecular beam. Central in these experiments is the use of a Stark decelerator, with which the velocity of a beam of neutral polar molecules can be continuously varied, using techniques akin to methods used in charged particle accelerator physics. These techniques allow for confining isolated gas-phase molecules in traps, but can also be used to advantage in a variety of molecular beam experiments where the velocity of the beam is an important parameter. Particularly, molecular beam inelastic scattering experiments can be performed with a variable collision energy and with a high intrinsic energy resolution.

1. Introduction

Over the last years, we have been exploring experimental schemes to achieve full control over both the internal and external degrees of freedom of neutral gas-phase molecules. In particular, we have developed schemes to accelerate or slow down beams of neutral polar molecules to any arbitrary value by the use of time-varying inhomogeneous electric fields [1]. When the molecules are decelerated to standstill, they can be confined in traps for times up to seconds [2, 3]. In these traps, the molecular packet has a temperature on the order of 10 mK. Electric fields are used to keep the molecules confined in a region of space where they can be studied in isolation from the (hot) environment. These experiments are of special interest to the growing cold molecules-community, as they represent one of the few techniques that have resulted in the trapping of neutral ground-state molecules. Interest in cold molecules stems from intriguing new physics and chemistry that can be studied in these systems, ranging from the observations of scattering resonances at low collision energies [4, 5, 6] to the observation of quantum degeneracy. Particularly interesting would be the study of dipole-dipole interactions in these quantum gases and the possibility to control the inter-particle interactions with external electromagnetic fields [7].

Central in these experiments is the use of a so-called Stark decelerator. A Stark decelerator is based on the notion that a polar molecule in a quantum state in which the dipole moment is anti-parallel to an external electric field will be attracted to regions of low electric field. Therefore, molecules in such a "low-field" seeking state will be decelerated on their way from a region of low electric field into a region of high electric field. If the electric field is switched off while the molecules are still in the region of high electric field, the molecules will not regain their lost kinetic energy. In a Stark decelerator, this process is repeated until the average velocity of the molecules is reduced to any arbitrary velocity [8].

In our experiments we use pulsed molecular beams to obtain a maximum initial density of state-selected molecules at a minimum initial temperature. A properly timed switching of the electric fields ensures that a bunch of molecules can be kept together in the forward direction ("phase stability") throughout the deceleration process [9, 10]. Transverse stability is achieved by using an electrode geometry that produces a minimum of electric field on the molecular beam axis, thereby continuously focusing the beam. The Stark deceleration process can thus be seen as slicing a packet of molecules with a narrow velocity distribution out of the molecular beam pulse. This packet can then be decelerated or accelerated to any velocity, maintaining the narrow velocity distribution and the particle density in the packet.

Although the forces that can thus be exerted on neutral polar particles are many orders of magnitude smaller than those exerted on charged particles, Stark effect ma-

nipulation allows one to perform all the operations on neutral polar molecules that are successfully used throughout on charged particles. In our group, this has been explicitly demonstrated by the construction of various types of decelerators [8, 11], a buncher [12], various types of traps [2, 13], and a storage ring for neutral polar molecules [14]. Recently, also a molecular synchrotron has been experimentally realized [15]. A Stark decelerator is operational or under construction in various groups, and to date, successful Stark deceleration has been demonstrated for the molecules CO^* [8], ND_3 [2], NH_3 [10], OH [3, 16], OD [17], H_2CO [18], NH^* [19], and SO_2 [20]. Of these, the molecules ND_3 [2], OH [3] and OD [17] have been trapped.

To be able to exploit the possibilities that this new technology offers, we have constructed in recent years a new generation molecular beam deceleration machine, that is designed such that a large fraction of the molecular beam pulse can be slowed down and trapped. This machine has been mostly used to perform experiments with Stark decelerated and/or trapped beams of OH. The role of the omnipresent OH radical as intermediate in many chemical reactions has made this a benchmark molecule in collision and reactive scattering studies. Together with its amenability to the Stark deceleration process, this makes the OH radical one of the seminal candidates for Stark deceleration experiments. In this paper, we briefly review the most important results of our Stark deceleration and trapping experiments using beams of OH radicals. For a more detailed description of the experiments and the results, the interested reader is referred to the existing literature.

