High-Resolution Momentum Imaging -

From Sterns Molecular Beam Method to the COLTRIMS Reaction Microscope

T. Jahnke[1], V. Mergel[2], O. Jagutzki[1+3], A. Czasch[1+3], K.Ullmann[1+3], R. Ali[4], V. Frohne[5], T. Weber[6], L. P. Schmidt[1], S. Eckart[1], M. Schöffler[1], S. Schößler[1+3], S. Voss[1+3], A. Landers[7], D. Fischer[8], M. Schulz[8], A. Dorn[9], L. Spielberger[10]; R. Moshammer[9], R. Olson[8], M. Prior[6], R. Dörner[1], J. Ullrich[11], C. L. Cocke[12], H. Schmidt-Böcking[1+3];

[1]) Institut für Kernphysik, Universität Frankfurt, 60348 Frankfurt, Germany
[2]) Patentconsult, 65052 Wiesbaden, Germany
[3]) Roentdek GmbH, 65779 Kelkheim, Germany
[4]) Department of Physics, The University of Jordan, Amman 11942, Jordan.
[5]) Department of Physics, Holy Cross College, Notre Dame, 46556 IN, USA
[6]) Chemical Sciences, LBNL, Berkeley, CA 94720, USA
[6])Department of Physics, Auburn University, Auburn, AL 36849, USA
[8]) Department of Physics, Missouri S&T, Rolla, MO 65409, USA
[9]) MPI für Kernphysik, 69117 Heidelberg, Germany
[10]) GTZ, 65760 Eschborn, Germany
[11]) PTB, 38116 Braunschweig, Germany
[12]) Department of Physics, Kansas State University, Manhattan, Kansas 66506, USA

Abstract:

Multi-particle momentum imaging experiments are now capable of providing detailed information on the properties and the dynamics of quantum systems in Atomic, Molecular and Photon (AMO) physics. Historically, Otto Stern can be considered the pioneer of high-resolution momentum measurements of particles moving in a vacuum and he was the first to obtain sub-atomic unit (a.u.) momentum resolution [1]. A major contribution to modern experimental atomic and molecular physics was his so-called molecular beam method [2], which he developed and employed in his experiments. With this method he discovered several fundamental properties of atoms, molecules and nuclei [2+3]. As corresponding particle detection techniques were lacking during his time, he was only able to observe the averaged footprints of large particle ensembles.

Today it is routinely possible to measure the momenta of single particles, because of the tremendous progress in single particle detection and data acquisition electronics. A "state-of-the-art" COLTRIMS reaction microscope [4-11] can measure, for example, the momenta of several particles ejected in the same quantum process in coincidence with sub-a.u. momentum resolution. Such setups can be used to visualize the dynamics of quantum reactions and image the entangled motion of electrons inside atoms and molecules. This review will briefly summarize Stern's work and then present in longer detail the historic steps of the development of the COLTRIMS reaction microscope. Furthermore, some benchmark results are shown which initially paved the way for a broad acceptance of the COLTRIMS approach. Finally, a small selection of milestone work is presented which has been performed during the last two decades.

1. Introduction

What have Stern's Molecular Beam Method (MBM) [2] and the COLTRIMS reaction microscope (C-REMI)[1] [4-11] in common? Both methods yield a very high, sub-atomic unit (a.u.) momentum resolution for low energy particles moving in vacuum. In both approaches the high resolution is obtained because the initial momentum state of the involved quantum particles is very precisely prepared. Conceptually, there is no theoretical limitation for the achievable precision of a momentum measurement of a single particle - the precision is only limited by the design of the macroscopic apparatus [1]. Developing novel experimental detection techniques and achieving higher experimental resolution are often required for advancements in science. Already Stern's second MBM experiment, the famous Stern-Gerlach experiment, performed from 1920 to 1922 in Frankfurt, yielded for silver atoms moving in a vacuum a sub-a.u. momentum resolution in the transverse direction of about 0.1 atomic units (a.u.). Stern and Gerlach achieved this excellent momentum resolution due to a very close collimation of the atomic Ag beam [12]. That way, Stern was able to show that the Ag atoms evaporated from solid silver obeyed the Boltzmann-Maxwell velocity distribution causing a momentum broadening along the beam direction. Later in Hamburg Stern used a double gear system to chop the atomic beam, which yielded also in beam direction a quite mono-energetic beam, further improving the momentum resolution of his apparatus.

At the time Stern performed his experiments (1919-1945), detectors for the detection of individual particles did not exist. Therefore, he was only able to analyze distributions of a large ensemble of individual particles. Today, because of revolutionary developments in the recent decades, as for example, in the electronic detection techniques for low energy particles, in the target cooling, and the advances in multi-parameter data storage, the AMO experimenter can detect and obtain information on single particles and even perform so-called "complete" high-resolution measurements on atomic and molecular many-particle systems. The C-REMI approach [4-11] uses detectors that can detect the position of impact of single particles with very good position resolution (50 micrometer or even less) and measure the arrival time of the particles with a precision of <100 ps. From these quantities the flight times of the particles and thus their velocities are determined with - conceptually - unlimited resolution. A C-REMI setup can reach a single particle-momentum resolution of below 0.01 a.u. and it can detect all fragments emitted from an individual atomic or molecular fragmentation process in coincidence. With such properties, it has been shown in the past, that the entangled dynamics occurring during such processes can be visualized and, in special cases, relative timing resolution of 1 attosecond or better can be inferred [13]. A further important aspect of the C-REMI concept lies in the multi-parameter data handling technique employed. It provides the ability to store the raw data of each detected particle in list-mode on a computer. Thus, the experiment can be replayed during the analysis of the data applying different constraints to the data and investigating different physical aspects of the process under investigation. This advantage is common in nuclear and particle physics, but has become prominent in AMO research with the C-REMI methodologies.

2. History of Stern's molecular beam method: the technological milestones

In 1919, when Otto Stern came back to Frankfurt he began to build his first atomic beam apparatus [3+12] stimulated by Dunoyer's experiment [14]. Already in 1911, Louis Dunoyer had published his famous work on the generation of a so-called atomic beam in the journal Le Radium 8. He had observed that the molecules of a gas that flow from a higher pressure volume

[1] COLTRIMS is the abbreviation for "Cold Target Recoil Ion Momentum Spectroscopy". Another, widely employed name for this technique is "reaction microscope" (REMI). Throughout this article we will use a combination of both acronyms, i.e., C-REMI.

through a small aperture into a vacuum (pressure $<10^{-3}$ Torr) move on a straight line. The development of the molecular beam method MBM became technically possible due to the rapid improvement of vacuum techniques during World War I. Diffusion pumps were invented which enabled a vacuum of below 10^{-5} Torr. Thus for a vacuum of about 10^{-5} Torr the mean free path-length of particles moving with a velocity of about 500 to 1000 m/sec is in the order of 10 m. In such a high vacuum the experimenter can perform controlled deflection and scattering measurements with very high momentum resolution. By deflection of the particle due to an interaction with a known external force (e.g. from electric, magnetic or gravitational fields) Stern could determine atomic properties as, e.g., magnetic or electric dipole moments. The MBM allowed, furthermore, to study the ground-state properties of atoms, which were not accessible by means of photon- or electron spectroscopic methods. The deflection observed in a MBM experiment corresponds to a transverse momentum transfer. This transverse momentum transfer can be determined on an absolute scale when particle velocity and mass are known. However, in all experiments performed by Stern or his group members, beginning 1922 in Frankfurt with the famous "Stern-Gerlach-Experiment" [3,15+16] continuing 1923 until 1933 in Hamburg and from 1933 to 1945 in Pittsburgh only deflection angles were measured using different particle detection techniques [2+15].

Although the Stern-Gerlach-Experiment had already demonstrated in an impressive manner what is achievable by the MBM, Stern and his colleagues continued to introduce improvements, especially during Stern's time in Hamburg. They tried to increase the sensitivity of the method and, more crucially, to further improve the momentum resolution and beam intensity.

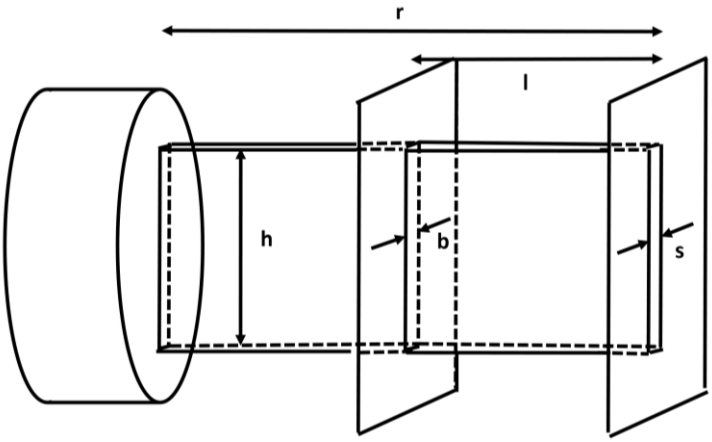

Figure 1: Stern's method of beam intensity amplification with simultaneous improvement of the momentum resolution [2].

In Frankfurt Stern used in his first experiment a heated platinum wire coated with Ag paste. Then in the Stern-Gerlach-Experiment the wire was replaced by an oven, which significantly increased the vapor pressure and thus the intensity of the atomic beam. A further increase in the beam intensity was achieved by using a slit diaphragm (see figure 1) instead of a small hole aperture. Since the MBM only required a high resolution in one transverse direction, the beam aperture could be made very narrow in the horizontal direction (see slit width "b" in figure 1) which improved the apparatus' resolution, but it could be enlarged in the other transverse direction (slit length "h" in figure 1) by a factor of nearly 100. Stern invented the so-called "Multiplikator" [2], where many parallel beams were created in the vertical direction, thus, de-facto allowing for many measurements to be performed in parallel, without affecting the transverse momentum resolution. Stern described in [2] further efforts for improvements of the transverse momentum resolution. The path lengths r and l (see figure 1) were increased by about a factor of 10 compared to the setup employed for the Stern-Gerlach-Experiment and by introducing rotating gears Stern obtained also a quite well-defined longitudinal beam velocity. To be able to measure the tiny magnetic moment of the proton the magnetic deflection force had to be increased (yielding larger deflection angles) and the beams (particular H_2 and He beam) had to emitted from sources operating at the lowest possible temperature. In the last

experiment performed in Hamburg before Stern's emigration in September 1933, Otto Robert Frisch [17] tried to observe the atom recoil momentum which is transferred when a photon is emitted or absorbed from/by an atom, which had been predicted by Albert Einstein. Frisch illuminated a sodium beam at right angle with sodium D_2 light, which caused a deflection of the atoms upon absorption of the light. Stern succeeded in his Hamburg time to improve strongly the momentum resolution thus Frisch was able to detect this very small transverse momentum transfer of about 0.001 a.u. in this experiment, which is considered as the pioneering experiment for the Laser cooling approach. The momentum resolution obtained by Frisch is even nowadays a "state-of-the-art" benchmark achievement.

In the years 1919 to 1922 Stern employed detection techniques where a large number of the silver atoms were deposited on polished brass or glass plates in order to observe a beam spot. Later, by chemical treatments Stern was able observe even a one-atom layer of beam deposition by silver sulfide formation observable as a black spot. In addition, the microscopic beam spot analysis (e.g. by photography) provided an excellent optical resolution in the low micron range, yet not allowing single atom counting. For beams consisting of lighter atoms or molecules, e.g., He and H_2, Stern used a different detection approach. He employed closed gas tubes with a tiny hole for beam entrance. Using very sensitive gas pressure meters he was able to obtain angle-resolved beam scattering distributions. When using H_2 beams, a further method he applied was to measure the heat increase on a metallic surface by a sensitive thermal element. Lastly, he used the Langmuir approach, as well, where the impacting atoms were ionized on a heated wire. The electric current in the wire was proportional the scattered beam intensity. The angular resolution of this method corresponded to the wire thickness [2+16]. Stern was never able to detect single atoms or molecules.

Stern's followers, like Rabi and his scholars, used the MBM mostly for preparation of beams into selected atomic or molecular states. E.g. Townes used a Stern-Gerlach device to produce population inversion to create the first MASER device. Ramsey used two cavities with two separated oscillating fields to excite Cs atoms. In this case one could not decide in which of the cavities the atom was excited. One had to add the excitation amplitudes coherently creating sharp interference structures from which the transition frequency could be determined with excellent resolution (10^{-9}). Both Rabi's scholars were awarded the Nobel Prize in Physics (Townes in 1964 for Maser development and Ramsey in 1989 for the invention of the atomic clock) [18].

3. The C-REMI approach

The multi-coincidence C-REMI approach [4-11] is a many-particle detection device imaging momentum space with high-resolution. The imaging is performed by measuring (in a high vacuum environment) the times-of-flight (TOF) and the positions of impact of low energy charged particles which started in a narrowly confined region in space. From these measurements the particles' trajectories inside the spectrometer volume are inferred yielding the particles' properties. This is similar to studies using the historic bubble chamber in high-energy particle physics.
In the late seventies many atomic physics groups worldwide working at accelerator laboratories investigated ionization processes in noble gas atoms induced by swift ion impact. Many research projects were dealing with the measurement of total and differential cross-sections for single and multiple ionization [19-26] (see in particular review article [26] and references therein). The resulting low-energy ions (referred to as "recoil ions" in the following) attracted interest mainly for two reasons: One research direction tried to measure the probability of ionization as function of the scattering angle by means of a projectile-recoil-ion coincidence. When measuring in thin gas targets at very small deflection angles (milli- and micro-rad) almost exclusively scattering of the projectiles from interaction with the collimation slits was observed. Therefore, to eliminate this slit-scattering problem, the projectile's deflection angle (i.e. it's very small transverse momentum) had to be measured in inverse kinematics, by

measuring the transverse momentum of the recoil ion. The measurement in inverse kinematics would provide, furthermore, a tremendously improved momentum and energy loss resolution if one could bring the target atom before the collision to a nearly complete rest in the laboratory system (which was achieved later by using a super-sonic jet target [9+11+27] or an optical trap [28]). As an example, if in a collision of a 1 GeV/amu Uranium ion on He the projectile energy loss shall be determined, one can measure the momentum change either of the projectile or of the recoiling target atom. When detecting the projectile, the achievable resolution is limited by the properties of the preparation of the incoming beam. Even at the best existing accelerators or storage rings a relative resolution of 10^{-5} is the limit. In case of detecting the recoil ion an energy resolution of below 1 meV can be achieved, yielding a relative resolution in the energy loss of the projectile of far below 10^{-10}.

The other area of interest in the research on very low energy recoil ions, was triggered in the late seventies by the Auger-spectroscopy work of Rido Mann and coworkers [29]. He observed in high energy heavy ion-atom collisions, that (in contradiction to expectations) inner-shell Auger transitions had very narrow line widths. This indicated that the Auger electron emitting recoil ions created in these collisions stayed nearly at rest.

As first step towards developing the C-REMI approach, Charles Lewis Cocke (Kansas State University) and Horst Schmidt-Böcking (Goethe University, Frankfurt) performed together in 1979 at KSU a first test experiment to measure recoil momenta in collisions of MeV heavy ions on He atoms. In this test experiment (using a diffusive room temperature gas target, non-position-sensitive recoil detector and non-focusing recoil-ion extraction field) they measured the TOF difference between the scattered projectile and the recoil ion by performing a scattered projectile-recoil ion coincidence. The measured TOF spectrum could not be converted into absolute values of recoil ion energies, since the spectrometer could not determine the recoil ion emission angle. The results of this test experiment were therefor not published.

In the period 1982-1987 Joachim Ullrich started (as part of his PhD thesis) to develop a new spectrometer approach to determine the absolute value of the transverse momentum of the recoiling target ion by measuring the TOF of the slow recoil ion emitted at 90° to the projectile beam. This development was, for the Frankfurt group, quite risky since the funding request at BMBF/GSI was officially not approved and thus one had to rely at the beginning on "self-made" equipment (e.g. detectors, spectrometers and electronic devices) [9]. Nevertheless, the project was started [30]. It was essential, that one could benefit from the experimental experience from the fields of nuclear and particle physics. C. L. Cocke and H. Schmidt-Böcking had both performed their PhD research in nuclear physics and were trained in using coincidence techniques.

To accomplish the envisioned approach, novel experimental equipment, not commercially available, had to be developed. New self-made position-sensitive detectors (Micro-Channel Plate electron multipliers, MCP, with Backgammon or Wedge & Strip anodes (WSA)) for measuring recoil ions with kinetic energies between zero and several keV were developed and successfully tested. Since 1973 Schmidt-Böcking and his group had developed position-sensitive gas filled Parallel-Plate-Avalanche-Detectors PPAD for performing x-ray/electron heavy-ion coincidence measurements [31]. The work with such detectors required also experience with fast timing electronics and multi-parameter data handling and storing. With a self-made gas filled PPAD the impact time and deflection angle of the high-energy projectiles could be measured in coincidence with the recoil-ion impact time and position. This experience gave confidence that the envisioned C-REMI project was feasible. However, it took until about 1993-1995 before the first full functioning C-REMI was operating. There were moments in these years before 1990 where parts of the project seemed unsolvable. But Joachim Ullrich never gave up! Without his efforts and ideas C-REMI would probably not exist. Besides that, the history of C-REMI is not only a chain of recoil-ion milestone experiments performed by different groups, it is in particular the history of technological developments.

In order to finally obtain sub-a.u. momentum resolution, the target had to be prepared in a state of very small momentum spread which led to using a super-sonic jet source. A further crucial piece was specifically designed electro-magnetic spectrometer fields, that provided optimal momentum focusing with maximum detection efficiency. When, in the early nineties, the detection power of C-REMI became apparent to the atomic physics community, the Frankfurt group was ready to help other groups to build up their own C-REMI systems. Schmidt-Böcking founded in 1990 the company "Roentdek" [32] to produce the C-REMI equipment components or later even deliver complete C-REMI systems to other laboratories. Equipment was delivered to research groups worldwide by selling or in a few cases by loan. The commercial availability of C-REMI systems was essential for the propagation of the C-REMI to several new fields in Physics (single photon research, strong-field and ultrafast sciences etc.). This provided in the last two decades for many groups in AMO physics, as well as in chemistry and biology the support to perform many milestone experiments and pioneering breakthroughs. The C-REMI has enabled insight into many-particle quantum dynamics at the few attosecond scale.

