Chapter 20
Manipulation and Control of Molecular Beams: The Development of the Stark-Decelerator

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Abstract State-selective manipulation of beams of atoms and molecules with electric and magnetic fields has been crucial for the success of the field of molecular beams. Originally, this manipulation only involved the *transverse* motion. In this Chapter, the development of the Stark-decelerator, that allows to also manipulate and control the *longitudinal* motion of molecules in a beam, is presented.

1 Introduction

"*Born in leaks, the original sin of vacuum technology, molecular beams are collimated wisps of molecules traversing the chambered void that is their theatre [...]. On stage for only milliseconds between their entrances and exits, they have captivated an ever growing audience by the variety and range of their repertoire*". This is how John B. Fenn affectionately phrased it over 30 years ago, when he reflected on the long and rich history of molecular beams in his foreword to one of the classic books on this subject [1]. He could not have foreseen the spectacular leap forward that the level of control over molecular beams would take. In particular, methods that have been developed since then to slow down and store molecular beams – thereby stretching the duration of their performance on stage by orders of magnitude – have made whole new classes of experiments possible.

The motion of neutral molecules in a beam can be manipulated and controlled with inhomogeneous electric and magnetic fields. Static fields can be used to deflect or focus molecules, whereas time-varying fields can be used to decelerate or accelerate beams of molecules to any desired velocity. In this paper we present an historical overview, emphasizing the important role of molecular beam deflection and focusing experiments in the development and testing of quantum mechanics. We describe the original attempts and the successful implementation of schemes to decelerate and accelerate molecular beams with electric fields, that is, the development of the Stark-decelerator. The various elements, using electric as well as magnetic fields, that have

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been developed for the manipulation and control of molecular beams since the first successful demonstration of the Stark-decelerator in 1998, have resulted in setups in which the molecules can be stored in stationary traps or injected in a molecular storage ring or synchrotron, for instance. Novel crossed-beam scattering studies at low collision energies, high-resolution spectroscopy studies on trapped or slow molecules and lifetime measurements of trapped molecules have become possible [2].

2 Deflection and Focusing of Molecular Beams

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 methods exist to perform sensitive and quantum state selective detection of the atoms and molecules in the beams. In the early days, such detection methods were lacking and the particles in the beam were detected, for instance, by a "hot wire" (Langmuir-Taylor) detector, by electron-impact ionization or by deposition and *ex-situ* investigation of the particles on a substrate at the end of the beam-machine. To achieve quantum state selectivity in the overall detection process, these methods were combined with inhomogeneous electric and magnetic field sections to influence the trajectories of the particles on their way to the detector.

The first paper discussing the degree of deflection for a beam of polar molecules passing through an inhomogeneous electric field was submitted almost a century ago, at the end of July 1921, to the *Zeitschrift für Physik* [3]. The paper was written by Hartmut Kallmann and Fritz Reiche, coworkers of Fritz Haber at the Kaiser Wilhelm Institute for Physical Chemistry and Electrochemistry in Berlin, the present Fritz-Haber-Institut der Max-Planck-Gesellschaft. Kallmann and Reiche write in their paper that they performed their analysis in support of experiments that "are ongoing at the institute" to determine whether the dipole moment is a property of an individual molecule or whether this is only induced in the molecule when it is in close proximity to other molecules, an issue that was intensively debated at that time. By passing a molecular beam, that is, a dilute but highly collimated sample of molecules, through an inhomogeneous electric field, they argued, it should be possible to monitor the deflection of the molecules and to thus determine the value of their dipole moment – provided they have one. In the introduction of their paper, they discuss in general terms the forces on a moving dipolar molecule due to inhomogeneous electric fields as well as due to inhomogeneous magnetic fields. In their analysis, however, they restrict themselves to the electric field case, and assume that the dipole moment of the diatomic molecule is perpendicular to the angular momentum vector, i.e., that the component of angular momentum along the internuclear axis is zero.