2. The Stark decelerator

A schematic representation of the deceleration and trapping machine is shown in Figure 1. A detailed description of the experiment is given in [21]. Briefly, a pulsed beam of OH radicals is produced via ArF-laser dissociation of HNO_3 near the orifice of a pulsed valve. Either Kr or Xe is used as a carrier gas to produce a beam with a low initial velocity. After the supersonic expansion, most OH radicals reside in the $J = 3/2$ rovibrational ground state of the $X\,^2\Pi_{3/2}$ electronic ground state. The beam passes though a 2 mm diameter skimmer and enters a small hexapole that focusses the beam into the Stark decelerator. Only OH radicals in the upper Λ-doublet component of f-parity are focussed and are of relevance to the experiments. The Stark decelerator consists of 108 electric field stages, with a center-to-center distance of 11 mm, and with a 4×4 mm^2 transverse acceptance area. To slow down the beam, a pulsed voltage difference of 40 kV is applied to the electrodes. The OH radicals are detected by imaging laser induced fluorescence (LIF) onto a photomultiplier tube (PMT).

The operation principle of the Stark decelerator can best be illustrated by the time-of-flight (TOF) profile of OH radicals exiting the decelerator that is measured for
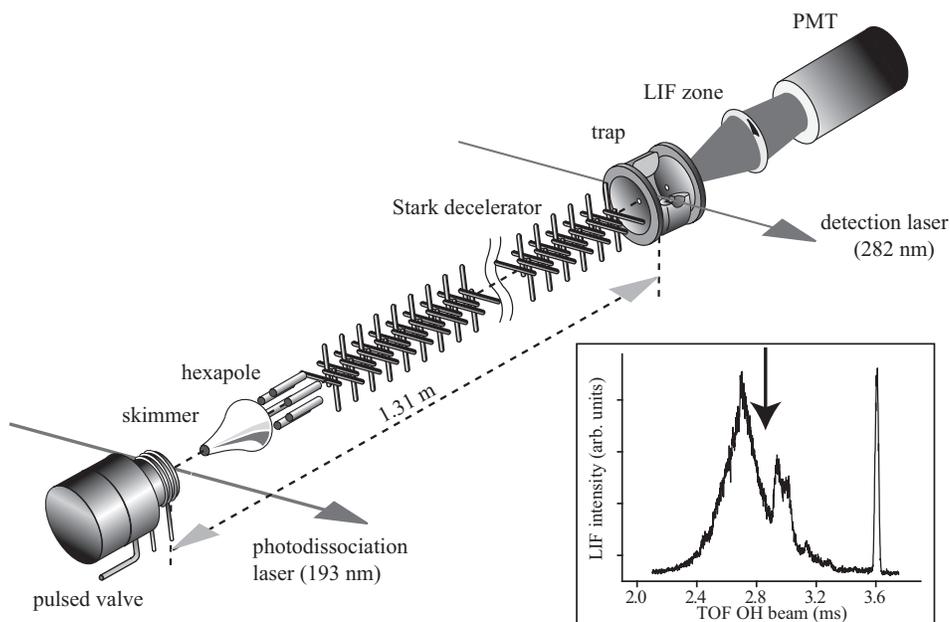
Figure 1. Scheme of the experimental setup. A pulsed beam of OH is produced via photodissociation of HNO_3 seeded in Xe. The beam passes through a skimmer, hexapole and Stark decelerator. The OH radicals can be brought to a standstill and confined in an electrostatic quadrupole trap. Molecules in the trap are state-selectively detected by imaging the laser induced fluorescence onto a PMT. In the inset, a typical time-of-flight profile is shown of OH radicals that exit the decelerator. The hole in the distribution due to the removal of molecules that are decelerated is indicated by the vertical arrow.

a typical setting of the decelerator (see Figure 1). The densest part of the OH beam, with an original velocity of 450 m/s, is selected and slowed down to a final velocity of 281 m/s. The hole in the TOF profile of the fast beam due to the removal of OH radicals that are decelerated is indicated by an arrow. It is noted that the final velocity of the packet is determined exclusively by the (computer controlled) sequence of switch times that is applied to the decelerator, and can be varied between standstill and conventional beam velocities.