3.1. The development of C-REMI components

In the late seventies and early eighties one of the main research activities in atomic collision physics was to measure total ionization cross sections as function of the recoil ion charge state in high energy heavy-ion rare-gas collisions using the TOF coincidence method [19-26], and to determine such cross sections differentially as function of the projectile scattering angle. These total cross sections had (for single ionization) sometimes macroscopic values [10^{+6} MBarn = 10^{-12} cm^2] and even the creation of completely ionized Ar^{18+} was possible (with a cross section about 1 MBarn in 15.5 MeV/u U on Ar collisions [19]). Both types of experiments required a coincidence measurement between scattered projectiles and recoiling ions. Detecting the recoil ion yielded the start signal, detecting the scattered projectile with a PPAD provided the stop signal.

3.1.1. Detectors

For the detection of the high energy projectile since 1973 self-made gas filled PPADs were available, which could monitor rates up to one GHz very stably in gas flow mode (see figure 2) [31]. Adapted to the experimental task they measured only scattering angles by annular shaped anode structures. At that time electronics were made that enabled a simultaneous measurement of 16 scattering angles.

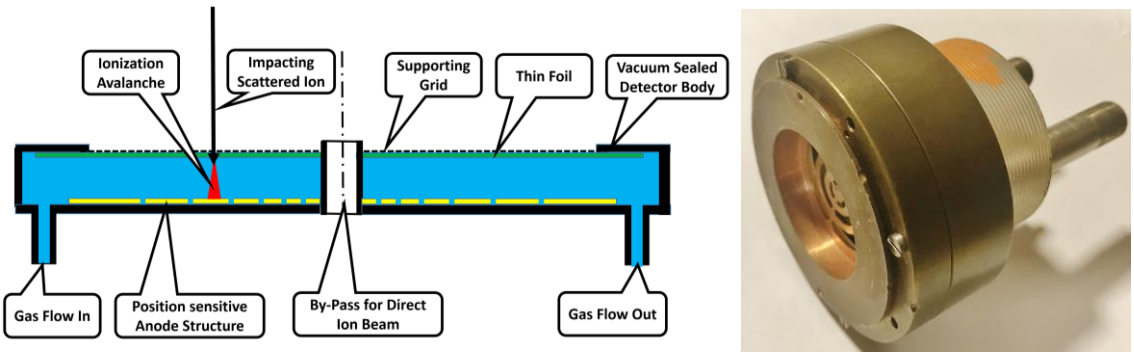

Figure 2: Left: scheme of a PPAD. The impacting ion ejects from the entrance foil several electrons into the gas filled detector volume. In the high electric field (between entrance foil and anode structure) the electrons are accelerated and create a secondary electron avalanche which is detected as function of the anode position. This detector can have a central hole to allow the un-scattered beam to pass through. Right: A photograph of the first PPAD built in 1973 [31]. This detector had three annular anode rings and could handle rates up to 1 GHz.

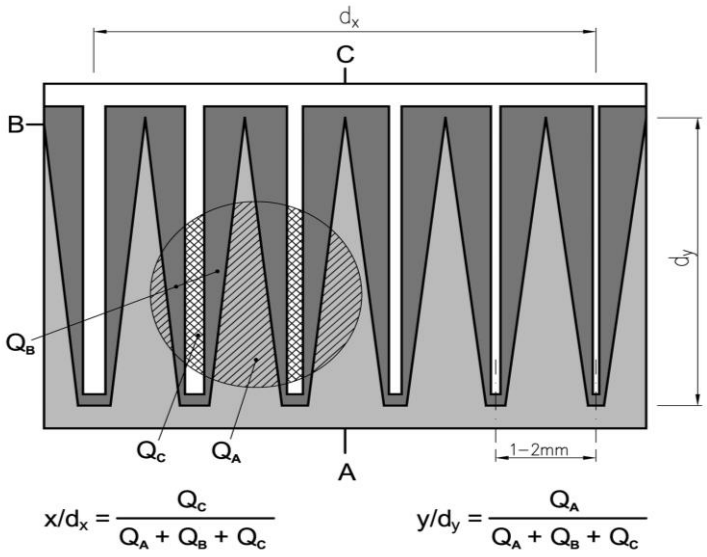

Figure 3: Scheme of the Wedge & Strip anode (WSA). If the charge cloud covers several (at least 2) pitches of three electrodes (B "wedge", C "strip" and A "meander"), measuring the relative charge portions Q_i allows to determine the centroid of the charge cloud [32+33].

A position-sensitive recoil-ion detector for such low energy ions was not commercially available in the early eighties of the last century. As an initial part of a recoil-ion detector so-called micro-channel plates MCP were used [32]. The slow recoils were post-accelerated close to the MCP surface and released, upon impact on the MCP, secondary electrons that induced an avalanche inside the very narrow MCP channels. The single particle detection efficiency of standard MCP is limited by the open area ratio (e. g. how much "hole-area" is present in the total surface). Typical values are 60 %. New developments of MCPs with surfaces, that look like a funnel, increase the efficiency up to 90 % [34]. The position readout of the MCP was performed using a "Wedge and Strip" anode structure. Located behind the MCP, this anode structure yielded information on the position of impact of the primary particle by means of a charge partition method (see figure 3 [32+33]). During the PhD work of Ullrich the anode structures were fabricated as printed circuits. Prior to use, they all needed a careful restoring work by using optical microscopes. In later years such anodes were carefully printed on ceramics and did not need any initial reconditioning. A breakthrough in ion detection was achieved by Ottmar Jagutzki [34] using the delay-line approach for determining the

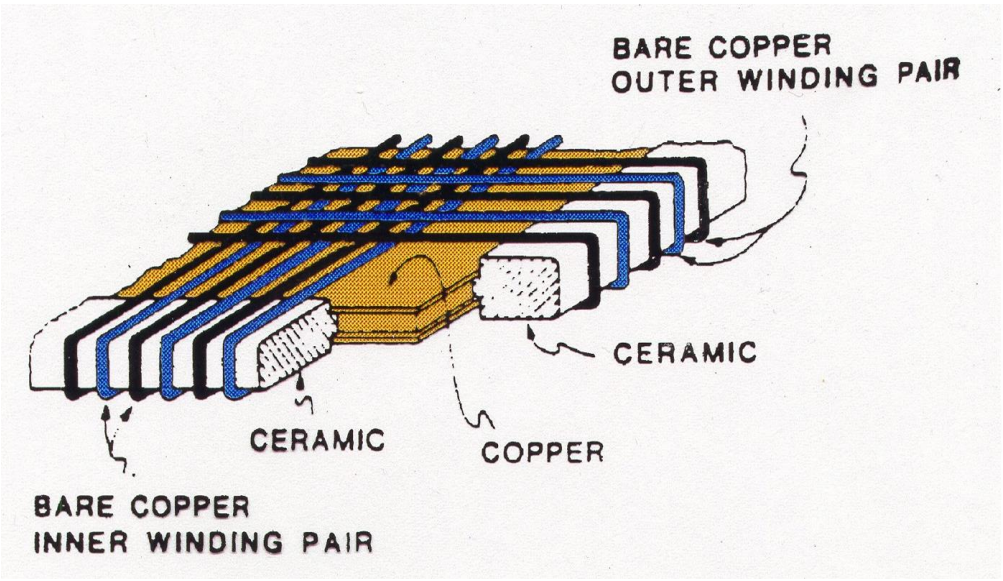

Figure 4: The principle set-up of the delay line anode and other references therein [34+35].

impact positions of the particles on the detector. For such detectors the signal read-out proceeds via a delay-line structure (see figure 4: double-wire structure). From the arrival-time difference at both ends of the delay-line system, the position of particle impact can be determined with a resolution of better than 100 µm. The delay-line approach yields several important advantages as compared to the charge-partition method. It can handle much higher detection rates, since it does not rely on slow charge collection processes. It can detect more than one particle at (almost) the same time (i.e., being multiple-hit capable) because the induced timing signals are very short (in the range of 5 ns to 10 ns) and, lastly, the use of a "timing approach" fits perfectly to the digitized world of computers and is easy to adjust and much cheaper to build.

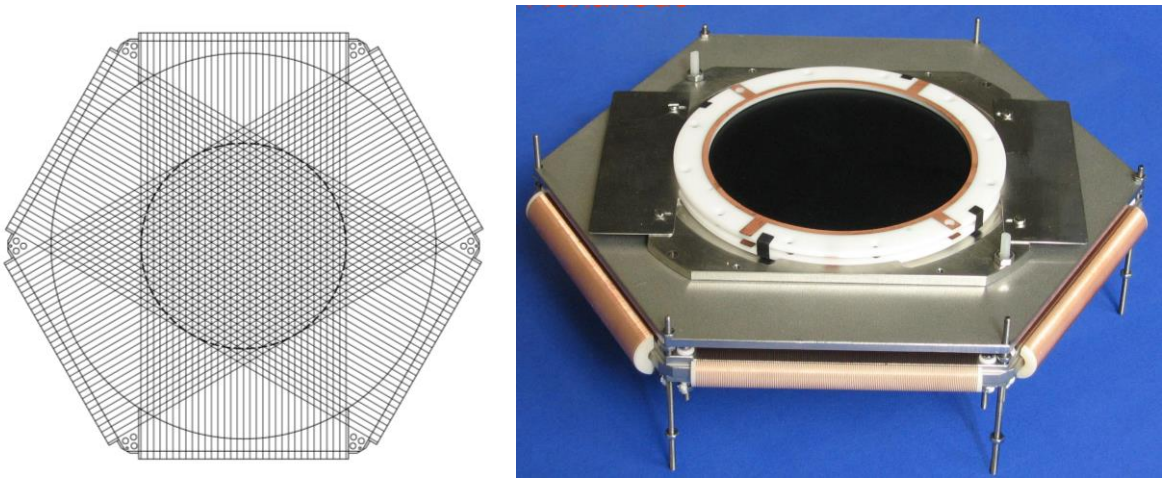

Figure 5: Left: the Hexanode structure and right: working detector system (active area 80 mm diameter) [34-36].

The first generation delay-line detectors consisted of two separate delay lines mounted at right angle. Later, Jagutzki developed a three-layer delay-line structure (a so-called hexanode) (see figure 5). The hexanode detector yields a better linearity and an improved multi-hit resolution with smaller dead-time blockade. The working diameter of these circular delay-line detectors can be as large as 120 mm diameter and recent developments target 150 mm. By using its three delay-lines the hexanode registers redundant information on each particle's position and impact time. Thus, it is possible to recover position and time information for several particles beyond the electronic dead-time limit: Even simultaneously arriving particle pairs can be detected as long as they have a minimum spatial separation of 10 mm.

3.1.2. Multi-parameter data handling

In the late seventies coincidence measurements, which were standard in nuclear physics, were very rarely performed in atomic physics. Thus, there was no need for fast electronics and many-parameter data handling. The electronic hardware for such measurements was quite expensive at that time and thus the comparably small groups of the atomic physics community could not afford to perform coincidence measurements where multi-parameter data had to be registered and stored. Only in nuclear and high-energy particle physics were multi-particle coincidence measurements commonly used. In order to have access to such measurement infrastructure, the Frankfurt atomic physics group, for example, performed all coincidence experiments in nuclear physics laboratories either at GSI-Darmstadt or at the MPI for Nuclear Physics in Heidelberg, where the needed electronics and data storing systems were available. The support by Ulrich Lynen [37] and Reinhold Schuch is highly acknowledged and was absolutely essential for the ongoing development of the C-REMI. Since about 1985 Klaus Ullmann [38] developed a PC based multi-parameter data storing system which was cheap and powerful enough to satisfy the needs of a two-particle or even 7 parameter coincidence measurement (implemented on an Atari ST mainstream personal computer). This development yielded a breakthrough enabling small groups to perform coincidence experiments. The inclusion of all these improvement steps took

about one decade from about 1984 to 1994. The steady progress of this project was published in the annual reports of the IKF-University Frankfurt in the eighties and nineties, i.e., 1984 to 1995.

In the early years the charge signals of the WSA detectors were registered by charge-sensitive preamplifiers and subsequently amplified by standard modules. The timing-signal was created by the "Constant Fraction Discriminator" scheme [39]. These preamplifiers, constant fraction discriminator units etc. were built in the electronics workshops of the Physics Institute in Heidelberg and GSI-Darmstadt. After the foundation of Roentdek GmbH [36] several members of the Frankfurt group were employed at Roentdek and they developed their own electronic circuits. These circuits (based on modern "state-of-art" digital chips) could handle nearly unlimited numbers of parameters per event and allowed high repetition rates. Furthermore, they were inexpensive.

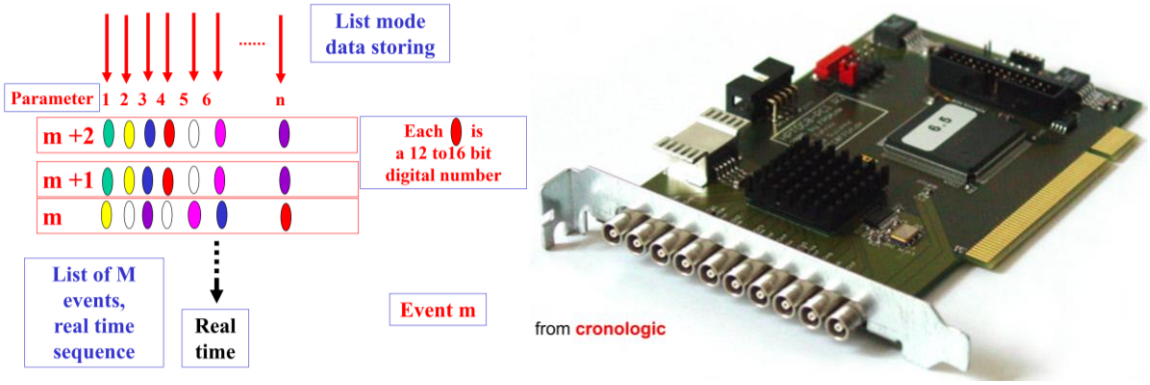

Figure 6: Left: scheme of multi-parameter "List Mode" data recording. Right: an 8-fold fast Time-to-Digital converter made by cronologic [40].

Simultaneously the interface between the electronic modules and the data storing PC changed from slow and expensive CAMAC to fast self-made TDC units (Time-to-digital Converters, with 25 ps timing resolution (see figure 6)) or even ADC units (Analog-to-Digital Converter) [40]. When using fast ADCs, the analog signal is sampled in e.g. 250 ps time-slices and its amplitude is digitized. A fast analysis program can determine several properties, as the "center" of each peak, its height or even disentangle double-peak structures (often referred to as "Camel peaks"). This development was crucial, since in case of the hexanode seven detector signals (2 for each of the three delay-line layers and one

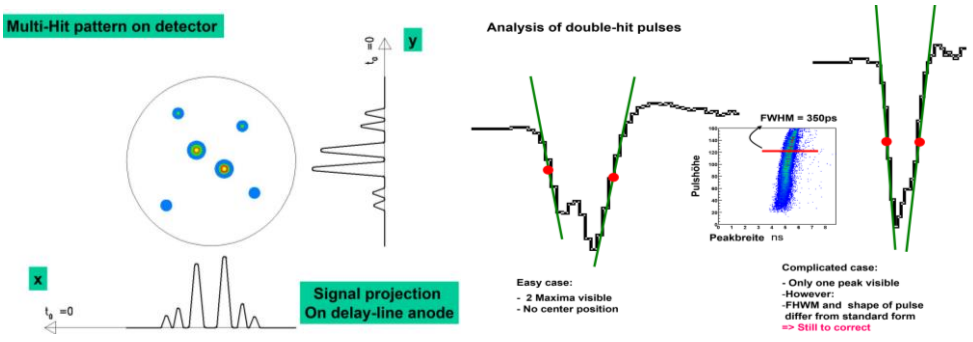

Figure 7: Left: the circle represents the active area of the DL detector. In the event shown 6 particles impacted within about 100 ns on the detector creating electron avalanches that differ in height, which induce in the delay-line structure localized charge clouds. Each of the six ends of delay lines are connected to a fast sampling ADC. Right: the sampled multi-hit signal of one channel is shown. It is analysed later (i.e. after the actual measurement) in high detail using a PC, which allows to resolve the multi-hit pattern.

signal from the MCP to obtain the time of impact) need to be detected for each impacting particle. Thus, a fast multi-hit recovery with very good timing resolution was needed (see figure 7). The present state-of-the-art C-REMI electronics including data list-mode storing can monitor coincidence rates up to several MHz.

3.1.3. Spectrometer design

The first generation of recoil-ion spectrometers (figure 8) was designed to measure only total cross sections for recoil-ion production in energetic heavy-ion collisions as function of the recoil charge state and as function of the final projectile charge state [19-24]. The collimated (1) projectile beam (2) intersected with a diffusive gas jet (3). In the collision with the projectile the target atoms were multiply ionized by, e.g., pure ionization or electron capture. The projectiles (9) were deflected after

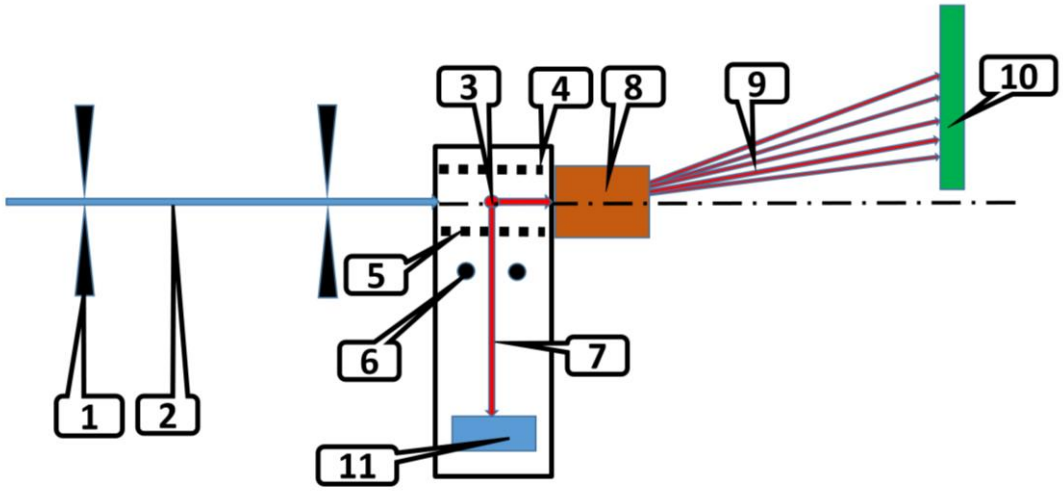

Figure 8: Scheme of the recoil-ion deflected projectile ions (see text).

the interaction with the target by a magnet (8) and detected in a position-sensitive PPAD (10). The final projectile charge states were distinguished by their bending angles behind the magnet. The low energy recoil ions were extracted by an electric field applied between plate (4) and a grid (5), which was on zero-potential. The extracted recoil ions were focused on the recoil detector (11) with the help of an einzel lens (6). The recoil-ion charge state was determined by the TOF of the ions (see figure 9).