Shortly after its submission, the paper by Kallmann and Reiche came to the eyes of Otto Stern, pressing him to write up the theory behind an experiment that – as he

wrote – "he had been involved in for some time with his colleague Walther Gerlach". Stern submitted his paper only about one month later, at the end of August 1921, to the same journal. In a footnote to his paper, he explicitly acknowledges that the reason for publishing his paper is the upcoming paper of Kallmann and Reiche, and concludes that both papers are nicely complementary as his paper "discusses the case in which the dipole moment is parallel to the angular momentum vector, as is generally the case for magnetic atoms" [4]. In his paper, Stern describes a method to experimentally test space quantization via measuring the deflection of a beam of atoms with a magnetic moment when moving through an inhomogeneous magnetic field [4]. No further account on the early electric deflection experiment in Berlin is to be found in the literature or in the archives, while Stern and Gerlach performed their famous experiment within one year [5], in February of 1922. Electric deflection of a beam of polar molecules was first demonstrated by Erwin Wrede, a graduate student of Stern, several years later in Hamburg [6]. It is interesting to note that up to our re-discovery of the article by Kallmann and Reiche in 2009, this article went largely unnoticed, being cited only seven times; the important mentioning of the article by Otto Stern in a footnote in his paper could not include the final reference yet. Since then, it has been cited more than once per year.

All the original experimental geometries were devised to create strong magnetic or electric field gradients to efficiently deflect particles from the beam axis. In 1939, Isidor Rabi introduced the molecular beam magnetic resonance method by using two magnets in succession to produce inhomogeneous magnetic fields of oppositely directed gradients. In this set-up the deflection of particles caused by the first magnet is compensated by the second magnet, such that the particles are directed on a sigmoid path to the detector. A transition to "other states of space quantization" induced in between the magnet sections can be detected via the resulting reduction of the detector signal. This provided a new method to accurately measure nuclear or other magnetic moments [7]. Later, both magnetic [8, 9] and electric [10] field geometries were designed to focus particles in selected quantum states onto the detector. An electrostatic quadrupole focuser, i.e., an arrangement of four cylindrical electrodes with alternating positive and negative voltages, was used to couple a beam of ammonia molecules into a microwave cavity. Such an electrostatic quadrupole lens focuses ammonia molecules that are in the so-called low-field seeking, upper level of the inversion doublet while it simultaneously defocuses those that are in the lower, high-field seeking, level. The inverted population distribution of the ammonia molecules that is thus produced in the microwave cavity led to the invention of the maser by Gordon, Zeiger and Townes in 1954–1955 [11, 12]. Apart from the spectacular observation of the amplification of microwaves by stimulated emission, these focusing elements more generally enabled the recording, with high resolution and good sensitivity, of microwave spectra in a molecular beam. 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 were exploited to study steric effects, that is, to study how the orientation of an attacking molecule affects its reactivity [13]. Variants of the molecular beam resonance methods as well as scatter-

ing machines that employed state-selectors 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.

3 Early Attempts to Decelerate or Accelerate Molecular Beams

The state-selective 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. In his autobiography, Norman Ramsey, who himself later invented the separated oscillatory fields method and wrote a very influential book on molecular beam methods [14], recalls that Rabi was rather discouraged about the future of molecular beam research when he arrived in Rabi's lab in 1937, and that this discouragement only vanished when Rabi invented the molecular beam magnetic resonance method [15]. However, even though the manipulation of beams of molecules with external fields has been used extensively and with great success in the past, this manipulation exclusively involved the *transverse* motion of the molecules.

When the velocity distribution in a molecular beam is rather broad, the state-selective deflection fields can be used to provide some velocity selection. Offset or angled molecular beam geometries have been used, in which deflection fields cause only the slow (fast) atoms or molecules to obtain sufficiently large (small) deflections to pass through the apparatus and to reach the detector, for instance. This approach has been attempted to selectively load slow ammonia molecules in a microwave cavity to produce a maser with an ultra-narrow linewidth [16]. At Bell telephone laboratories an electrostatic parabolic reflector was designed to selectively couple slow ammonia molecules in the microwave cavity, that is, after deflecting them by 180 degrees [17]. These approaches suffered from the deficiency that, as stated then, "it is generally known that the velocity distribution of molecules emanating from a hole in a box is not Maxwellian, but departs from it by having fewer low velocity molecules", possibly caused by collisions with fast molecules "from behind" [17].

At the end of the nineteen-fifties, alternative approaches, with electric fields designed such as to create "multiple retardation barriers" have been proposed to actively manipulate the *longitudinal* motion of molecules in a beam, that is, to slow down ammonia molecules [18]. A rather compact experimental setup consisting out of a source chamber, a deceleration chamber and a slow-molecule deflection and detection chamber was constructed for exactly this purpose in the physics department of MIT, under the supervision of John G. King [19]. The approximately 20 cm long decelerator consisted of a linear array of ten parallel plate capacitors, capable of maintaining a voltage difference of 30 kV across 1 mm plate separation. Ammonia molecules in the low-field seeking, upper level of the $J = K = 1$ inversion doublet