3. Trapping of OH radicals

In principle, the selected packet of molecules can be decelerated to a standstill inside a Stark decelerator. However, for confinement of the cold cloud a trap is required. For this, the decelerator is extended with an electrostatic quadrupole trap. The trap consists of a ring electrode and two hyperbolic end caps. To be able to load the OH radicals into the trap, the beam needs to be decelerated to approximately 20 m/s. When this slow packet of molecules exits the decelerator, voltages are applied to the trap electrodes that create a last potential hill for the molecules. The parameters can be chosen such that the packet comes to a standstill near the center of the trap. At this time, the trap voltages are reconfigured to produce a (nearly) symmetric 500 mK deep potential well in which

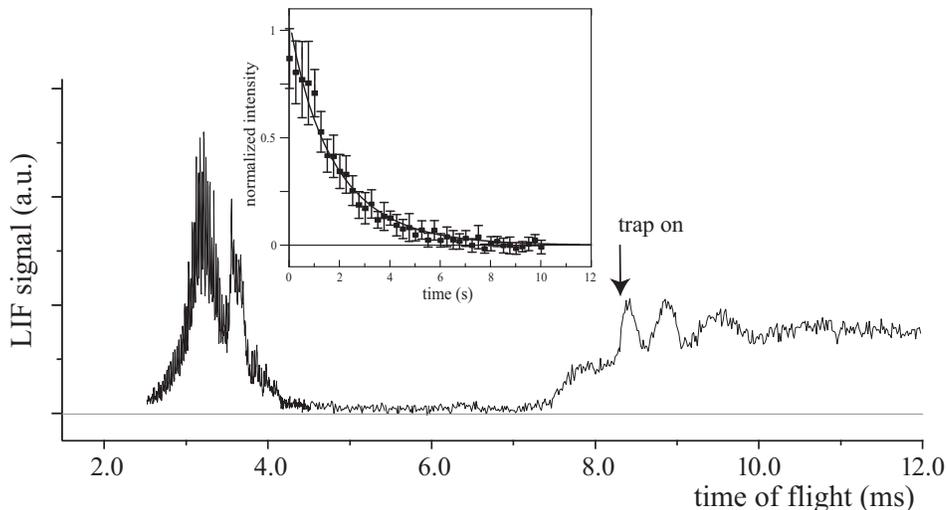the molecules are confined. Figure 2 shows a typical observed TOF for a deceleration

Figure 2. LIF signal of OH radicals at the center of the quadrupole trap as a function of time after production for a deceleration and trapping sequence. The time at which the trap is switched on is indicated by a vertical arrow. In the inset, the signal of the trapped OH radicals is shown on a longer timescale.

and trapping experiment. The OH radicals come to a standstill approximately 8.3 ms after their production. At that time, indicated by the arrow, the trap is switched into the confining geometry. After some initial oscillations, a steady LIF signal is observed from the OH radicals in the trap. The inset of Figure 2 shows the LIF signal intensity on a longer timescale. From these experiments, a $1/e$ trap lifetime on the order of 1.5 s is deduced. Depending on the details of the trap-loading sequence, the density of the trapped cloud is about 10^7 radicals cm^{-3}, and the temperature can be as low as 50 mK [3].

The experimental ability to store neutral molecules, isolated in the gas phase, in a trap opens up the possibility for a variety of experiments. The trapping technology has matured now sufficiently to start exploiting these possibilities to study the molecules and their interactions in detail. For instance, the radiative lifetime of long-lived excited states can be measured directly by monitoring the temporal decay of the population in the trap. We have demonstrated this for vibrationally excited OH ($X\,^2\Pi_{3/2}, v = 1, J = 3/2$) radicals, and measured a radiative lifetime of 59.0 ± 2.0 ms [22]. This measurement is in excellent agreement with calculations, and benchmarks the Einstein A coefficients in the important Meinel system of OH. It is also possible to use the signal of the trapped molecules to automatically optimize the Stark deceleration and trap loading process using evolutionary strategies [23]. Most recently, we studied the interaction of trapped OH radicals with room temperature blackbody radiation. By comparing the trapping times of OH and OD radicals, the individual contribution of blackbody radiation and collisions with background gas could be quantified [17].