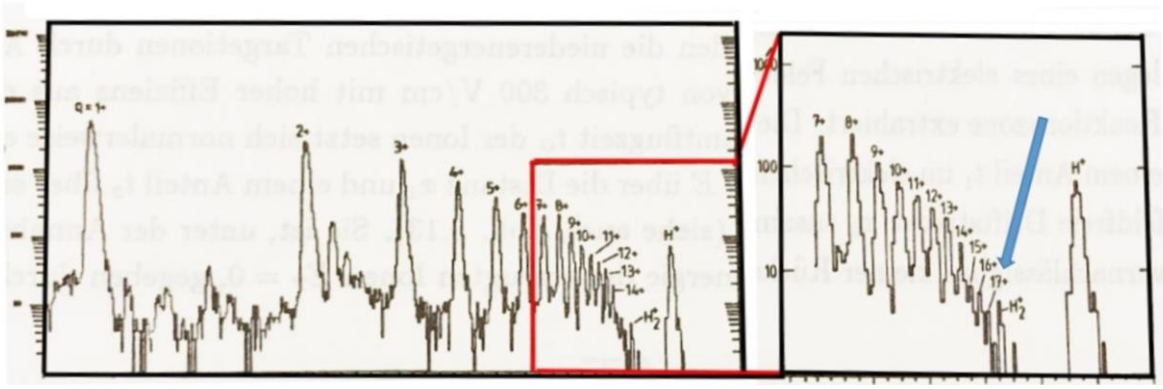

Figure 9: Ar recoil-ion TOF spectrum after the collision with 15.5 MeV/amu U^{75+} projectiles [19]. Even the Ar^{17+} fraction is clearly visible (blue arrow). The Ar^{18+} contribution is covered by the H_2^+ molecular ion charge state. The Ar^{18+} production cross section is approximately 1 MBarn.

The second generation recoil-ion spectrometer was aimed at measuring the transverse recoil-ion momenta. This was the first working recoil-ion momentum spectrometer of the Frankfurt group. In figure 10 the spectrometer used by Ullrich et al. [41] is shown. The projectiles intersected with the diffusive gas target inside a field-free cylinder and were detected downstream with a PPAD. Between the inner and outer cylinder 700 V were applied to post-accelerate the recoil-ions transverse into the recoil-ion spectrometer. These accelerated ions were focused by an einzel lens. In a small magnet the recoil-ion charge states were separated and monitored by a one-dimensional position-sensitive channel-plate detector (the anode structure was only "backgammon"-like). Only such recoil ions were post-accelerated which passed through a tiny hole in the inner cylinder. The recoil velocity (i.e., its momentum) was determined by performing a recoil-ion projectile coincidence yielding the recoil-ion TOF inside the inner (field free) cylinder. The first successful experiments investigating recoil-ion production could be performed in the mid-eighties at the Heavy-ion accelerators (UNILAC) at GSI. The first publication on the new recoil-ion momentum spectrometer with reliable small angle data appeared in Phys. Lett. A [41+42].

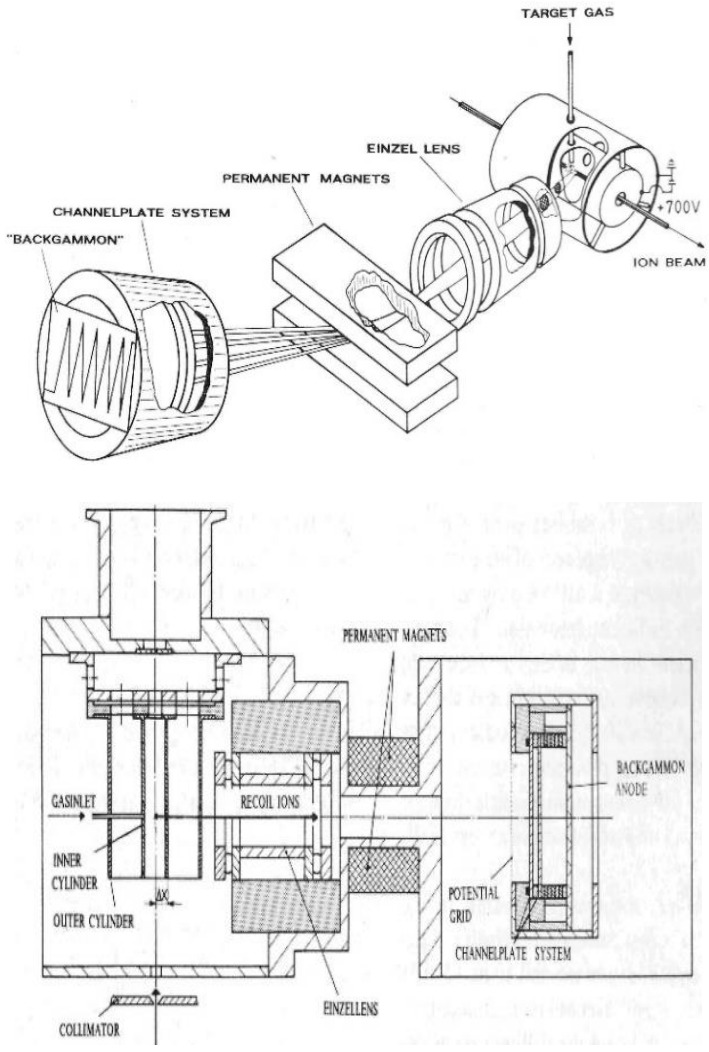

Figure 10: Recoil-ion momentum spectrometer design [41+42] Upper part: 3-dimensional view, lower part: cross section seen from above.

For collisions of Uranium ions on Ne the transverse absolute recoil ion and the scattered projectile momenta were obtained in coincidence (see figure 11). The data showed that at the very small scattering angle of only µrad the sum of each recoil-ion and corresponding projectile transverse momentum did not add up to zero as expected for a two-body collision. By comparing the data to the CTMC theory of Ron Olson [43+44], it became clear that the observed deviations were due to the influence of the emitted electron in the

ionization process and due to the target temperature (the target was at room temperature), too.

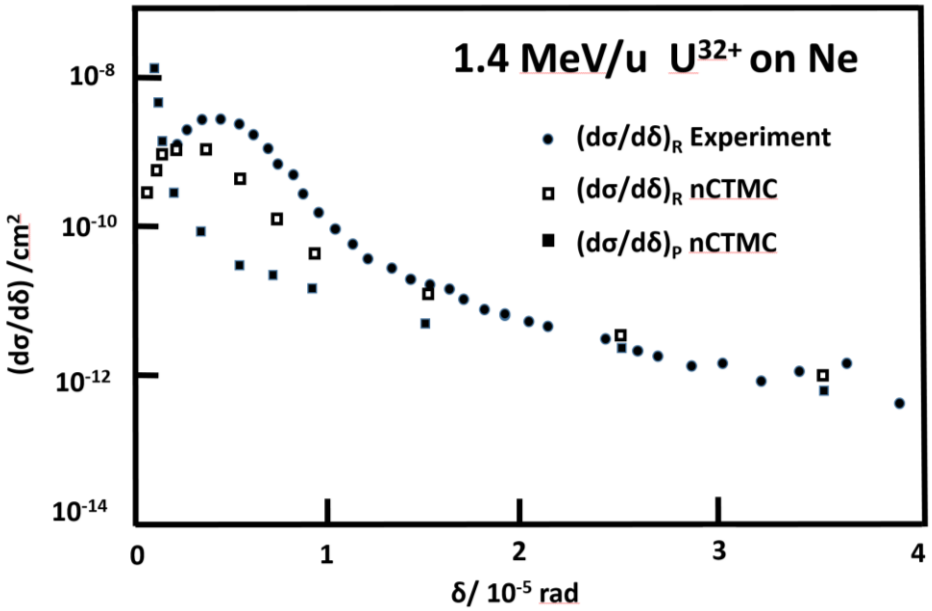

Figure 11: These experiments were the first where - in high-energy heavy-ion rare-gas collisions ionization - probabilities at very small scattering angles < 10 μrad were successfully measured. In parallel, the group of Ivan Sellin at Oak Ridge [46-48] measured mean energies of low energy recoil ions, too.

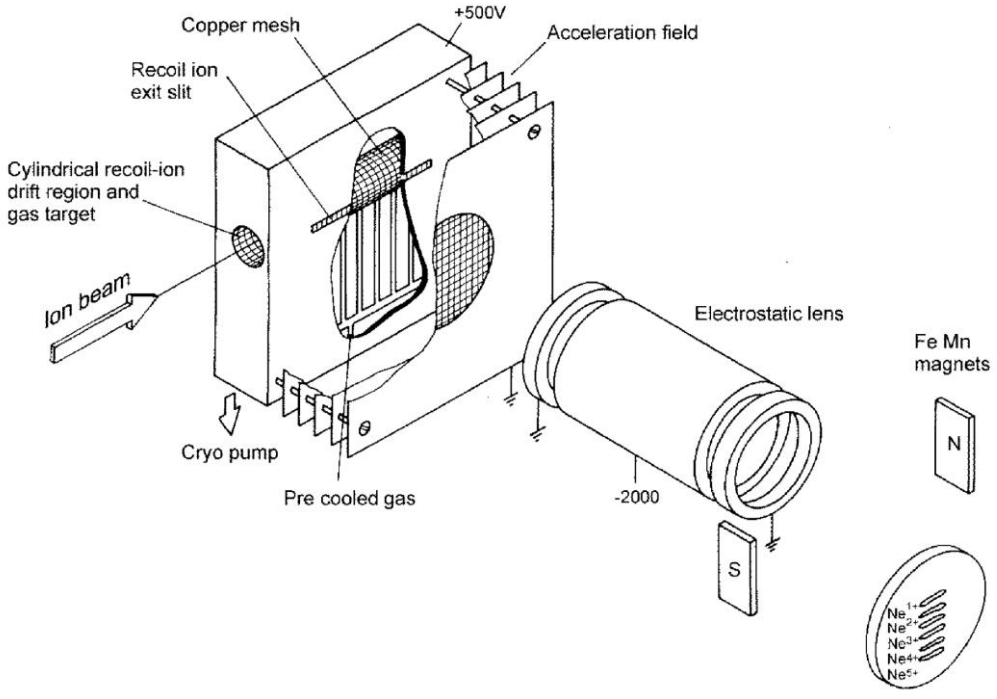

Figure 12: Recoil-ion spectrometer of Dörner et al. [45] (see text).

From Ron Olson's calculation it became clear that internal motion of the gas target (due to its temperature) had to be strongly reduced to obtain quantitative information on the electron momenta in such measurements. Using ultra-cold targets, the method could be improved that much in resolution that electron momenta could be obtained solely by deducing them from the measured momenta of the involved ions. In his PhD work Reinhard Dörner [45] started to build a cooled gas target. He achieved, using a static-pressure gas

target, a temperature reduction down to approximately 15 K. This cooling improved the resolution, but by far not enough. In figure 12 the Dörner-spectrometer is shown. Inside a cooled gold-plated brass housing (connected to the head of a cryogenic pump) the cold He target is intersected by a fast, well-collimated proton beam. The He recoil ions can exit through a slit aperture towards the recoil-ion detector. Behind the slit the recoil ions are post-accelerated, focused by an einzel lens and magnetically deflected. The impact position

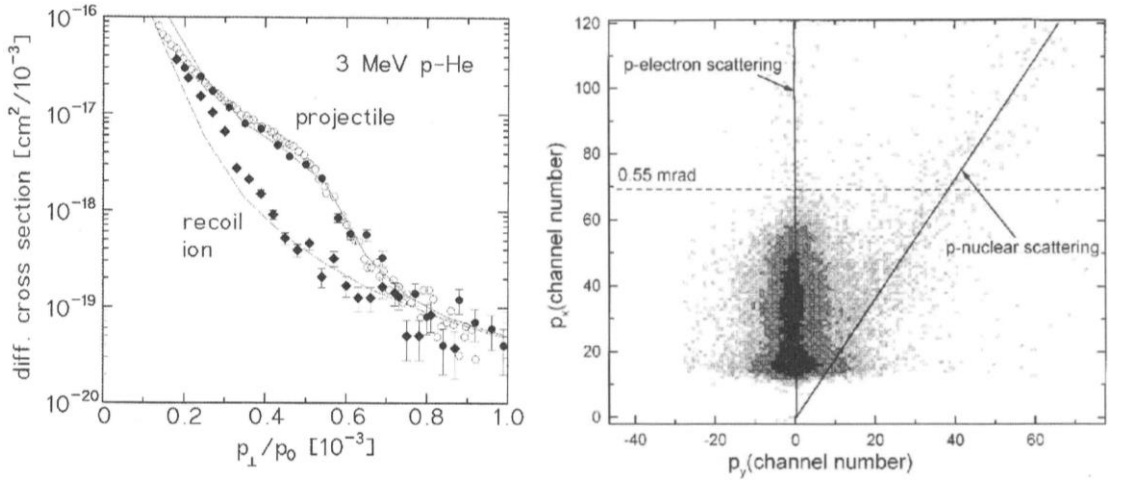

Figure 13: Left: differential single-ionization cross sections of He in 3 MeV Proton collisions [45]. The circles show the identical data plotted versus the measured projectile transverse momentum, the diamonds versus the recoil-ion momenta. As seen in the right plot the projectile can be scattered by the He nucleus (diagonal line p-nuclear scattering) or by the He electrons (vertical line p-electron scattering). Above 0.55 mrad the projectile cannot be deflected by an electron at rest. The p-electron scattering above 0.55 mrad and the broadening below 0.55 mrad is due to the initial electron momentum (plus target temperature).

of the recoil ions is measured by a position-sensitive MCP detector (back-gammon anode) and the TOF of the recoil ions by a coincidence with the scattered projectiles. The measurements of Reinhard Dörner showed that the expected two-body correspondence between projectile and recoil-ion transvers momentum was broken below angles of about 0.6 mrad. The first perception was: this is the principle limitation of recoil-ion momentum spectroscopy. However, with the help of CTMC calculations of Ron Olson and numerous discussions with him (Olson was a Humboldt Award fellow in Frankfurt from 1986 to 1987) it became clear, that the method (recoil-ion momentum spectroscopy) was not limited to projectile scattering angles above 10^{-5} rad, but that the method was at lower angles even sensitive to the momenta of the involved electrons if the target temperature could be decreased by several orders of magnitude. The experiment performed by Reinhard Dörner demonstrated that further target cooling would improve the momentum resolution and that it should be possible to measure the momentum exchange between nuclei and electrons with high resolution. This observation was a milestone perception towards the realization of C-REMI.

Rami Ali and Charles Lewis Cocke at KSU used the first recoil-ion extraction system [49+50], which was time-focusing [51]. Thus the KSU group was the first to determine the Q-value (inelastic energy loss or gain) in an ion-atom collision process by measuring the longitudinal momentum component of the recoil ion [49] (i.e. parallel to the incident projectile momentum). They investigated the multiple electron capture process in 50 keV Ar^{15+} on Ar collisions and obtained a Q-value resolution of about 30 eV. Relative to the projectile kinetic energy this corresponds to a resolution just below the 10^{-3} level (see figure 14). The method of Q-value determination by the longitudinal recoil momentum

component has been discussed before in an invited talk by Dörner et al. at the ICPEAC in Brisbane 1991 [52]. In this invited lecture, it was shown that in high-energy heavy ion collisions a relative Q-value resolution far below 10^{-6} can be obtained.

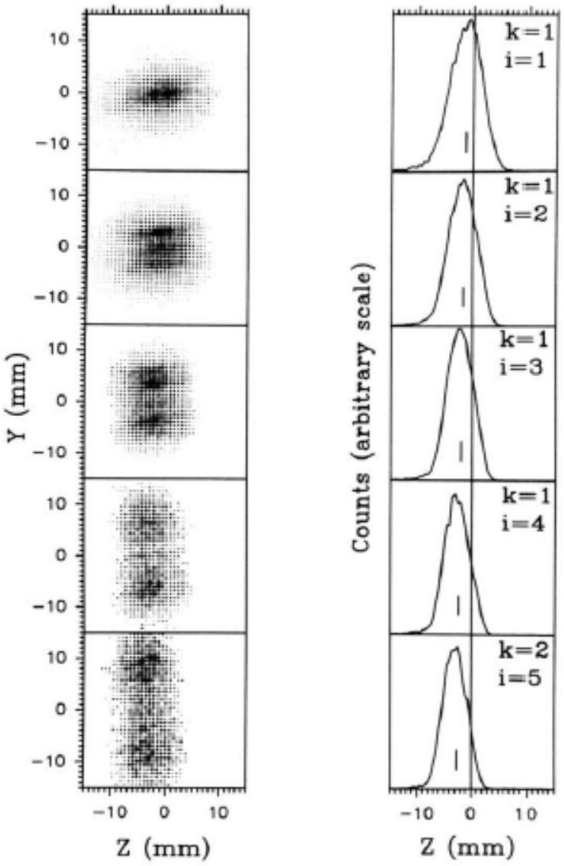

Figure 14: Left: two-dimensional recoil-ion momentum distributions (z-abscissa: longitudinal component, y-axis: transverse component) for different projectile charge change k and recoil-ion charge state i. Right: their projections on the z-abscissa. The vertical bars indicate the center of the projections [49+50].