lose kinetic energy when entering the high electric field region of the capacitor. When the electric field is slowly turned to zero while the molecules are in the homogeneous electric field inside the capacitor, the molecules do not regain the lost kinetic energy when exiting the capacitor and the process can be repeated. The experiments were performed using continuous beams and the same high voltage was applied to the whole array of electrodes in the form of a sine wave with a fixed frequency of 6 kHz. Therefore, the distances between the adjacent parallel plate capacitors as well as their lengths needed to be made such that it takes the ever slower molecules always exactly the same amount of time to reach the next stage, i.e., these distances and lengths needed to gradually decrease along the molecular beam. The machine was designed to slow down ammonia molecules from an initial speed of 200 m/s to a final speed of 35 m/s, selectively detecting the slowed molecules at the exit. The project suffered from the same deficiency as mentioned earlier, namely that there were not enough ammonia molecules at the initial speed of 200 m/s to yield a detectable signal. The work is described in detail in the Ph.D. thesis of Robert Golub [20], but no further publication has resulted from this work.

In the physical chemistry community the experimental efforts of Lennard Wharton, to demonstrate electric field *acceleration* of a molecular beam, are much better known. In the mid nineteen-sixties, at the University of Chicago, he constructed a molecular beam machine, containing a thirty-three foot long accelerator. The accelerator consisted of an array of 600 acceleration stages intended to increase the kinetic energy of LiF molecules in high-field seeking states from 0.2 eV to 2.0 eV, that is, speeding the LiF molecules up from 1200 m/s to 3800 m/s, with the aim to use these high energy beams for reactive scattering studies. Each acceleration stage consisted of two hemispherically ended rods with a diameter of 0.5 mm, spaced 0.5 mm apart. The beam was transversely kept together using additional alternate-gradient lenses. A popular scientific account of this work, together with a schematic drawing of the acceleration principle and a photograph of the "Chemical Accelerator", appeared in *Scientific American* in 1968 [21]. The photograph of the about eleven meter long machine is reproduced in Fig. 1. Also in this case, continuous molecular beams were used and the high voltage was applied as a sine wave with a fixed frequency to the array of electrodes, implying that the adjacent electric field stages needed to be put ever further apart to compensate for the molecules being ever faster, explaining the length of the molecular beam machine. An excellent paper, in which the focusing of beams of polar molecules in high-field seeking states was theoretically analyzed, resulted from this work [22]. The acceleration experiment was not successful, however, not only because the alignment of the array of electrodes is very critical in an alternate-gradient setup but also due to an overly optimistic view on the magnitude of the electric fields that could be stably obtained when designing the accelerator module. This is what the Ph.D. student who was working on this project, Edward A. Bromberg, concluded in his Ph.D. thesis in which he summarised that "it has not been possible to show either that particles have been accelerated, or that neutral particles cannot be accelerated" [23]. Both, the deceleration experiments of John King and the acceleration experiments of Lennard Wharton were not continued after the Ph.D. students completed their theses. Whereas interest in slow molecules as a maser
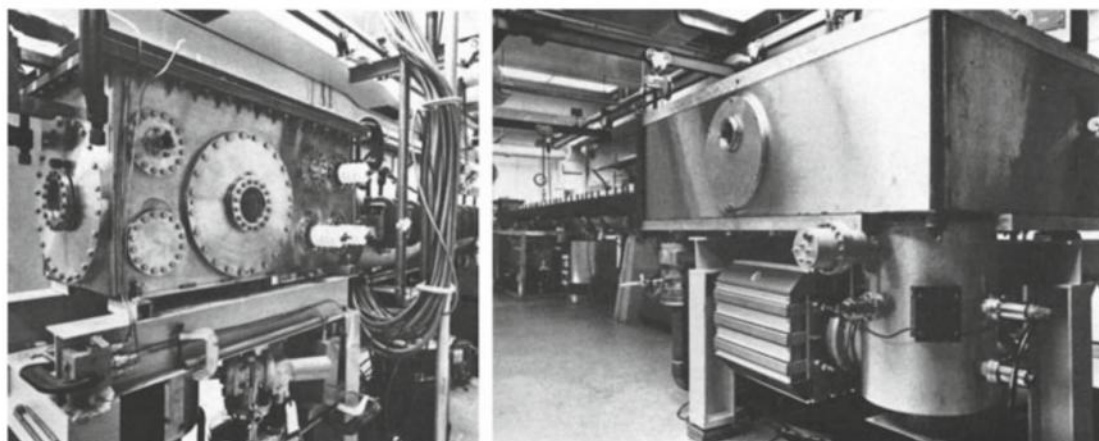
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Fig. 1 Photograph of the "Chemical Accelerator" built at the University of Chicago by Lennard Wharton, to produce molecular beams with an energy of about two electron volts. The photographs show the long dipole accelerator from both ends. Molecules produced by heating in an oven (left) are accelerated down the long tube by electric fields to the reaction chamber (right). Reproduced from [21]