4. Collision experiments

The long-term goal of these trapping experiments is to reach densities that are high enough and temperatures that are low enough to study collective quantum effects in these cold gases. But also in more conventional molecular beam experiments a Stark decelerator can add interesting new possibilities. In fact, all beam experiments in which the velocity (distribution) of the beam is an important parameter can benefit from using a Stark decelerator. Arguably one of the most interesting of these is to exploit the velocity tunability of the beam to perform scattering experiments as a function of the continuously variable collision energy. The low velocity spread of Stark decelerated beams enable high intrinsic energy resolutions. In addition, the state purity of the bunches of selected molecules that emerge from the decelerator is close to 100 %.

The applicability to use Stark decelerated beams in crossed beam experiments was demonstrated recently by scattering a Stark decelerated beam of OH ($X\,^2\Pi_{3/2}, v = 0, J = 3/2, f$) radicals with a conventional beam of Xe atoms at right angles [24]. The velocity of the OH radicals was varied from 33 to 700 m/s. The total collision energy could therefore be varied in the 50 to 400 cm^{-1} range, with a nearly constant energy resolution of 13 cm^{-1}. Particularly interesting in this experiment is that the collision energy could be varied over the energetic thresholds for inelastic scattering into the excited rotational levels.

In Figure 3 the measured relative inelastic cross sections as a function of the collision energy are shown for scattering into 4 inelastic channels. The highest cross section is observed for scattering into the ($X\,^2\Pi_{3/2}, J = 3/2, e$) state. This Λ-doublet changing collision is the only exo-energetic channel, and the relative cross section for this channel approaches 100 % at low collision energies. The other channels show a clear threshold behavior. These measurements provide a very sensitive probe for theoretical potential energy surfaces, from which a detailed understanding of the collision dynamics can be obtained. The solid curves that are shown in Figure 3 result from the quantum scattering calculations that Gerrit Groenenboom at the Radboud University Nijmegen in the Netherlands performed independently from the experiment, and show an excellent agreement with the experiment.

5. Conclusions and outlook

The Stark decelerator is a versatile new tool in molecular beam technology that enables exquisite control over the velocity(distribution) of the beam. Molecular beam velocities ranging from standstill to a few hundred meters per second can be dialed in and scanned similar to the variation of the wavelength of a laser. This allows for confining molecules in traps, but can also add new dimensions to for instance crossed molecular beam scattering experiments. The densities that can ultimately be reached in these beams
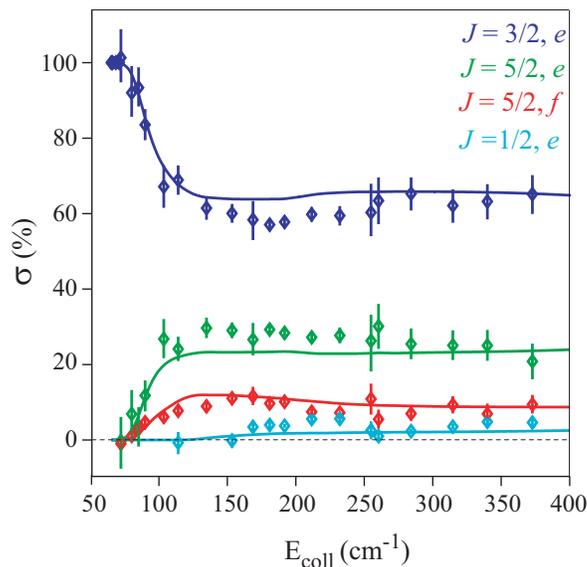
Figure 3. Relative inelastic cross sections for scattering of OH ($X\,^2\Pi_{3/2}, J = 3/2, f$) radicals with Xe atoms as a function of the collision energy. Collisions populating the ($X\,^2\Pi_{3/2}, J = 3/2, e$), ($X\,^2\Pi_{3/2}, J = 5/2, e$), ($X\,^2\Pi_{3/2}, J = 5/2, f$), and ($X\,^2\Pi_{1/2}, J = 1/2, e$) states were studied.

and/or in the trap suffice to study interactions between the molecules.

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