The first breakthrough into the high-resolution domain was achieved in the "Diplomarbeit" of Volker Mergel [53] by performing an experiment with a He super-sonic jet as target and using a spectrometer design with strongly improved time-focusing properties. A further significant improvement was then achieved in the PHD-thesis work of Volker Mergel [53], which used the first three-dimensionally focusing spectrometer. This spectrometer included the so called time focusing conditions and the focusing of the extension of the gas jet in the direction of the electrical field for the recoil-ion extraction. For the first time this spectrometer combined the time focusing with a two-dimensional focusing lens in the extraction field focusing the gas jet projectile reaction volume with respect to the two dimensions perpendicular to the extraction field (figure 15). Using this three-dimensional focusing in combination with a pre-cooled (17 K) super-sonic gas jet, a momentum resolution of 0.05 a.u. was achieved in all three dimensions, which was a breakthrough in momentum resolution of C-REMI and was the best resolution achieved at that time [53][1].

Footnote:

[1] As Volker Mergel remembers: The three-dimensional focusing was invented in the early nineties during a night-shift performing an experiment at the tandem accelerator at KSU. In that night-shift, an experimental resolution was observed which was better than expected. These surprisingly good experimental results triggered a discussion between Charles Lewis Cocke and Volker Mergel searching for the reasons. Performing in that night-shift some calculations on the possible electric field configuration Volker Mergel could show that the reason for the improved resolution must be an inhomogeneity of the electrical field, which accidentally caused a focusing of the extension of the reaction volume. As a result of this discussion Volker Mergel built a new three-dimensionally focusing spectrometer as shown in figure 15. This technology was then also patented [DE 196 04 472 C1].

In the early nineties, the Göttingen group of Udo Buck and Jan Peter Toennies [27] provided important support in constructing a super-sonic jet as target. Using this recoil technique, in the late nineties, Daniel Fischer in the group of Joachim Ullrich obtained a resolution of 10^{-6} in the Q-value measurement [11].

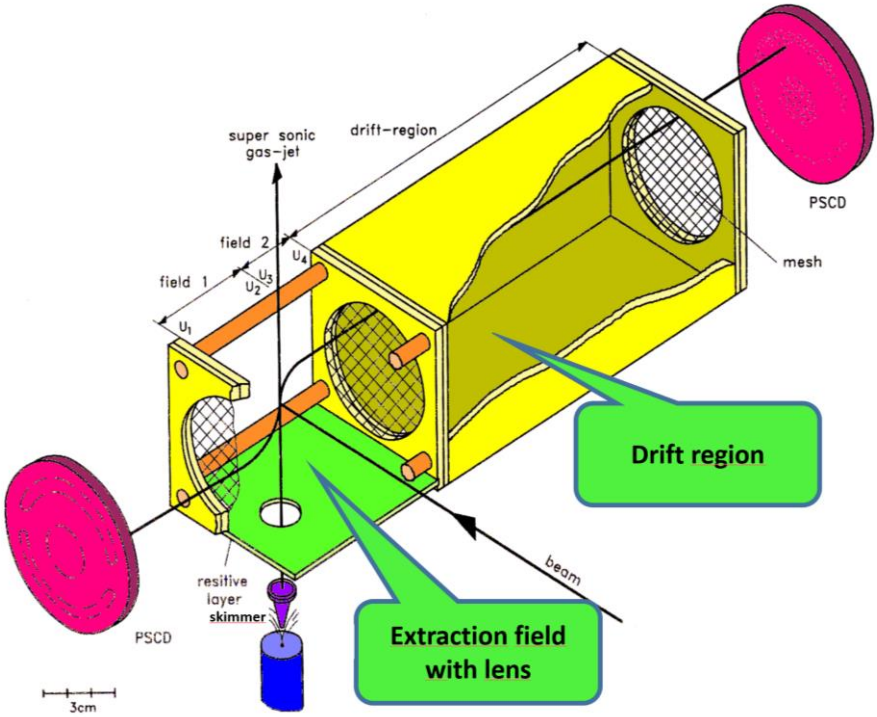

Figure 15: Recoil-ion spectrometer used by Mergel et al. [53] with transverse extraction. The extraction field and drift zone are designed in length and field strength to obtain time focusing conditions. The detector PSCD on the right side monitors recoil ions and the detector on the left side electrons.

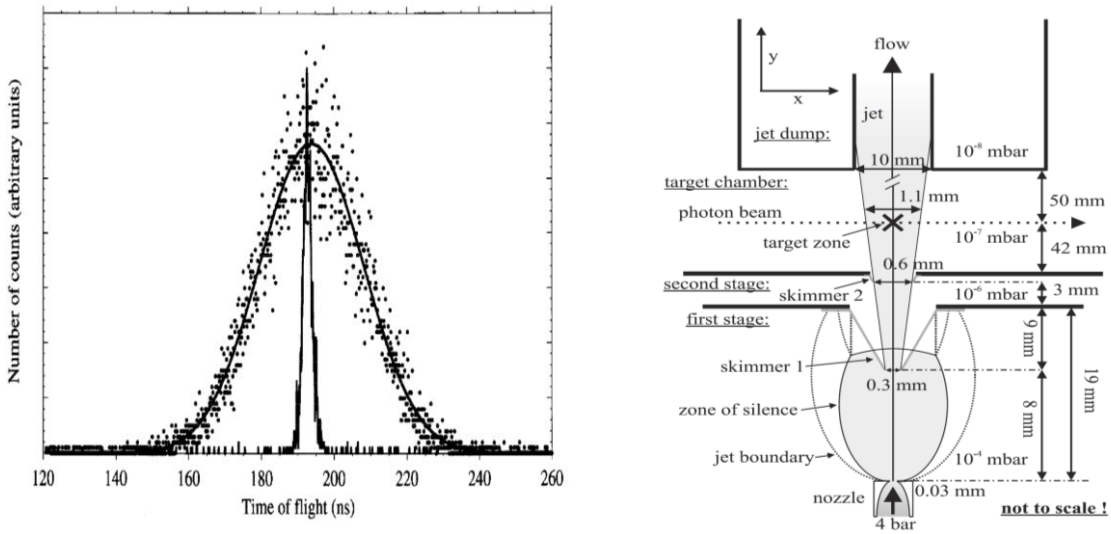

Figure 16: TOF distribution of recoil ions emitted from a room temperature target in comparison to ions emitted from a super-sonic jet. The full line shows the thermal momentum distribution [9]. Right: Two-stage super-sonic jet [58].

In the early nineties also the group of Amine Cassimi at Caen [54-57] started to use a super-sonic jet-target for recoil-ion production. Their original motivation was to build a source for intense, cold and highly-charged ions based on recoil-ion production. The effect of target cooling is visible in figure 16, where the recoil-momentum distribution for a room temperature and a super-sonic jet target are compared. The small transverse spread in the

momentum distribution of a super-sonic jet can still be improved by collimating the super-sonic beam with skimmers, thus nearly unlimited sub-atomic resolution in the transvers momentum space can be obtained.

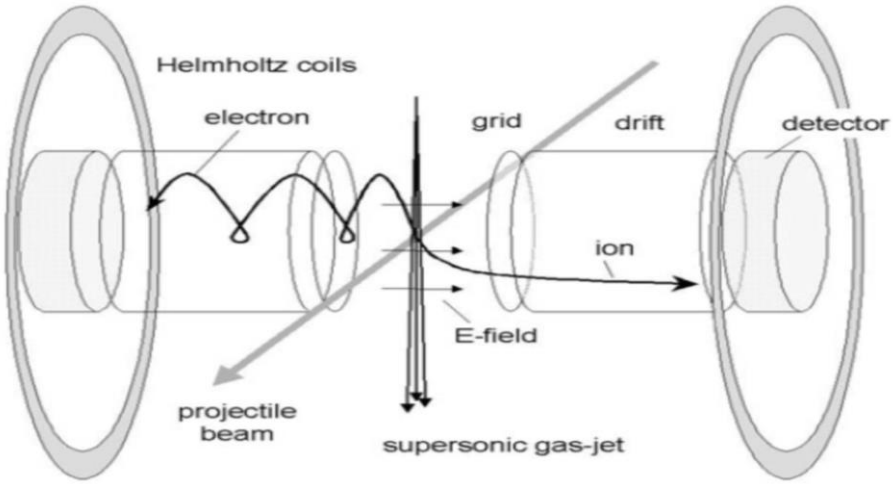

Figure 17: The COLTRIMS reaction microscope with guiding magnetic field [11].

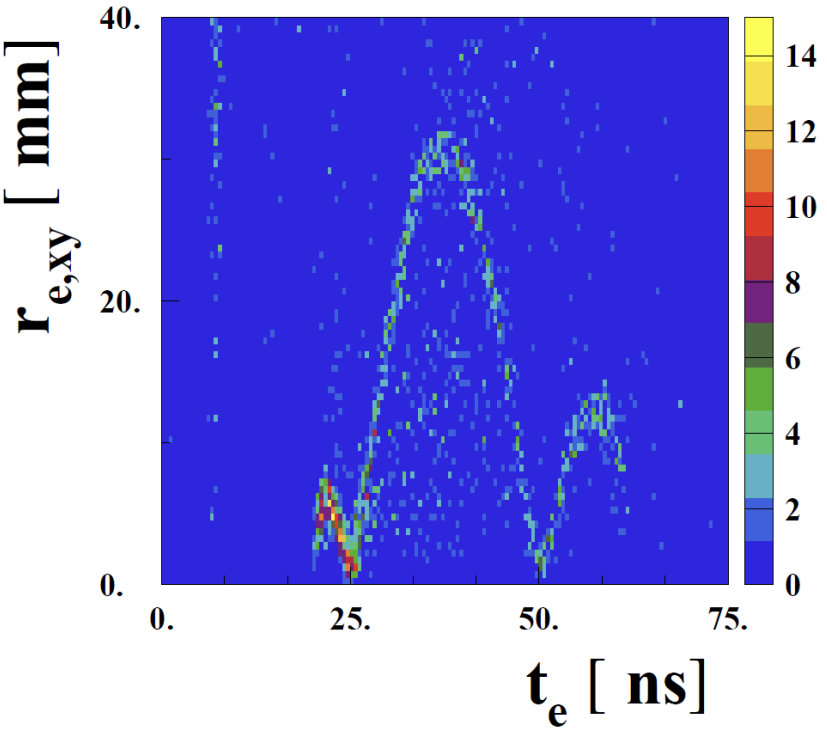

Figure 18: Plotted is the TOF versus radius of electron trajectories inside the spectrometer in the presence of the magnetic field B [58]. The time between minima in the radius is the cyclotron motion period, which is used to measure the magnetic field.

The next very important milestone step towards today's "state-of-the-art" C-REMI system as a multi-particle momentum-imaging device was the incorporation of the magnetic field confinement of high-energy electrons. With this improvement, one could finally achieve a nearly 4π geometrical solid angle for the detection of even high energy electrons. Joachim Ullrich and Robert Moshammer conceived this benchmark development in the early nineties [5+11]. It increased the multi-coincidence detection efficiency by orders of magnitude. As shown in [5+11] the guiding magnetic field pointing in parallel to the axis of the electric extraction-field provides an unambiguous determination of the initial electron momentum, as long as the electron is not detected at flight times where its detection radius is re-approaching zero.

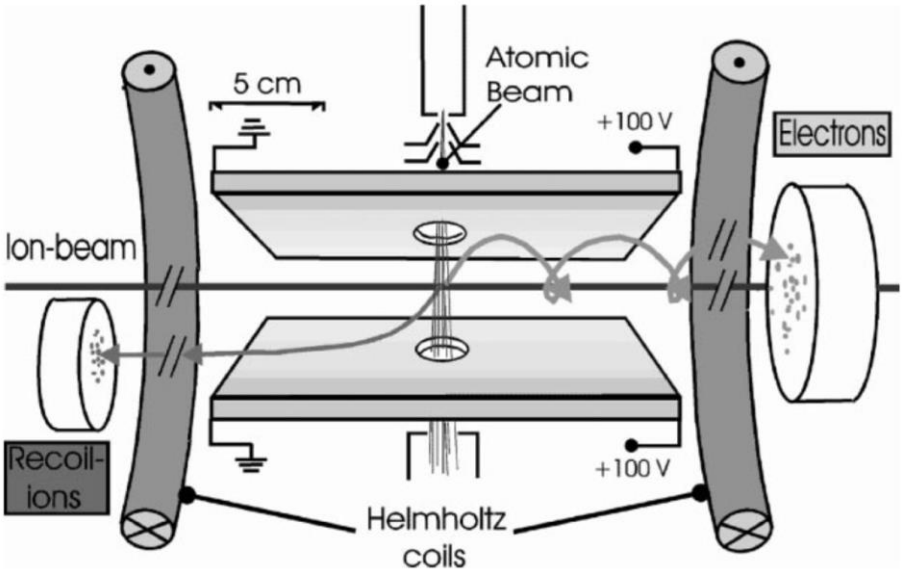

Figure 19: The C-REMI system of Ullrich and Moshammer installed at the GSI storage ring (ESR) [5+11].

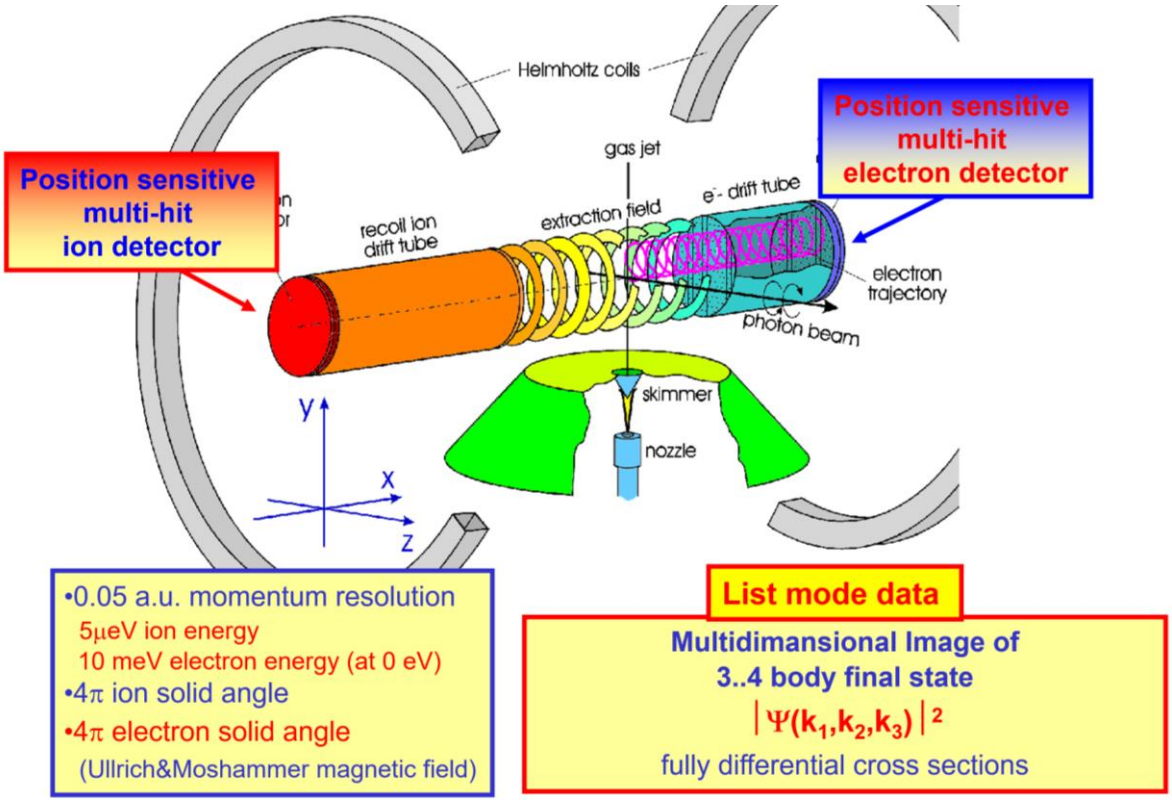

Figure 20: Schematic view of a C-REMI system.

Joachim Ullrich, Robert Moshammer et al. performed several of their measurements on the physics of the recoil-ion momentum at the GSI storage ring ESR. The operation of the ESR required a wide-open spectrometer for the circulating ion beam. Therefore, they designed a spectrometer where a particle-extraction can be performed in principle in any direction (see Figure 19), but with preferential extraction in the longitudinal direction along the ion beam. The recoil-ion as well as the three electron detectors were positioned in time-focusing geometry [5+11], which allowed a high Q-value resolution. Figure 20 shows a schematic view of a modern, "state-of-the-art" C-REMI system with transverse extraction and time-focusing.

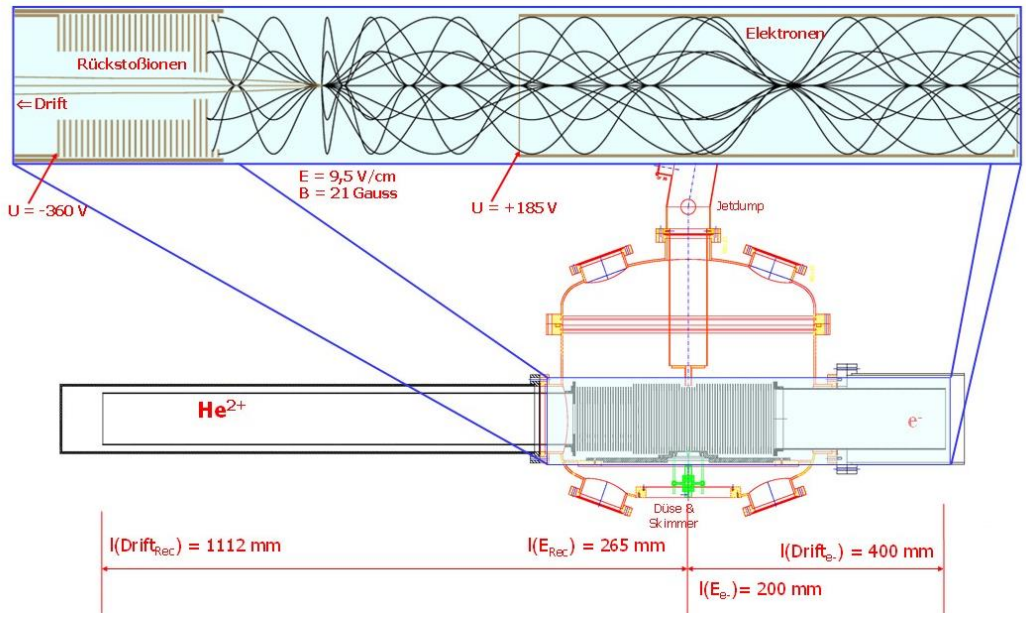

Figure 21: Electrostatic lens-system of a C-REMI with transverse extraction showing ion and electron trajectories and time marker [59].