medium declined owing to the invention of the laser, the molecular beam accelerator was made obsolete by the seminal demonstration of John Fenn and co-workers of gas dynamic acceleration of heavy species in seeded supersonic He and H_2 beams [24]. Because of these unsuccessful early attempts, researchers who wanted to pursue molecular beam deceleration with electric or magnetic fields for high-resolution spectroscopy and metrology in the following decades, had difficulties getting this financed [25].

4 Deceleration of CO ($a^3\Pi$) Molecules with Electric Fields

I studied physics at the Katholieke Universiteit Nijmegen in Nijmegen, The Netherlands – since 2004 renamed into Radboud University – and performed my undergraduate and Ph.D. research in the Atomic and Molecular Physics department, headed by Antoni Dymanus and Jörg Reuss. In this department, I was exposed from day one, that is, from February 1984 on, to a wide variety of molecular beam machines; electrostatic and magnetic state-selectors and focusing elements were used throughout. The magnetic properties and the molecular quadrupole tensor of the water molecule [26] as well as the electric and magnetic properties of carbon monoxide [27], for instance, were measured with those machines by Antoni Dymanus and his students already in the nineteen-seventies and are still the standards in the field. During my time as Ph.D. student, Jörg Reuss wrote the chapter entitled "State-selection by non-optical methods" as contribution from Nijmegen to the earlier mentioned classic book on "Atomic and Molecular beam methods" [1].

After post-doctoral periods abroad, I was offered the opportunity to start up my own research program in Nijmegen, in what was then just renamed the Molecular

and Laser Physics department, and I became the successor of Jörg Reuss per January 1995. With my Ph.D. student Rienk Jongma, we performed two-dimensional imaging of metastable CO ($a^3\Pi$) molecules, to study with modern tools the phenomenon of mass-focusing in a seeded molecular beam as well as the performance of an electrostatic hexapole focuser [28]. We showed that metastable CO molecules, prepared with a pulsed laser at a well-defined time, at a well-defined position and in a single quantum state, are ideally suited to study velocity distributions and spatial distributions in molecular beams in general, and to study electric and magnetic field manipulation of molecular beams in particular. The metastable CO molecules live several milliseconds [29], long enough for molecular beam experiments, and with about 6 eV internal energy they can be efficiently detected – temporally and spatially resolved – via the Auger electrons that are released when they impinge on a surface. Whereas the Stark shift of the rotational levels of CO in its electronic ground state is very small, the shift of the $J = 1$ rotational level in the metastable $a^3\Pi_1$ state is on the order of 1 cm^{-1} in electric fields of about 100 kV/cm. When the CO molecules are state-selectively prepared in the metastable state with a pulsed laser *inside* a strong electric field, the interaction with the electric field is suddenly "switched on". This led us to propose a scheme for confining metastable CO molecules in stable "planetary" orbits in an appropriately shaped electrostatic trap [30]. As this confinement scheme would only work for CO molecules with speeds below about 22 m/s, we discussed how such slow molecules could be obtained, and in the final section of our paper we mentioned the possibility to slow down polar molecules with electric fields [30]. The latter was critically commented upon by the (anonymous) referee of our paper, which motivated us to try to experimentally demonstrate that when CO ($a^3\Pi$) molecules are prepared in a high-field seeking state *inside* a large electric field, the molecules will indeed slow down when leaving the high-field region, loosing an amount of kinetic energy that is identical to the Stark-shift of the levels. Even when seeded in Xe, however, the kinetic energy of the CO molecules in our beam was too large, to be able to unambiguously detect the deceleration effect of a single parallel-plate electric field stage of 140 kV/cm. It was clear that we would need either a considerably higher electric field or more electric field stages. When I informed Jörg Reuss in the spring of 1997 about these experiments, he told me that he vaguely remembered an experiment along similar lines by "Wharton in Chicago", but he did not know any further details. This was the first time I learned about Lennard Wharton's earlier experiments, which was comforting as I had been wondering for quite a while already why acceleration or deceleration of molecular beams with electric fields had never been demonstrated – at least now I knew it had been tried.