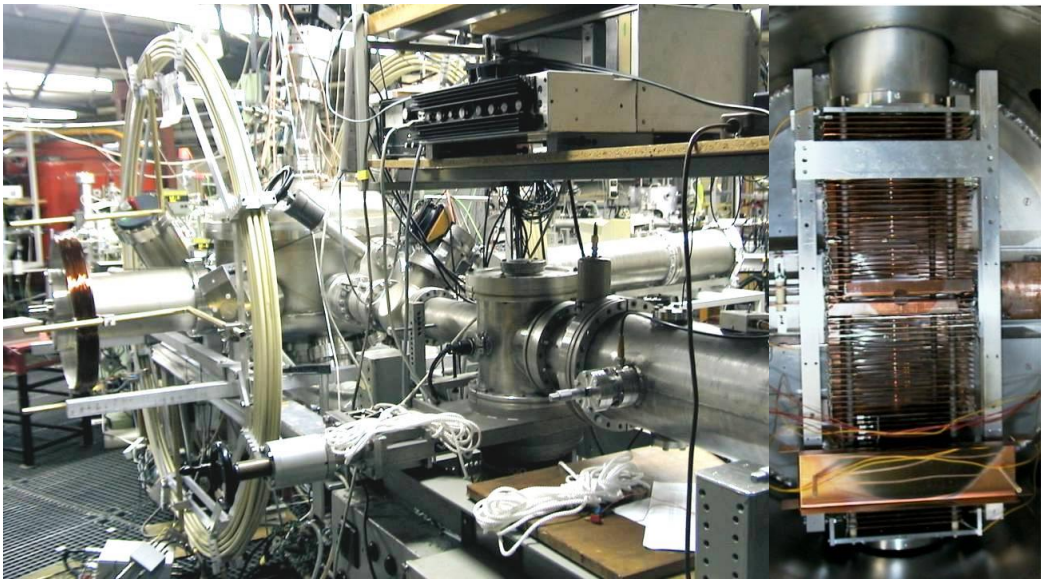

Figure 22: One of the Frankfurt C-REMI systems used for MeV p on He collisions. Left: View from outside. Right: Inside the vacuum system with view on the C-REMI spectrometer [36].

4. The early benchmark results

In the eighties many experts in atomic physics were skeptical that a C-REMI-like approach could actually work. The high resolution and detection efficiency of the C-REMI method was slowly recognized and acknowledged by the physics community, when in the nineties first benchmark results were obtained and published. Visiting Frankfurt, the Russian physicist Afrosimov [60] from the Joffe Institute in Leningrad (now St. Petersburg) told Schmidt-Böcking that in the fifties Russian physicist were discussing a detection method similar to a C-REMI. But they did not pursue this concept, since they did not believe that it could work because of the thermal motion of the target atoms and molecules. Using room temperature targets first angular resolved recoil-ion measurements were performed in the late seventies [61+62]. In lectures on "Inelastic Energy-Loss Measurements in Single Collisions" Bent Fastrup [63] discussed the advantages of recoil ion momentum spectroscopy, i.e. the method of inverse kinematics. But he also did not pursue recoil-ion momentum spectroscopy, since a target at room temperature did not allow a good

momentum resolution. To our knowledge in the late seventies or early eighties no group was developing recoil-ion detection devices with larger solid angle imaging features. The required equipment like detectors, electronics, coincidence and vacuum equipment and cold target preparation methods were not available at that time to give a C-REMI a real chance of success.

4.1. Q-value measurements

At ICPEAC XVII in 1991 in Brisbane, the Frankfurt group presented theoretical estimates and first experimental results on the high-resolution obtainable by the cold-target recoil-ion method [52]. The prediction was that in MeV/u heavy-ion atom collisions a Q-value resolution relative to the projectile energy of below 10^{-8} and deflection angles below 10^{-8} rad could be measured. The relation between **Q**-value and recoil longitudinal momentum $\mathbf{p}_{r\parallel}$ is: $\mathbf{Q} = -(\mathbf{p}_{r\parallel} + q/2) \bullet v_p$ [9+11], where q is the number of electrons transferred from the target to the projectile and v_p is the projectile velocity (all values are in a.u.). The summand $(q/2 \bullet v_p)$ is due to the mass transfer of electrons from the target atom at rest into the fast moving projectile system. The experimental verification of the high-resolution power of the C-REMI approach was demonstrated in the period of 1992 to 1994 by Volker Mergel [53] when he assembled the first fully working **COLTRIMS** (**CO**ld **T**arget **R**ecoil **I**on **M**omentum **S**pectrometer) system (see figure 15). It included a super-sonic He jet as target. The He gas was pre-cooled down to about 15 K and expanded under high pressure (> 10 bar) through a nozzle of 20 µm diameter into vacuum. By the expansion process the inner temperature of the super-sonic beam decreased to a few mK.

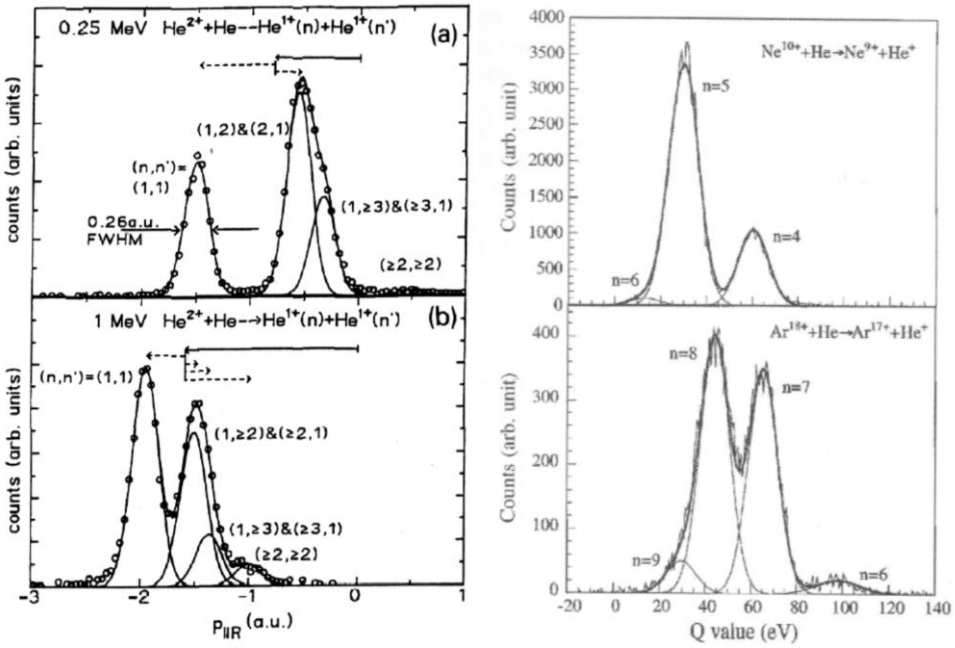

Figure 23: Left: $\mathbf{p}_{r\parallel}$ measurement of Mergel et al. [53]. The numbers in the brackets indicate the mean shell of the transferred electron in the initial and final state, respectively. 1 a.u. corresponds to a Q-value of 27.2 eV. Right: Q-value measurement of the GANIL group [54]. The kinetic energy of Ne^{10+} was 6,82 keV/u and 6.75 keV/u for Ar^{18+}.

The beam was collimated by a skimmer (about 1 mm circular opening) to reduce the transverse momentum spread of the gas jet. Furthermore, static electric extraction fields of the C-REMI were designed to provide perfect time focusing [9+11+53]. Using a predecessor of the spectrometer as shown in figure 15 in 250 keV He^{2+} on He collisions, Volker Mergel et al. [53] obtained an energy loss/gain resolution of 0.26 a.u. (i.e. 7 eV) by measuring the longitudinal recoil-ion momentum (figure 23). Relative to the kinetic energy of the impacting He^{2+} projectiles this is an energy loss resolution in the order of 10^{-5}. It is to notice that one can determine the energy loss/gain of projectiles extremely precisely

without accurate knowledge of the projectile beam energy. By using the three-dimensional focusing technology of the spectrometer as shown in figure 15, the resolution could be improved by a factor of about 5 yielding a resolution of 0.05 a.u. in all three dimensions. The method allowed one to visualize details of electron transitions in a collision and to determine the involved electronic energy transfer with high resolution (see also [54-57, 64-66]).

4.2. Electron-electron contributions in the ionization process of ion-atom collisions

In the nineties, several groups tried to separate the contributions of target-nuclei-electron (n_t-e) and electron-electron (e-e) interaction in ion-atom collisions. The electron-electron interaction can only knock out the bound electron if the mean relative velocity (projectile velocity) exceeds a certain barrier. Thus measuring the projectile ionization cross section as function of the projectile velocity the (e-e) contribution would contribute only above a certain velocity. Using the C-REMI approach, however, both contributions should become distinguishable in the recoil-ion momentum distribution. In the (e-e) process both involved projectile and target electrons are knocked-out and the recoil ion would act only as an observer, thus its final momentum remains at target temperature (close to zero momentum). In the (n_t-e) process the recoil ion must compensate the momentum of the electron knocked-out. In figure 24 (left side) the two mechanisms are explained by diagrams and the measured recoil ion momentum data for He^+ on He collisions are shown [67]. At this impact energy the two peaks in the distribution are clearly separated. On the right side of figure 24 the data are shown for F^{8+} on He [68] at two impact energies (left below the barrier, right above the barrier).

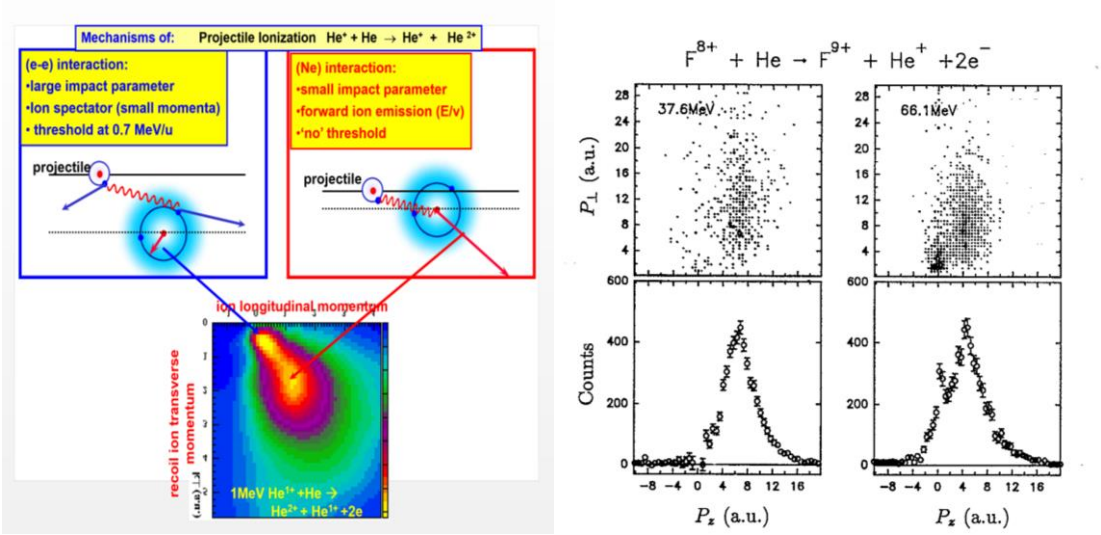

Figure 24: Recoil-ion momentum plots for projectile ionization. Left: 1 MeV He^{1+} on He [67]. Right: 37.6 and 66.1 MeV F^{8+} on He. Density Plots and corresponding projections, the z-component is the longitudinal momentum axis [68].

4.3. Momentum spectroscopy in high-energy heavy ion atom collisions

A further benchmark experiment by Moshammer et al. [69] demonstrated the high resolution power in measuring Q-values and deflection angles. In 3.6 MeV/u Ni^{24+} on He-collisions the full kinematics of the ionization process was measured by a recoil-ion electron coincidence. In figure 25 the sum-momentum of the electron and the He^{1+} recoil-ion is presented as function of the longitudinal momentum. More than 90% of all electrons are ejected in forward direction and their momentum is mainly balanced by the backward recoiling He^{1+} ion showing that binary projectile-electron collisions are of minor importance. The full width half maximum (FWHM) is 0.22 a.u., which corresponds to a

relative projectile energy loss of $\Delta E/E_p = 3.4 \cdot 10^{-7}$. The obtained resolution in the transverse momentum corresponds to a resolution in the deflection angle below 10^{-7} rad. In this publication for the first time the acronym COLTRIMS was defined.

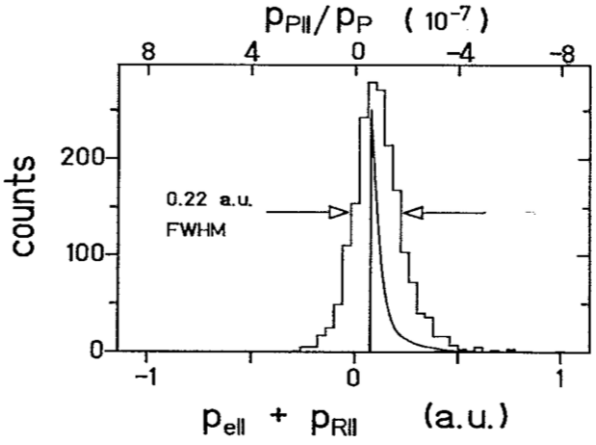

Figure 25: Sum of electron and recoil-ion longitudinal momentum (in atomic units). Upper scale: $\Delta p_{r\|}$ relative to the incoming projectile momentum p_p. The histogram shows the experimental data and as a full line results from CTMC theory (normalized) [69+70].

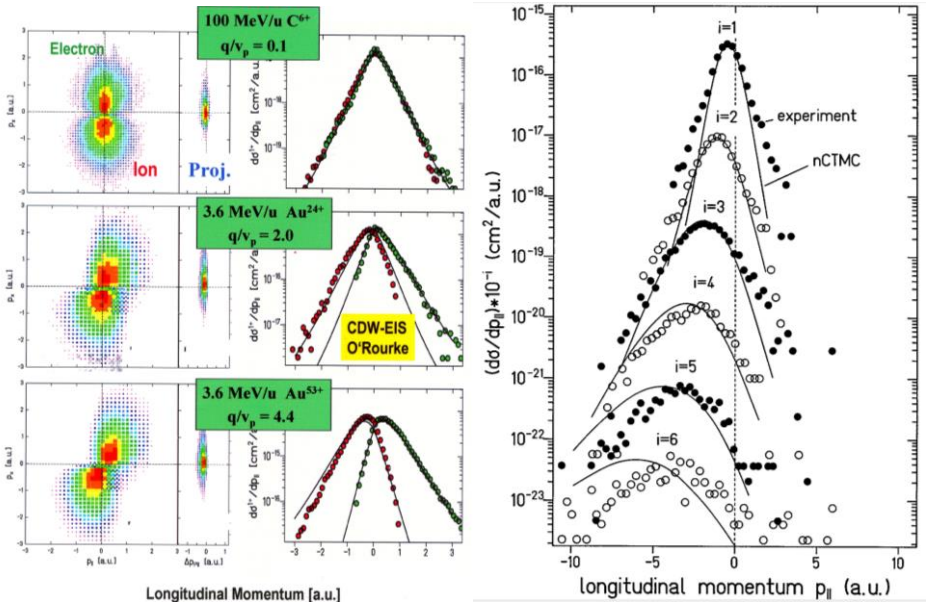

Figure 26: Left side: electron-recoil ion momentum plots for heavy-ion impact on He measured in single event coincidence (projected on the plane of incoming projectile - recoil momentum vectors). The plotted projectile momentum vector is the sum of recoil ion and the electron momentum vectors. In the middle the projection of the projectile momentum change is plotted in a.u. [70]. Right: projections of the recoil-ion momentum in multiply ionizing 5.9 MeV/m U^{65+} on Ne is plotted (in a.u.) [70].

In the nineties, the GSI group of Ullrich and Moshammer in cooperation with the Frankfurt group and partly with the CAEN group of Amin Cassimi explored in several research projects the mechanisms of multiple ionization of rare gas atoms in high-energy heavy ion impact. The C-REMI method allowed visualization even for GeV projectiles the energy loss at the few eV level (below 10^{-8} precision) as function of the projectile deflection angle. In figure 26 examples of such data are shown for He (left side) and Ne (right side) as a target [69-75]. At higher projectile velocities the momentum distribution of the ejected electrons and of the recoil ions [73] becomes more photon-like. The projectile provides by virtual photon interaction the energy for the ionization process. With higher projectile charge and slower the projectile velocity the electrons are increasingly ejected in the

forward direction. In another kinematically complete benchmark experiment on single ionization of He in collisions with 100 MeV/amu C^{6+} the collaboration found small but significant discrepancies between experiment and theory which were interpreted to be the result of higher-order effects in ionization [76]. Today, the puzzling contribution of such presumed higher-order contributions remains a matter of discussion.

4.4. Single-Photon ionization

Since 1993 the C-REMI technique also contributed strongly to the field of single-photon induced ionization processes. At HASYLAB/DESY-Hamburg and the ALS/LBNL-Berkeley first experiments with C-REMIs were performed. The C-REMI apparatus installed at Berkeley in the group of Michael Prior was mainly funded by the Max Planck Forschungspreis (200.000 DM) awarded together to Cocke and Schmidt-Böcking in 1991. Additionally, Kansas State University provided fellowships for PHD students and Postdocs. LBNL supported the Berkeley-KSU-Frankfurt collaboration with electronic and computer equipment.

The first achievement was the measurement of the ratio R of He^{2+} to He^{1+} by single photon ionization [77]. The absolute value of this ratio was debated since the methods did not allow a reliable calibration of detection efficiencies. The C-REMI approach had the advantage that the He^{2+} and He^{1+} ions were simultaneously recorded and hence the product of photon beam intensity times target thickness and geometrical solid angle was identical, only the detection efficiencies of the channel-plate detector for He^{2+} and He^{1+} could differ.

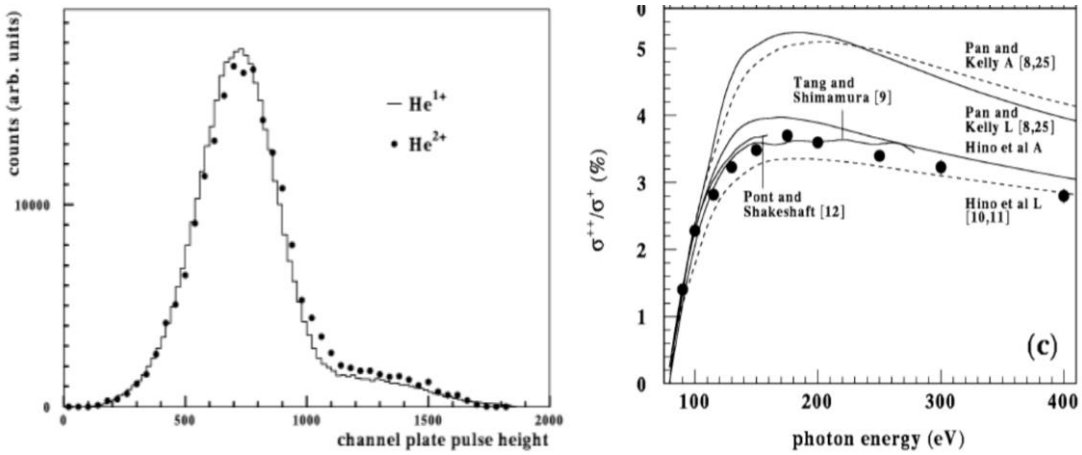

Figure 27: Left: pulse height distribution from the channel-plate detector for He^{1+} and He^{2+}. Right: the ratio R full circles from Dörner et al. [77] as function of the photon energy.

Since the height of the detector signal was recorded for every event, too (see figure 27 left side), it was evident that the efficiencies for both He charge states were identical, as well. When the ratios were analyzed, however, they did not agree on absolute scale with the standard data available in the literature at that time. The data by Dörner et al. were about 30% lower than the "official" numbers published as reference values. Thus, Dörner et al. began a long search for possible unknown systematic errors in their data analysis. On a meeting at RIKEN/Tokyo in 1995 the Dörner et al. data were compared to new theoretical calculations of Tang and Shimamura. These experimental and theoretical data agreed nicely on an absolute scale within their error bars. Consequently, both were immediately published. The "photon ionization community" reacted friendly and acknowledged immediately that the standard data used so far were, for an unknown reason, increased by 40% and could be wrong. The new published data were then accepted as reliable reference.

The ratio of the total ionization cross sections of Helium occurring due to the photo-ionization and the Compton effect was another fundamentally important problem in photon

physics at these times. The traditional methods of ion counting could not distinguish by which mechanism the atom was ionized. Both processes, however, differ in their recoil-ion momentum. In case of the photo effect the momentum p_e of the ejected electron is fully balanced by the recoil-ion momentum with $p_{recoil} = - p_e$ (see figure 28, left side) [78]. In case of the Compton effect the recoil ion acts only as a spectator and its final momentum peaks at zero (see figure 28, right side) [78]. Using the C-REMI approach, these different momentum distributions could quite easily be measured and separated. For photoionization one obtained, furthermore, information on two-electron correlations were the second He electron is simultaneously excited to higher n states (see figure 28, left side, rings of smaller electron momenta).

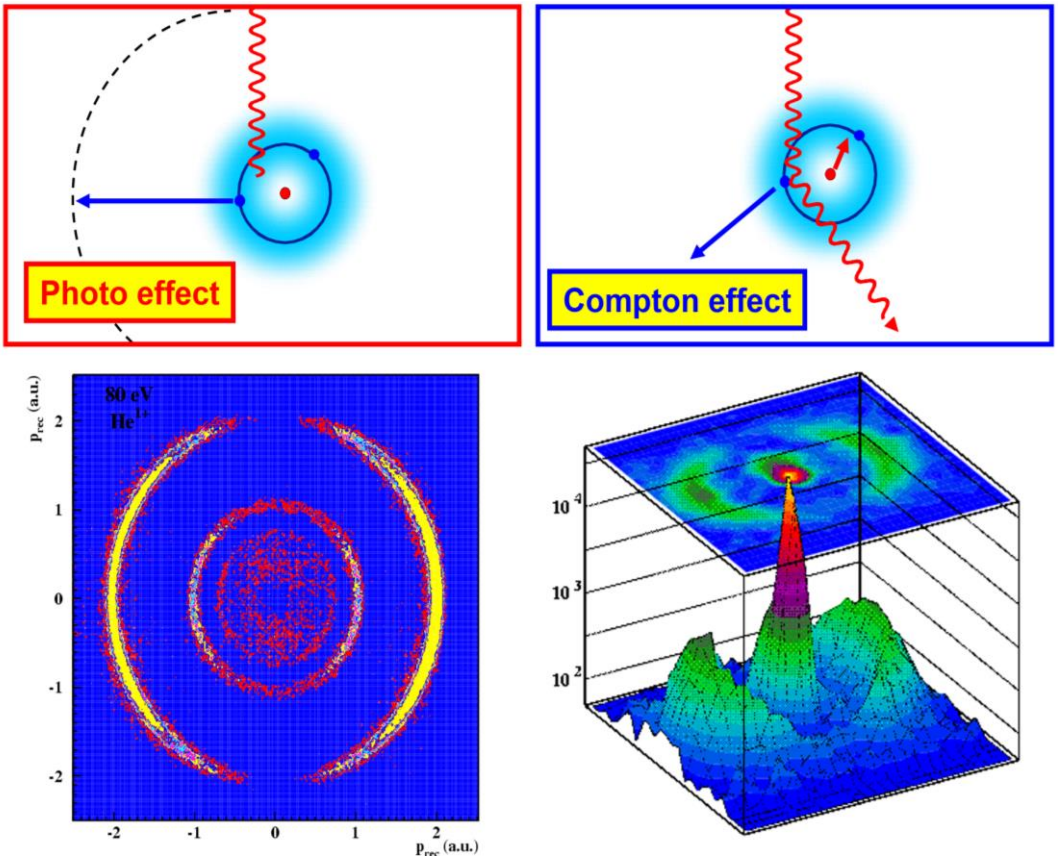

Figure 28: C-REMI measurement of He photon ionization [78]. Left: Photo effect, right: Compton effect [79].

Correlated two-electron processes, like the double ionization of He by a single photon, were, in the nineties very hot topics in the field of photon physics performed at synchrotron machines. Pioneering, fully differential data on the subject were measured by Volker Schmidt's [80] and Alan Huetz`s groups [81] by performing electron-electron coincidences. They used traditional electron spectrometers which had compared to C-REMI very small solid angles (resulting in a coincidence efficiency below 10^{-6}). The C-REMI approach has a coincidence efficiency of almost 50% and could image in quasi “one shot” the complete differential distribution (see figure 28, left side). Thus C-REMI revolutionized the field of double ionization processes by photon impact. Even the multi-TOF electron spectrometers of Uwe Becker [82] did not reach the C-REMI coincidence efficiency.

The first fully differential He double ionization data for circularly polarized photons were measured by Volker Mergel, Hiroshi Azuma and Matthias Achler [85] at the synchrotron machine at Tsukuba. Figure 29 shows the momentum distributions of one electron with respect to the momentum vector of the other electron for linearly polarized photons. In

figure 30 the same plot is shown for circularly polarized photons. The asymmetric, chiral electron emission patterns are clearly visible in the distributions.

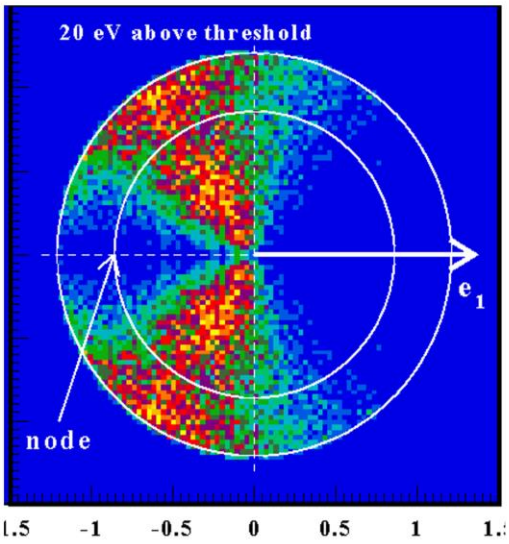

Figure 29: Fully differential He double ionization data for linearly polarized photons of 79 eV. The momentum distribution of one electrons is plotted with respect to the momentum vector of the other electron [83+84].

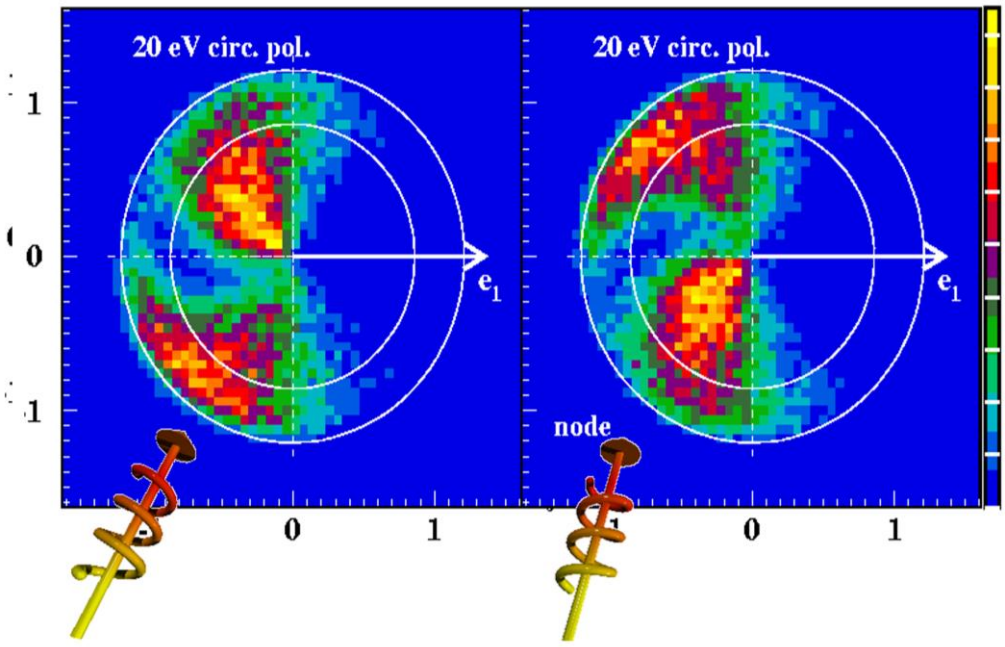

Figure 30: Fully differential He double ionization data for circularly polarized photons of 99 eV. The momentum distribution of one electron is plotted with respect to the momentum vector of the other electron [82].

4.5. Saddle point ionization mechanism in slow ion-atom collisions

In slow ion-atom collisions the mechanism of so-called saddle-point emission played an important role in the ionization process. Even when the projectile velocity was so slow that in a binary projectile nucleus-target electron collision the electron cannot be knocked out, theory predicted that the electron can be promoted to the continuum via quasi-molecular orbitals. Riding finally in the middle of the two nuclei like on a saddle, the electrons end up in the continuum in forward direction with about half the projectile velocity. Using the C-REMI approach Reinhard Dörner and Michael Prior [86] investigated, at the Berkeley ECR ion source, the ionization process in slow p on He collisions, measuring the recoil-

ion momentum vector in coincidence with two momentum vector components of the ejected electron. Because of the conservation of total momentum and total energy, the collision dynamics is kinematically fully defined. The surprising result was that the electrons did not ride on a saddle but their emission was kinematically steered by angular momentum conservation. The maxima of the “banana”-like electron distributions (see figure 31) vary in emission angle as function of projectile velocity. These shapes are centered in the nuclear collision plane.

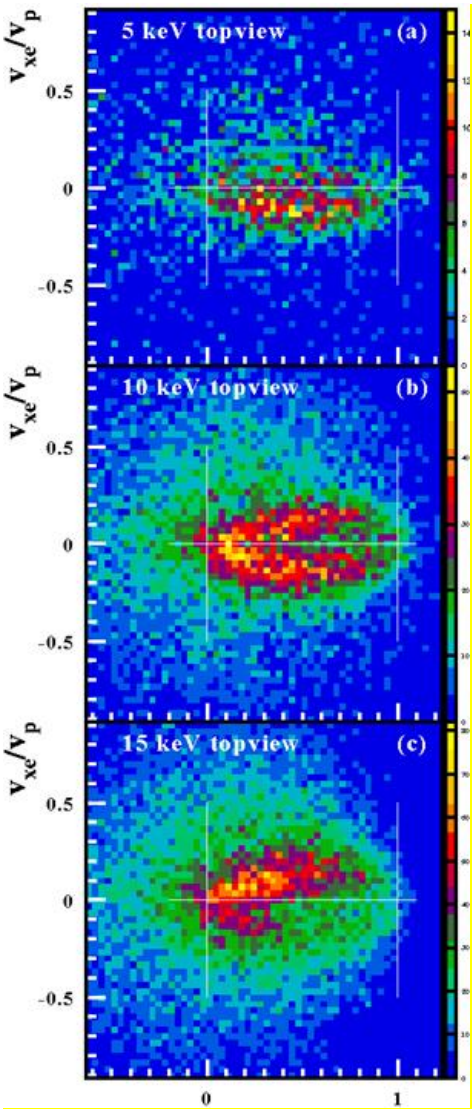

Figure 31: Electron momentum distributions projected on the nuclear scattering plane for so-called "saddle point electron emission" in slow p-He collisions [86]. The momenta are plotted in relative units of the electron velocity (v_p is the projectile velocity).

4.6. Visualization of virtual contributions to the He ground state

In 1983 Eric Horsdal Pedersen and Charles Lewis Cocke at KSU [87] and in 1986 Reinhold Schuch in Heidelberg [88] could verify, by examining the scattering angle dependence of the transfer ionization process in 7.4 MeV p + He => H° + He^{2+} + e collisions, the existence of the Thomas ionization mechanism [89]. These findings triggered great attention on the Thomas process in the whole atomic physics community. In this process the projectile nucleus can kick the bound He target electron 1 in a binary collision under 60°. On its way to the continuum electron 1 collides in a subsequent binary process with the second electron 2, thus one electron is ejected under 90° in the laboratory system and the other electron under 0°. This forward going electron is then captured by the parallel moving proton

projectile resulting in He-double ionization. These billiard like two-step processes require that the projectile is deflected under the angle of $\delta_p = 0.55°$. Thus the He double ionization as function of δ_p should show a peak structure at 0.55°. Varying the projectile velocity Horsdal-Pedersen found that this maximum gets even more pronounced when the projectile velocity increases [90]. Theory, however, predicted a v_p^{-11} law [91]. Therefore, the question arose, is the peak structure at about $\delta_p = 0.55°$ really related to the Thomas process?

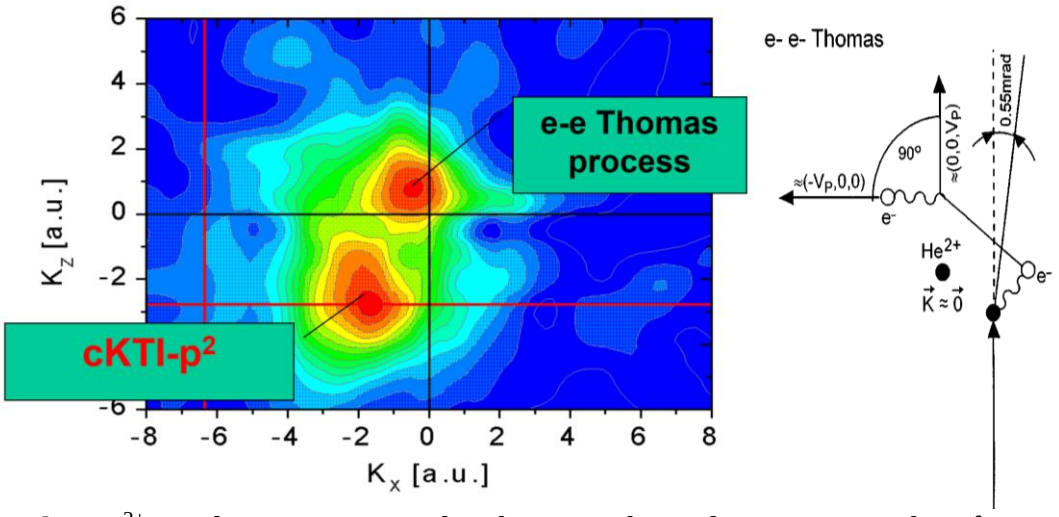

Figure 32: He^{2+} recoil-ion momentum distribution in the nuclear scattering plane for 1 MeV on He transfer ionization process [92]. Right: The kinematics of the Thomas process.

In the PHD work of Volker Mergel the complete kinematics of the transfer ionization process in fast proton He collisions was measured by an H° and He^{2+} coincidence using a C-REMI [92+93]. Determining the He^{2+} momentum vector and the H° transverse momentum components, the kinematics is fully controlled. In figure 32 the measured He^{2+} recoil-ion momentum distribution is shown for protons of 1 MeV scattered under 0.55°. Surprisingly two strong maxima appear. One at $p_{rec\parallel} = + 0.8$ a.u. and $p_{rec\perp} = - 0.5$ a.u. which coincides with the expected Thomas peak position, but the second unexpected maximum (named cKTI-p^2) at $p_{rec\parallel} = - 2.8$ a.u. and $p_{rec\perp} = - 1.8$ a.u. indicates there must be another, so far, unconsidered mechanism enabling transfer ionization at $\delta_p = 0.55°$. The analysis of the kinematics showed that one electron is captured at large impact parameters into the H° ground state (Brinkmann Kramer mechanism) and the second He electron is emitted backward under about 135° with a momentum of approx. 3 to 4 atomic units. Mergel found the total cross section for this maximum follows a $v_p^{-7.4}$ law, thus, compared to the Thomas peak, it is the dominant transfer ionization channel at higher projectile velocities.