In December 1997, Rick Bethlem started in my department as Ph.D. student. He was hired on a project to study the physical properties of endohedral fullerenes, which would include the synthesis of small molecules like CO encapsulated in a fullerene cage, and the subsequent spectroscopic characterization of the motion of such a "dumbbell in a box". In one of our first meetings, prior to his start as Ph.D. student, I informed him about our proposed scheme to confine slow CO molecules in stable orbits and about our attempt to demonstrate molecular beam deceleration with a single electric field stage. Rick was fascinated by this topic, that actually

fitted better to his background and interest, and we decided that he would switch research projects and start on the deceleration of metastable CO molecules with electric fields. At that time, I was also actively involved with other members in my department in experiments with an infrared free electron laser [31] and over lunch we had discussions about the operation principle of a linear accelerator (LINAC) for electrons, and whether and how the equivalent device for neutral, polar molecules could be constructed. We discussed in particular whether, as proposed by Basov [18], the "multiple retardation barriers" would need to be combined with electromagnetic radiation fields to be able to repeat the deceleration process, or that one could also rapidly switch the fields on and off; we concluded that both should be possible and decided to go for the approach with the time-varying electric fields. Searching for the term "slow molecules" on the internet gave us then – quite unexpectedly – as one of the first hits the one-page conference abstract by John King from 1959 in which he briefly described the ongoing ammonia deceleration experiments in his laboratory [19] (he there mentioned a 1 m long, 25-stage array of parallel-plate capacitors, using electric fields of 100 kV/cm, i.e., longer but with lower electric fields than what Robert Golub reported upon later [20]), to which we had found no reference in the later work from Lennard Wharton. Different from the earlier attempts by John King and Lennard Wharton, we used seeded pulsed beams and we did not have to rely on time-varying high voltages at a certain fixed frequency. Instead, we could make use of commercially available high voltage switches that can be rapidly (sub-μs) switched on and off in any pre-programmed time-sequence. This made it possible to design an array of – in our first experiment – 63 equidistant electric field stages with a center-to-center distance of 5.5 mm and to have complete flexibility in the input and output velocity that we would like to use. The electric field in each stage is formed by applying a high voltage to two parallel 3-mm-diameter cylindrical rods, centered 4.6 mm apart, leaving 1.6 mm opening for the molecular beam. The two opposing rods are simultaneously switched by two independent high-voltage switches from ground potential to voltages of +10 kV and −10 kV, yielding maximum electric fields of 125 kV/cm in a geometry that also provides transverse focusing. To obtain transverse focusing in two dimensions, adjacent electrode pairs are alternately positioned horizontally and vertically. All horizontal and all vertical electrode pairs are electrically connected and alternately switched, requiring a total of four independent high-voltage switches. A photograph of the prototype, 35 cm long, so-called "Stark-decelerator" as well as a scheme of the experimental set-up is shown in Fig. 2. The two electric field configurations between which switching takes place are schematically shown on the right hand side of Fig. 2.