According to multi-configuration theory the He ground state contains a small contributions of 1% - 2 % of the so-called pseudo-states like p^2, d^2 etc.. In the p^2 pseudo-state the two electrons have opposite angular momenta and in a He atom at complete rest the target nucleus balances at any moment in a fully entangled motion the sum electron momentum to zero. If one electron in the p^2 state is captured at large impact parameters by the proton the electron 1 velocity and its direction are identical with that of the moving proton. In this moment the other He electron in the p^2 state and the nucleus must move backward. Although electron 2 is in a p pseudo-state it must enter in this moment a real continuum state. In figure 33 the angular distribution of the emitted electron 2 is shown in comparison to theoretical predictions. The agreement between experimental data and theory is rather good giving confidence that the presented explanation is valid. It is really surprising that such “virtual states” can be visualized in the real experimental environment. Even the kinematics at a given virtual excitation energy is visible. These tiny contributions represent an extremely small part of states contributing e.g. to the Lamb shift. However, the C-REMI approach is sensitive enough to probe the kinematics of such very small fractions of virtual states.

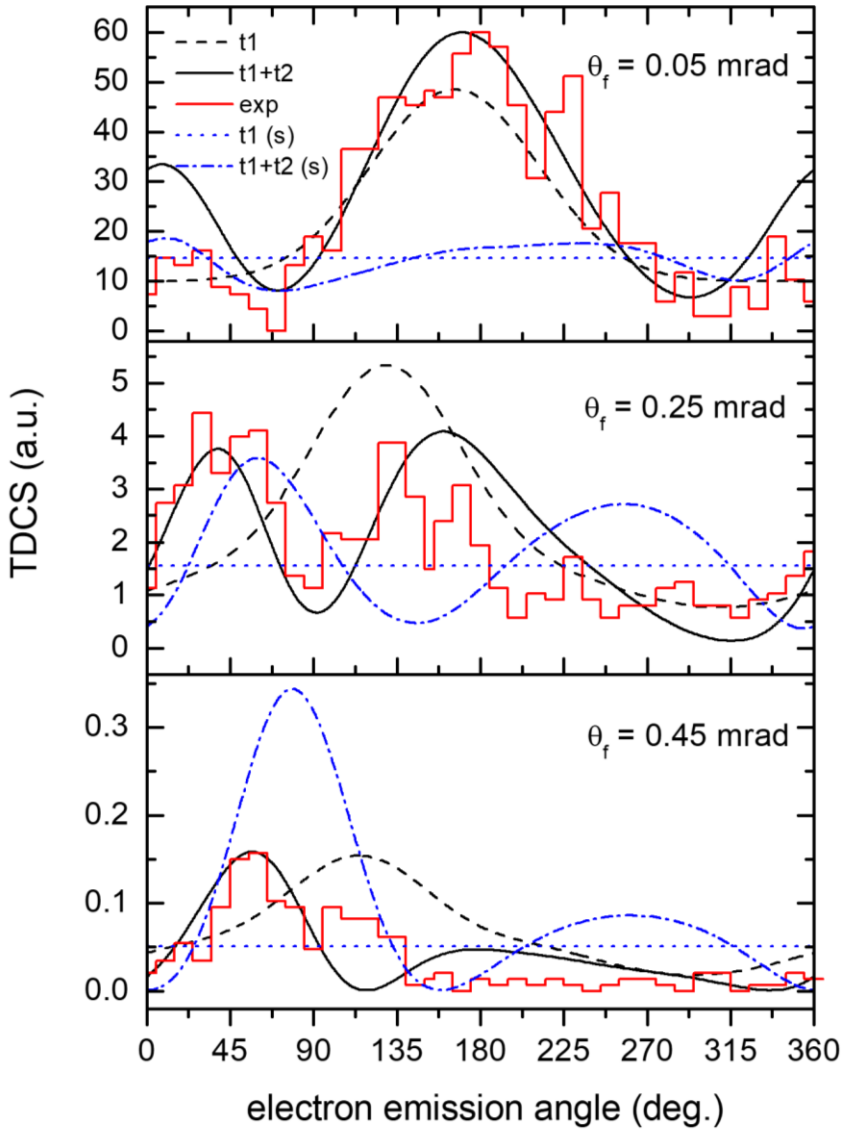

Figure 33: Triple differential cross sections of Transfer Ionization in 630 keV p on He collisions and 20 eV kinetic energy of the electron corresponding to maximum two (figure 32) for three different projectile scattering angles. The black solid line is the theoretical prediction for the non- s^2 contributions. Theory and experiment are relatively normalized [94-96].

5. Milestone discoveries

The C-REMI had grown into being an established experimental approach to study dynamics in quantum systems in Physics, Chemistry and other fields in the mid-nineties. In several hundred laboratories worldwide C-REMI systems are operating, partially commercially purchased or self-made. By using the C-REMI imaging technique many groups have produced numerous milestone discoveries. However, reference to all of these in this review paper would exceed the purpose of this article. To present all milestone results produced by the authors of this paper would also overshoot the capacity of this review. Thus only a few of those achieved by the authors of this paper are presented here.

5.1. Multi-Photon processes - experimental verification of re-scattering mechanism

To explain the processes underlying multiple ionization and the high double ionization probability of He and other rare gases in intense Laser pulses, Paul Corkum (1993) and

Kenneth Kulander (1995) proposed the so-called re-scattering model [97]. There, emitted electrons are oscillating in the strong Laser field and are re-scattered at their parent atom. At that time period, the strong-field community did not have the proper detection device to verify experimentally this hypothesis. To visualize the dynamics of this re-scattering process one had to measure the momenta of two or more ejected electrons (and if possible of the recoil ion, too) in coincidence with high resolution. Thus, two independent collaborations, which stayed in very close contact, performed, in parallel, such coincidence experiments.

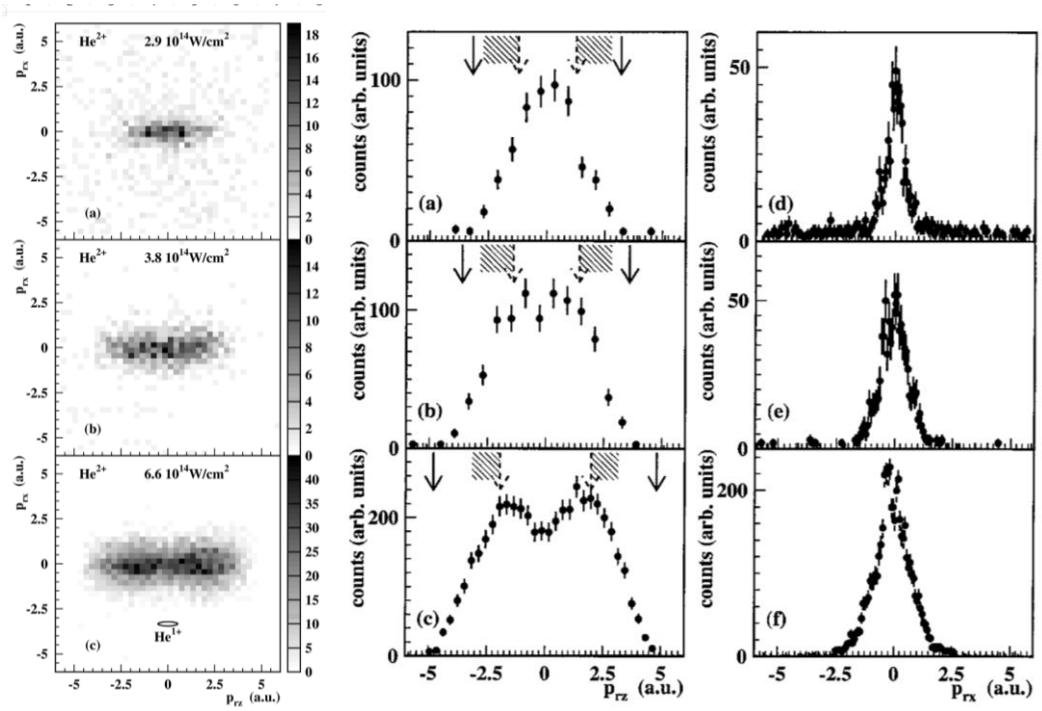

Figure 34: Left column: He^{2+} recoil ion momentum plots for three different Laser intensities. The horizontal axis is parallel to the direction of the electric field of the Laser. The vertical axis is given by the Laser propagation. The small ellipse in (c) shows the half-width of the He^{1+} distribution. Middle and right column: projections of the plots of left column onto the horizontal and vertical axis. The arrows indicate the maximal momentum due to the ponderomotive energy. The dashed areas represent the in the rescattering model calculated values [99].

The collaborations consisted of, first, the Heidelberg group of Joachim Ullrich and Robert Moshammer, who supplied a C-REMI and joined the Laser group of Wolfgang Sandner and Horst Rottke in Berlin [98] and, second, the Frankfurt group of Reinhard Dörner and Thorsten Weber supplying the C-REMI and joined Harald Giessen in Marburg providing the Laser [99]. Presented here, in figure 34, are only the data of the experiment performed in Marburg [99]. For 220 femtosecond long Laser pulses of 800 nm wave length at intensities of 2.9 till 6.6 • 10^{14} W/cm^2 the He^{1+} and He^{2+} recoil ion momenta were simultaneously measured.

The surprising result was: the He^{1+} recoil momenta are strongly directed parallel to the Laser electric field with much smaller momenta in the transverse direction- even much less than in case of single photon ionization. The He^{2+} recoil ion momenta are in transverse direction of the Laser field similar to the He^{1+} momenta, but parallel to the field 5 to 10 times larger. In addition, they show two maxima separated by a minimum at zero. In case of single photon ionization, the recoil-ion momentum distribution reflects mainly the momentum distribution of the electron in its initial bound state, in case of double ionization by a single photon it reflects possible electron-electron correlations in the initial state. But the He^{2+} recoil momenta never exceed the He^{1+} recoil momenta by more than a factor two.

Thus, in case of Laser induced double ionization only Corkum's re-scattering mechanism can explain the observation of such large He^{2+} recoil-ion momenta parallel to the Laser field. In his model, the electron can gain in the Laser field a high ponderomotive energy yielding finally a large recoil momentum. Similar work can be found in [100]. This work provided the experimental proof that the re-scattering process does explain the dynamics of the double ionization in intense Laser field and that both electrons act coherently.

The two-Laser pulse pump-probe technique is well established to measure timing (delays) in the femtosecond-regime. However, using this technique in combination with the C-REMI coincidence imaging one can – despite of employing pulses of many femto-second duration – obtain timing information in the atto-second regime [101]. In 2005 the collaboration between Paul Corkums group in Ottawa and Reinhard Dörners group in Frankfurt performed such timing measurements in the attosecond regime [102] by using the Laser pump-probe scheme for ionization and for detection of the emitted low-energy electrons the C-REMI approach. The first Laser pulse aligned a nitrogen molecule and the subsequent strong probe pulse ionized the molecule. Recoil ions and electrons were detected in coincidence and from the measured recoil-ion momentum vectors the spatial alignment of the molecule in the lobaratory frame was determined. They found that both electrons did exit the molecule more likely in the same direction when the polarization of the probe pulse was parallel to the direction of the alignment. Double ionization was less probable and takes longer for the perpendicular alignment (a few hundred atto-seconds longer).

In a coincidence experiment where several momentum vectors of the emitted fragments resulting from the same reaction are detected, one can deduce from the relative angular vector directions phase differences and thus determine relative time delays. Thus a multi-coincidence momentum-imaging approach like the C-REMI method is the key to explore atto- and even zeptosecond dynamics by measuring streaking effects of Laser fields on the momenta of emitted particles. This technique allows to measure, as outlined before, time differences shorter than present Laser pump-probe technique can resolve. Ursula Kellers group at the ETH Zürich in cooperation with the Frankfurt group performed such measurements on the tunneling times in He [103]. From the observed phase shifts in the recoil ion and electron-momentum distributions it was claimed that the tunneling process takes a finite time of about 20 as, triggering strong debate on the topic in the following years. Many more important experiments have been performed in recent years in the field of ultrafast processes. References [104-113] are a few selected papers on this topic.

5.2. Single Photon Ionization of Molecules

Since Max von Laues X-ray diffraction experiment in 1912 in Munich the scattering of X-rays and electrons has been used to explore the structure of molecules. In all these studies the molecules had to be in an ordered structure (e.g. crystal) to know the molecular orientation. The C-REMI allows the study of freely moving non-oriented molecules in a gas phase. By performing multi-hit electron-ion coincidence measurements the orientation of the molecule with respect to the detection device is determined from the ionic momenta. The first successful experiments employing the idea of inferring molecular orientation from fragment emission directions were performed by Eiji Shigemasa et al. in 1995 [114] and Franz Heiser et al. [115]. They used traditional electron spectrometers with small solid angles and had to scan the electron energy. Thus these measurements were very time consuming and gave results only for discrete angles.

The first such experiments on single photon ionization of simple molecules using a C-REMI were performed in Berkeley and in parallel in Paris. When the Advanced Light Source (ALS) started operation in 1993 Michael Prior of the LBNL in Berkeley, Charles Lewis Cocke and his group at KSU together Reinhard Dörner and Horst Schmidt-Böcking

from the University Frankfurt installed a C-REMI system at the LBNL, which could be used either at the ECR source in the 88" cyclotron building or the ALS. At the Oji-Workshop (Atomic Mol. Photoionization, September 1995) in Tsukuba, Paul Guyon and Horst Schmidt-Böcking arranged to use the C-REMI coincidence system with position-sensitive detectors to perform collaborative experiments on single photon ionization of molecules at the Paris synchrotron. Paul Guyon's group had used so far the ZEKE technique [116] to study such processes. This method had extremely small coincidence efficiency because of tiny solid angles accepted in the direction transverse of the photon beam. The Paris group provided the photon beam and gas target, the Frankfurt group the detection and data acquisition system.

First experiments on single photon ionization of simple molecules and their fragmentation by photo ionization started at Berkeley in the late nineties with Alan Landers (at that time at KSU) and Thorsten Weber (at that time in Frankfurt) being the responsible investigators. Alan Landers et al. [117] measured the two fragment ions, their charge state and the photo electron upon C-K-shell ionization of CO in coincidence. Following the inner-shell photoionization, Auger electrons are emitted after a short delay leading to a Coulomb explosion of the molecule. Therefore, the ions' emission directions correspond to the molecular orientation at the instant of the photoionization and, from the ions' relative momenta, the kinetic energy released in the fragmentation was also obtained. For this concept to work, it is important that the delay between the fragmentation of the molecule and the initial photoionization is short compared to possible molecular rotation periods. As the photoelectron was measured in coincidence, its angular emission distribution with respect to the molecular axis was obtained.

Figure 35 shows the angular distributions of the C-K-shell photoelectrons in a polar representation, where the distance of a data point to the center of the plot represents the intensity. The double arrow with the two balls in each plot indicates the direction of the photon polarization and the molecular orientation. With the help of theory [117] details of the three-dimensional molecular potential could be deduced from such measurements. Parallel in time to the measurements by Alan Landers and Thorsten Weber et al., also the group of Anne Lafosse et al. located in Paris in cooperation with the Frankfurt group performed such measurements using C-REMI approach [118].

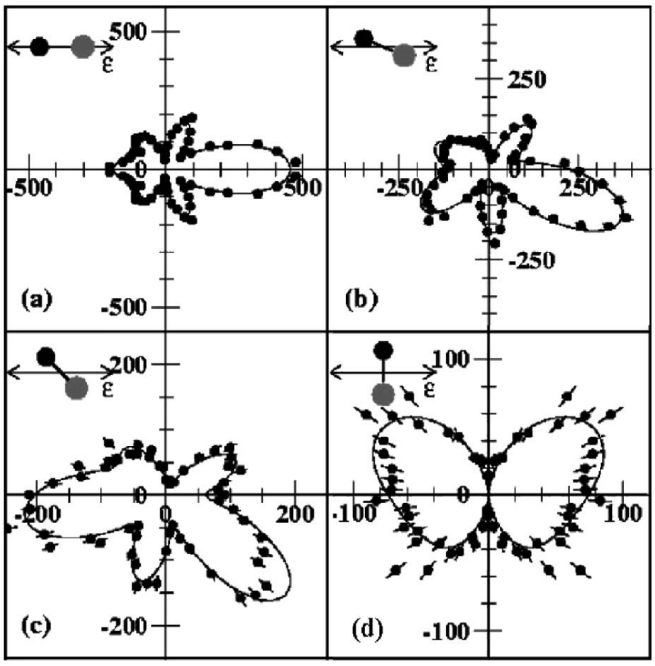

Figure 35: Polar distribution of 10.2 eV photo-electrons in the frame of the CO molecule (small ball carbon, large ball oxygen). The solid line represents a fit to the data [119].