In Fig. 3, the original measurements are shown that gave us the first hint of signal due to decelerated molecules, indicated by the blue dashed line as a "guide to the eye" – which is needed here. Rick Bethlem had been scheduled to give an oral presentation on the deceleration experiments at the annual meeting of the Dutch AMO community in Lunteren, The Netherlands, in November 1998, and there he presented these results, that had been obtained just a few days before that. In the weeks following this meeting, we managed to significantly improve upon these first results and we demonstrated deceleration of metastable CO ($a^3\Pi_1, J = 1$) molecules in low-
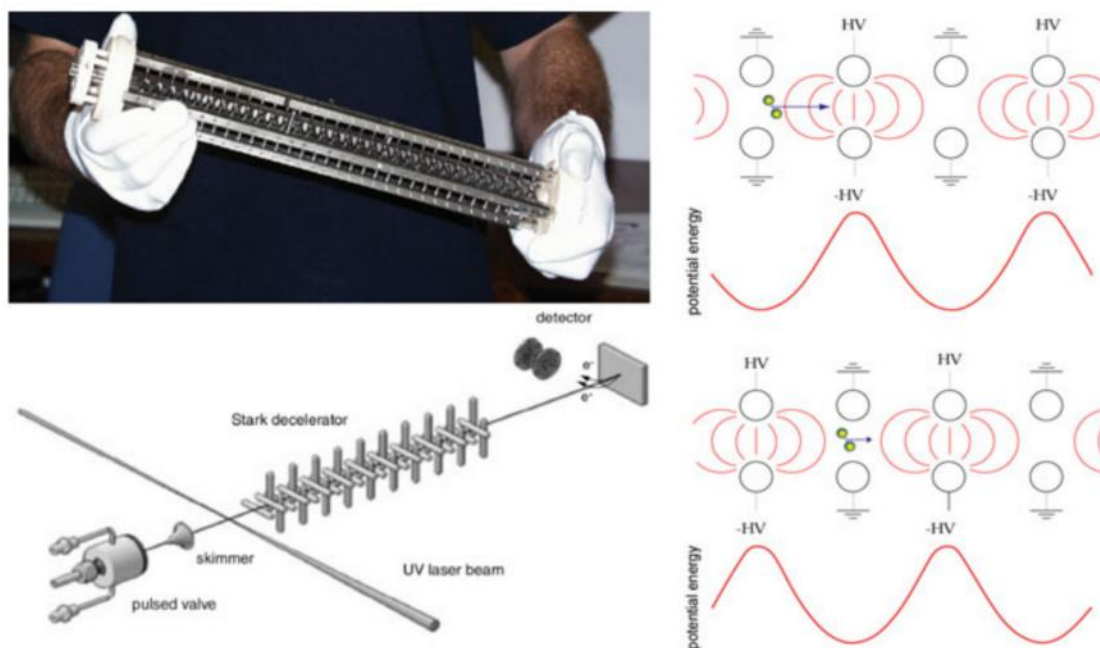
Fig. 2 Rick Bethlem, holding the prototype Stark-decelerator in his hands. The hexapole that is used to optimize the coupling of the beam into the Stark-decelerator can be seen sticking out on the left. A scheme of the experimental setup, omitting the hexapole, is shown underneath the photograph. A pulsed molecular beam is produced by expanding a mixture of 5% of CO seeded in Xe through a pulsed valve into vacuum. After passing through a skimmer, the CO molecules are excited with a pulsed laser to the low-field seeking, upper Λ-doublet component of the $J = 1$ level in the $a^3\Pi_1$ state. The metastable molecules then pass through the Stark-decelerator and are detected by monitoring the electrons that are emitted when they impinge on a clean gold surface. On the right, the two electric field configurations that are alternately used to slow down the metastable CO molecules are schematically shown.

field seeking states from 225 m/s to 98 m/s reducing their kinetic energy by almost 0.8 cm^{-1} per electric field stage [32]. Two years after this, we demonstrated phase-stable acceleration (and deceleration) of – again – CO ($a^3\Pi_1$, $J = 1$) molecules, but this time in high-field seeking states, using an array of twelve dipole lenses in alternate-gradient configuration [33]. These were the successful demonstrations of the experiments that John King and Lennard Wharton set out to perform almost forty years earlier, made possible by the choice of our system, that is, by the advantages that laser-prepared, metastable CO molecules offered, and by the advances in high-voltage switching technology.

When our first manuscript on the Stark-decelerator was still under review, I presented the main results and our future plans during a workshop at ITAMP, in July 1999. There, Daniel Kleppner informed me that although none of the attempted deceleration work of John King had been published, there should exist a Ph.D. thesis from one of his students, and we subsequently traced down the Ph.D. thesis of Robert Golub. Interestingly enough, Robert Golub mentions in his Ph.D. thesis from 1967 that "there has recently been a proposal to use a similar scheme to accelerate molecules by L. Wharton" [20]; it remains unclear in how far Lennard Wharton was
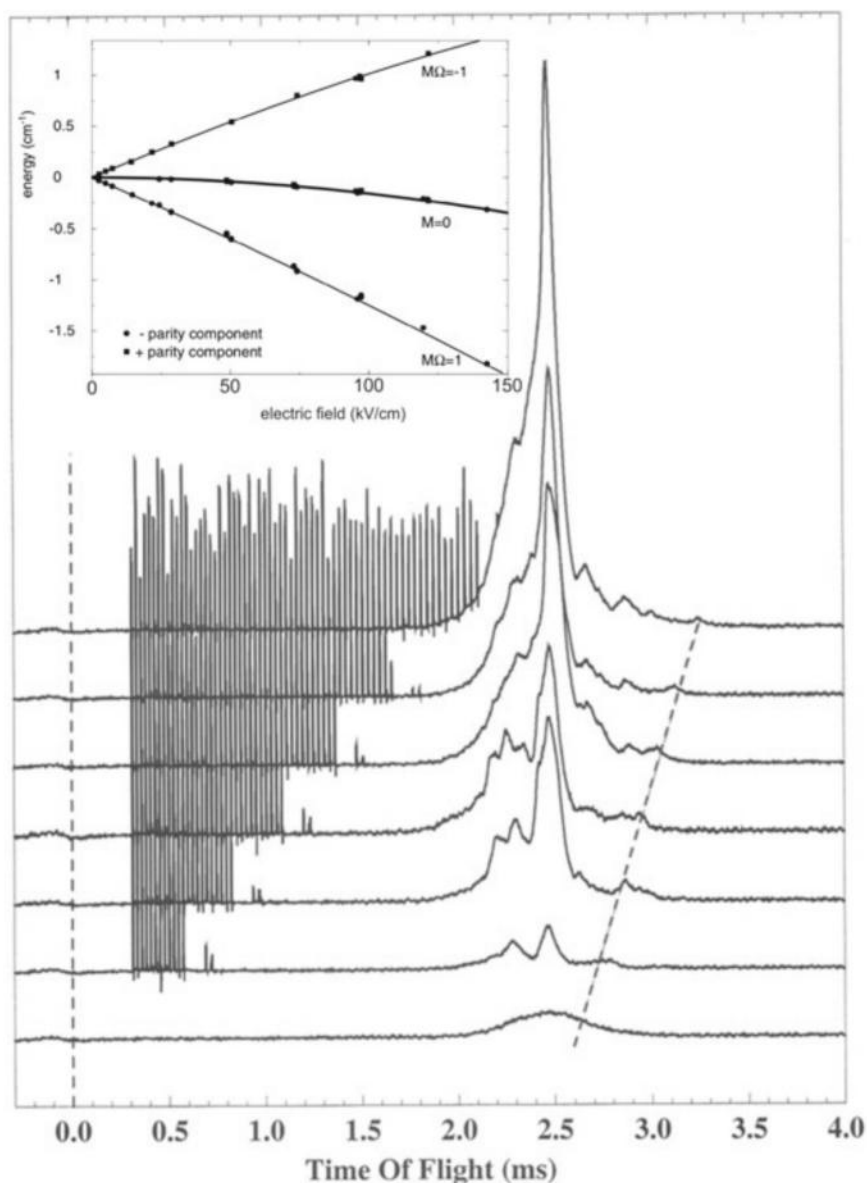
Fig. 3 Observed arrival time distribution of metastable CO molecules when an increasing number of deceleration stages is being used, from none at all (lowest curve) to all of them (uppermost curve). With no fields applied, the arrival time is centered around 2.5 ms after firing of the pulsed laser, i.e., 2.5 ms after preparation of the metastable CO molecules, with a more-or-less Gaussian distribution. The spikes in the signal prior to the arrival of the molecules on the detector are due to electrical noise from the high-voltage switches, and indicate how often the fields have been switched. When electric fields are applied, transverse focusing takes place, and more molecules are seen to arrive on the detector, with a highly structured temporal distribution. The blue, dashed line indicates the arrival time of the synchronous molecules, i.e., those CO molecules that experience the aimed-for, constant amount of deceleration per stage, and these are seen to arrive later when more deceleration stages are being used. In the inset, the measured (solid dots and squares) and calculated Stark shift of the components of the $J = 1$ level in the a$^3\Pi_1$ state of CO are shown in electric fields up to 150 kV/cm.

aware of John King's experiments. During the same workshop, Hossein Sadeghpour informed me on ongoing experiments in the group of Harvey Gould in Berkeley, aimed at decelerating molecules with electric fields. Shortly after our work was published, they published an article in which they presented data on the deceleration of Cs atoms with time-varying electric fields [34].

5 Concluding Remarks

The Stark-decelerator has made it possible to produce packets of state-selected molecules, oriented in space, with computer-controlled six-dimensional phase-space distributions. This level of control of molecular beams has first been demonstrated with electric fields, as outlined in this Chapter, but has also been obtained using time-varying magnetic fields and – to a lesser extent – using electro-magnetic radiation fields by now. Together, these methods have made a whole variety of new experiments possible. It would go too far, to (try to) list these here, and the interested reader is referred to the earlier mentioned Review from 2012 [2], and the references therein. As selected highlights that have appeared since then, I would like to mention the experimental realization of a molecular fountain by Rick Bethlem and co-workers [35], the demonstration of a cryogenic molecular centrifuge in the group of Gerhard Rempe [36], the magnetic deceleration and trapping of molecular oxygen in the group of Edvardas Narevicius [37] and the high-resolution collision experiments in the group of Bas van de Meerakker [38].

"If one extends the rules of two-dimensional focusing to three dimensions, one possesses all ingredients for particle trapping." This is how Wolfgang Paul stated it in his Nobel lecture [39], and as far as the underlying physics principles of particle traps are concerned, it is indeed as simple as that. To experimentally realize the trapping of neutral particles, however, the main challenge is to produce particles that are sufficiently slow that they can be trapped in the relatively shallow traps that can be made. When the particles are confined along a line, rather than around a point, the requirements on the kinetic energy of the particles are more relaxed, and storage of neutrons in a one meter diameter magnetic hexapole torus could thus be demonstrated first [40]. Trapping of atoms in a 3D trap only became feasible when Na atoms were laser cooled to sufficiently low temperatures that they could be confined in a quadrupole magnetic trap [41]. The Stark-decelerator enabled the first demonstration of 3D trapping of neutral ammonia molecules in a quadrupole electrostatic trap [42] even before it was used in the demonstration of an electrostatic storage ring for neutral molecules [43].

There obviously are large similarities between the manipulation of polar molecules and the manipulation of charged particles, and concepts used in the field of charged particle physics can and have been applied to neutral polar molecules, and *vice versa*. Both Hartmut Kallmann and Wolfgang Paul worked on the deflection and focusing of beams of neutral molecules before they turned their attention to controlling the motion of charged particles; it is interesting to realize that multipole fields were actually used
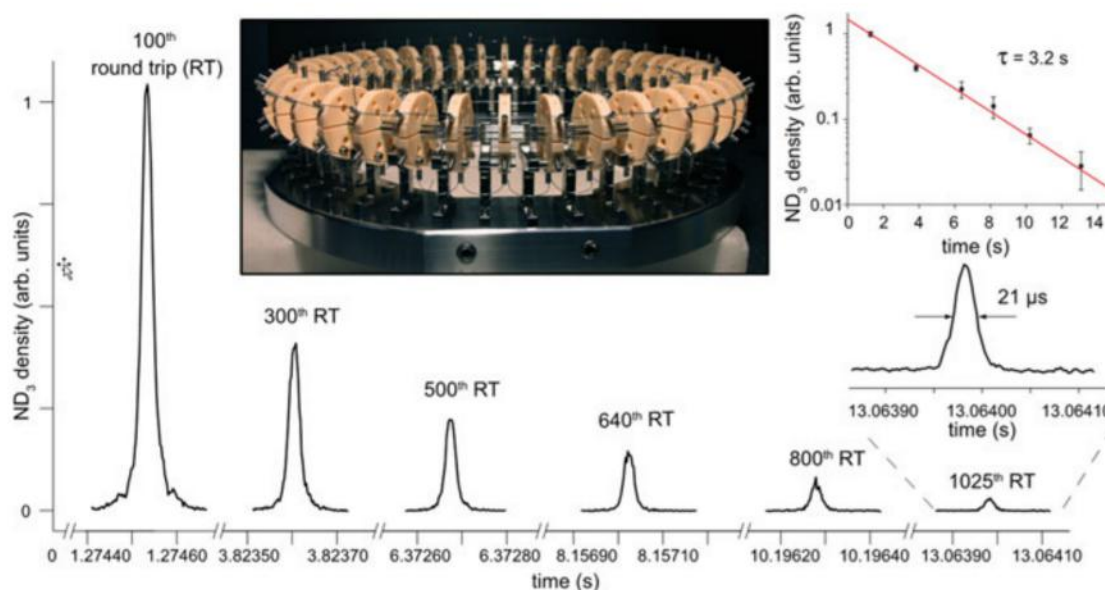
Fig. 4 Photograph of the 0.5 m diameter molecular synchrotron, consisting out of 40 straight hexapoles arranged in a circle. Deuterated ammonia molecules with a velocity of 125 m/s are injected in the synchrotron and stay confined to a 2.5 mm long packet, also after having made more than thousand round trips, i.e., after having travelled a distance of over one mile. The inset shows that the amount of trapped molecules decays with a $1/e$ time of 3.2 s, caused in equal parts by optical pumping due to black-body radiation and collisions with background gas.

in molecular beam physics first. Inspiration from charged particle physics has been instrumental for the development of the Stark-decelerator, the LINAC for neutral, polar molecules. It has also inspired the realization of a molecular synchrotron, shown in Fig. 4, in which state-selected, neutral molecules are kept together in a compact packet for a distance of over one mile, extending their duration on stage to many seconds [44].

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