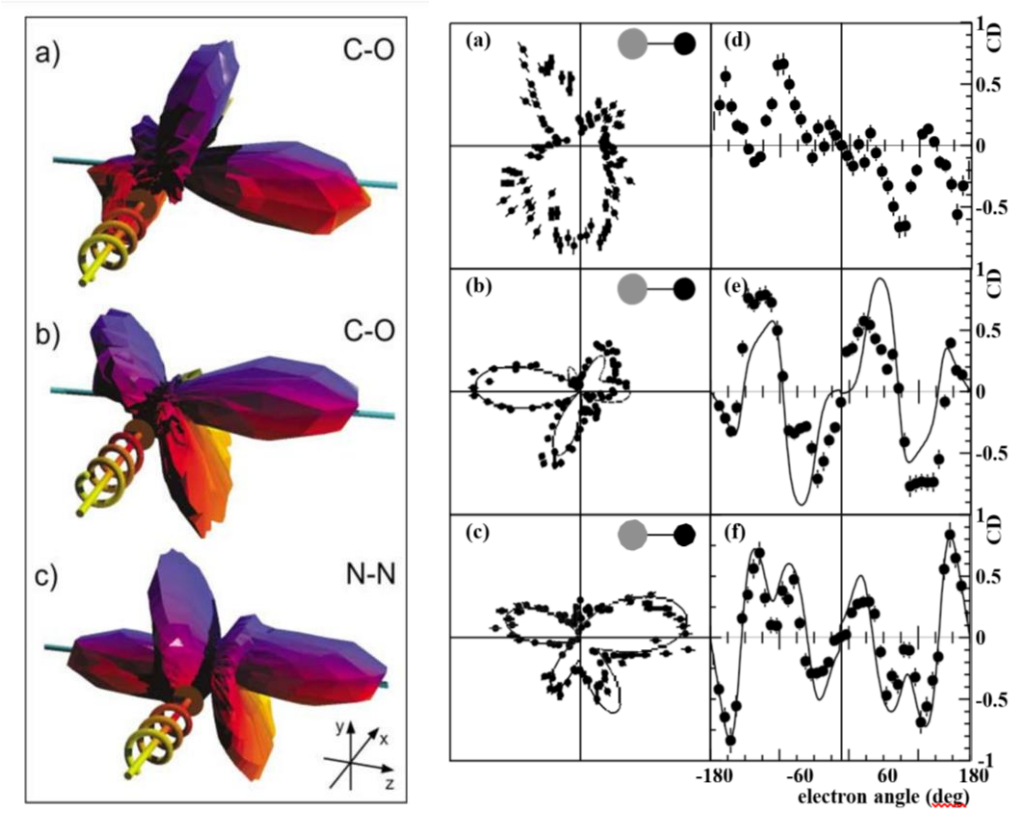

Figure 36: Left: three-dimensional molecular-frame angular distribution of C and N 1s photoelectrons emitted from CO and N_2 molecules. The molecular orientation is indicated by the green line and the label. The handedness of the photons and their impact direction are indicated by the spirals. Right (a,b,c): projections of the data (left) on the plane perpendicular to the photon propagation, right (d,e,f): extracted circular dichroism [119]. The corresponding distributions for different molecular orientations can be found at: www.atom.uni-frankfurt.de/research/20_synchrotron/30_photon_molecule/20_K-shell_CO_N2/.

More photo-ionization measurements of molecules have been performed in the last two decades using the C-REMI approach (see, e.g., [119-123]). Jahnke et al. [119] performed corresponding measurements using circularly polarized photons providing first full 3-dimensional molecular frame photoelectron angular distributions, as shown in figure 36, left. Furthermore, they found a strong circular dichroism (CD) in the photoelectron emission (see figure 36, right).

At the ALS in Berkeley Thorsten Weber and Michael Prior, in collaboration with the KSU and Frankfurt groups performed several further studies [121-122] including the measurement of the complete photon induced fragmentation of D_2. With the support of the theory groups of Bill McCurdy in Berkeley and Fernando Martin in Madrid, fundamental information on symmetry breaking in the D_2 fragmentation processes was deduced.

5.3. Multi-fragment vector correlations in inner shell single-photon ionization processes of atoms and molecules - dynamics of entangled systems

This kind of measurement approach delivers insight into two or more new fundamental aspects of atomic physics research:
1. the study of oriented very short living excited atomic and molecular ionic configurations, which can never be produced by any other preparation technique (e.g. like Laser orientation and excitation).

2. the study of dynamical entanglement in sequential cascading decay processes, exploring memory effects and dynamically induced symmetry breaking.

The multi-coincident fragment detection from a Coulomb-exploding molecule can provide insight into fundamental aspects of entangled many-particle Coulomb dynamics. In figure 37 the scheme of such a multistep process is indicated. From left a circular polarized photon with a well-defined energy (momentum) and angular momentum is absorbed by a two-atom molecule and creates a K vacancy thus a low energy photo electron is emitted (step 1). The electron-momentum vector (three dimensions) is measured. After a short delay, in step 2 a K-Auger electron (probably from the atom where the K vacancy was created) is ejected. The momentum vector of the Auger electron is measured too. Vacancies and excitation energy can be shared by the two atoms. In the following steps 3 to 5 more Auger electrons are emitted. From the measured Auger electron momenta, the experimenter knows the electron energy and thus the time sequence of the different steps. The delay times, however, remain unknown. Finally, with increasing degree of ionization the molecule undergoes Coulomb explosion. Measuring the ionic momenta and its final charge state the experimenter has a full dynamical control on the orientation of the molecule and on the dynamics of the fragmentation (i.e. dynamic entanglement). Finally, in this example of fragmentation using C-REMI the experimenter has measured all together 24 momentum components and two charge states (the angular momentum vector of the photon, 5 electron momentum vectors and two momentum vectors of the ionic fragments).

Comparing this approach with the two-pulse Laser Pump & Probe technique one can "pump" (ionize) a molecule by a single high energy circular-polarized photon with subsequent photo-electron and multiple Auger-electron emission (i.e., multiple probe technique MPT). The angular momentum of the system recoil-ion and photo-electron is identical with the one of the photons (assumption: the initial state of the molecule has no angular momentum) and is therefore known by the experimenter. In this way the experimenter "pumps" the molecule by single photon absorption without destroying the dynamic entanglement of the system. Thus this MPT establishes a new field in atomic and molecular physics allowing the investigation of extremely short-lived excited molecular states.

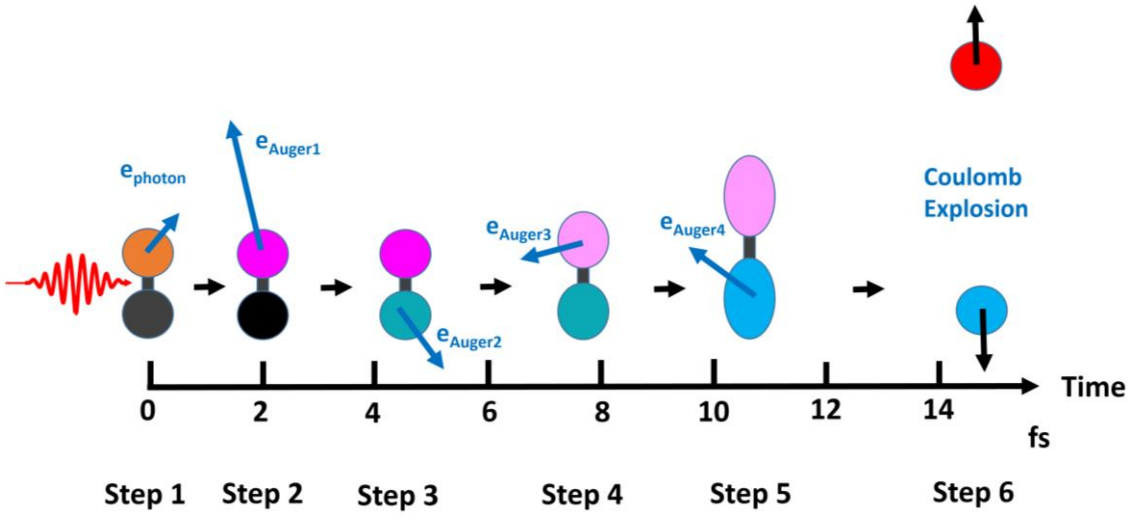

Figure 37: Scheme of a fragmentation chain with intermediate steps 1 to 6.

The MPT allows one to ask whether the delayed emitted electrons have a "memory" of the earlier fragmentation steps and whether any dynamically induced symmetry breaking (in

time or parity) may occur. From the measured vectors $\boldsymbol{L_\gamma}$ and $\boldsymbol{p_{fn}}$ ($\boldsymbol{L_\gamma}$ angular momentum vector of the photon and $\boldsymbol{p_{fn}}$ the momentum vector of the n-th emitted electron) one can define new dynamical coordinate systems, e.g. $\boldsymbol{L_\gamma}$ x $\boldsymbol{p}_{f1} = \boldsymbol{Z_{y1}}$ and $\boldsymbol{p_{f1}}$ **x** $\boldsymbol{p_{f2}} = \boldsymbol{Z_{12}}$ and plot the delayed electron-emission probabilities emitted in step 2,3,.. with respect to these new coordinates [122]. This new **pump & multiple-probe MPT** approach enables the investigation of fundamental dynamical processes in many-particle systems, like time or parity symmetry breaking. If time symmetry is broken then the distribution of the Auger electron of step 2 with respect to vector $\boldsymbol{Z_{Ly1}(+t)}$ ($\boldsymbol{p}_{e1}$ is the momentum vector of the photo-electron) and to vector $\boldsymbol{Z_{Ly1}(-t)}$ should be asymmetrical. This vector equation shows

$$\boldsymbol{Z_{Ly1}(-t) = L_\gamma (-t)\ x\ p_{e1}(-t) = (-)L_\gamma (+t)\ x\ (-)p_{e1}(+t) = +\ Z_{Ly1}\ (t)}$$

that in case of time inversion the vector does not change its sign.

Vector product	$t \to -t$	$\boldsymbol{r} \to -\boldsymbol{r}$
$\boldsymbol{Z} = \boldsymbol{A}_\gamma \times \boldsymbol{p}_{\text{ephoto}}$	$Z(t) = +Z(-t)$	$Z(\boldsymbol{r}) = -Z(-\boldsymbol{r})$
$\boldsymbol{Z}' = (\boldsymbol{A}_\gamma \times \boldsymbol{p}_{\text{ephoto}}) \times \boldsymbol{p}_{\text{K-Auger}}$	$Z'(t) = -Z'(-t)$	$Z'(\boldsymbol{r}) = +Z'(-\boldsymbol{r})$
$S = (\boldsymbol{A}_\gamma \times \boldsymbol{p}_{\text{ephoto}}) \cdot \boldsymbol{n}$	$S(t) = +S(-t)$	$S(\boldsymbol{r}) = +S(-\boldsymbol{r})$

Table 1: Vector products with respect to time and parity symmetries.

In [122] for 306 eV right and left handed photons on Carbon Monoxide CO the vector correlations between the 10 eV photo electron, the K-shell Auger electron (Carbon) and the singly charged ionic fragments were measured. Florian Trinter et al. [122] analyzed their coincidence data with respect to possible dynamically induced symmetry breaking. In table 1 some "dynamical" vector products are shown with respect to time and parity symmetries.

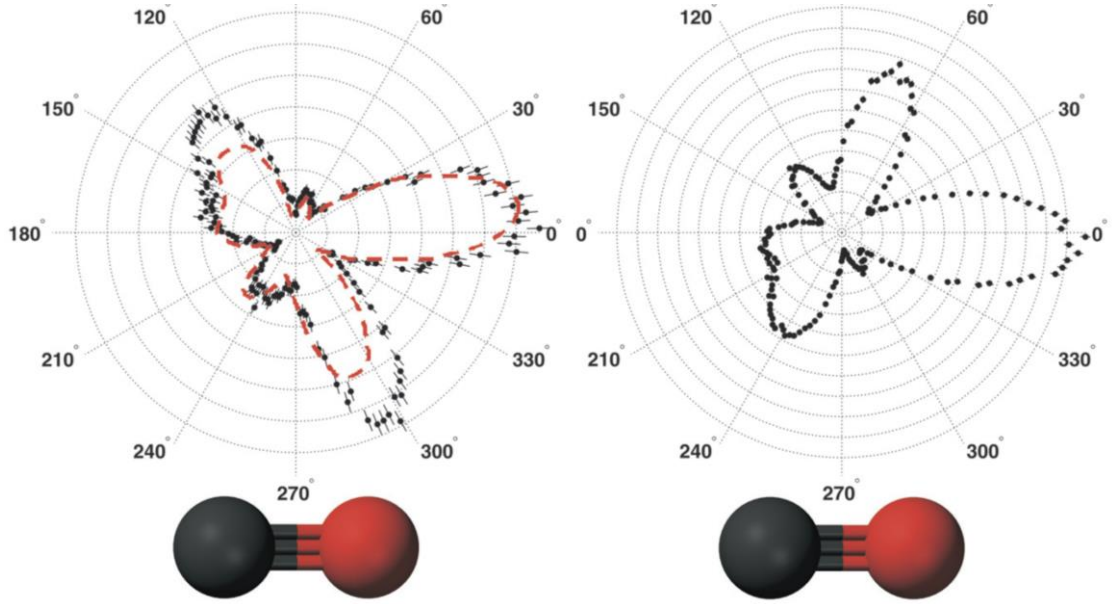

Figure 38: Polar plot of angular photoelectron distributions in the plane perpendicular to the propagation of the photon. Left: The red dashed line represents the distribution of the right side (right handed photon) but mirrored in time. The molecular orientation is indicated by the bar-bell with Carbon on the left. Only events with a KER value > 11 eV are selected, which ensures the axial recoil approximation [122].

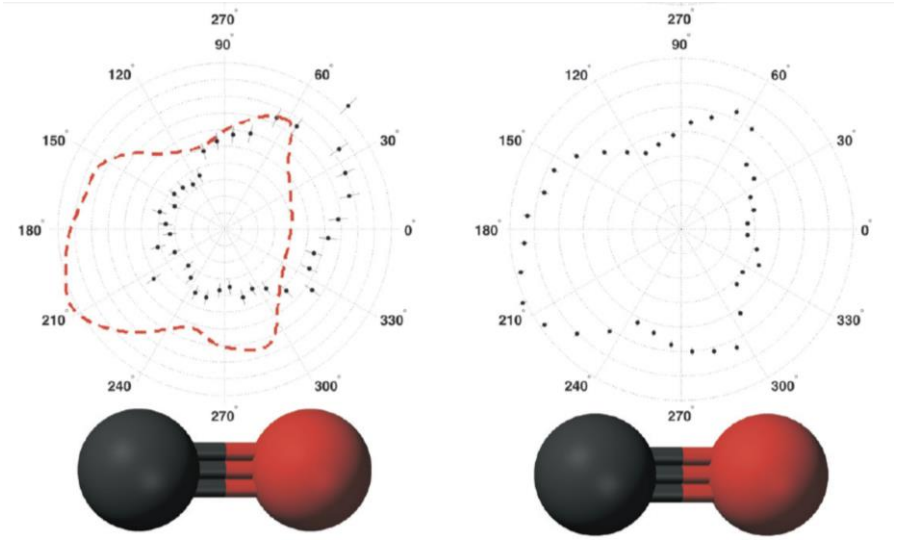

Figure 39: Polar plot of angular K-Auger-electron distributions in the plane perpendicular to the propagation of the left and right handed circular polarized photons. Left: The red dashed line represents the distribution of the right side (right handed) [122].

Trinter et al. [122] have analyzed also the K-Auger electron distributions for different conditions on the momenta of the emitted photo electrons for both left and right handed photons. In figure 39 the K-Auger electron distributions are shown for left and right handed polarized photons with the identical conditions on the photo-electron momentum vector (in the same planes as in figure 38). In case of complete symmetry with respect to dynamics both distributions should have the same shape. I.e. mirroring the time the corresponding distributions did not agree within the statistical error bars. However, these preliminary measurements do not allow within their error bars any reliable conclusion on time reversal we only assert that such fundamental aspects of quantum dynamics can be explored with the C-REMI approach in these kinds of measurements.

5.4. Single photon induced interatomic Coulombic decay

Electronically excited atoms or molecules decay by photon or electron emission. More than twenty years ago Lorenz Cederbaum et al. [124] predicted another very fast decay channel in loosely bound matter, where the excitation energy can be exchanged by means of a virtual photon between an excited atom and its neighboring atom. This decay channel was named "Interatomic Coulombic decay" (ICD). This process occurs in very weekly bound molecules. For example, the Ne dimer, which is a prototype system for ICD, is bound by the van der Waals forces with a binding energy of 2 meV at an inter-nuclear distance of 3.4 Å. First experimental evidence for the existence of ICD was reported by observation of slow electrons emitted in large photon excited Ne clusters by Uwe Hergenhahn und Thomas Möller [125]. ICD in Ne dimers can, however, be unambiguously identified by coincident detection of two Ne^{1+} fragments and the low-energy ICD electron. To yield a unique fingerprint of this ICD process, Till Jahnke and Achim Czasch have performed at BESSY II in Berlin a corresponding multi-fragment coincidence experiment using the C-REMI approach [126]. The photon energy was chosen such that only a 2s electron in one Ne atom could be ejected, but a subsequent Auger transition in the same ionized atom was energetically not possible. As ICD occurs, the excitation energy is transferred to the other atom of the dimer causing the ionization of its outer shell. The energy released in the process is shared by the ICD electron and fragment ions. Therefore, the total sum of the kinetic energies is fixed and can be used for an unambiguous identification of the ICD process. A scheme of the ICD process is shown in Figure 40. The quantitative values of the shared energies are plotted in a two-dimensional plot, KER energy versus electron energy in figure 41. The ICD feature forms a diagonal line depicting the constant

energy sum, as predicted for the ICD process. In [127-132] more recent work on the ICD process is presented.

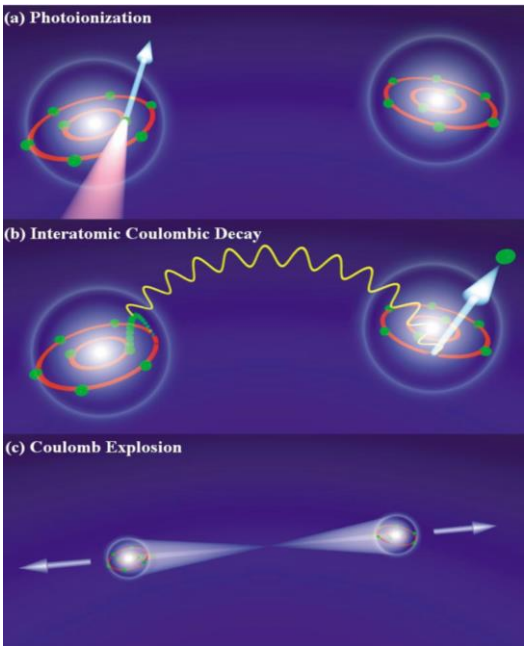

Figure 40: Scheme of the ICD process. a) Photoionization with ejection of a 2s photoelectron; b) Virtual photon transfer from the ionized atom to its neutral partner atom yielding the emission of 2p electron from the partner atom; c) Coulomb explosion of the doubly charged dimer [126].

The experimental proof and verification of the existence of the ICD process was only possible by the coincident detection of all charged fragments occurring in the process. Most hydrogen or van der Waals bound systems, most prominently liquid water, will often release or transfer energy via the ICD channel. A recent, comprehensive review on ICD can be found in [133].

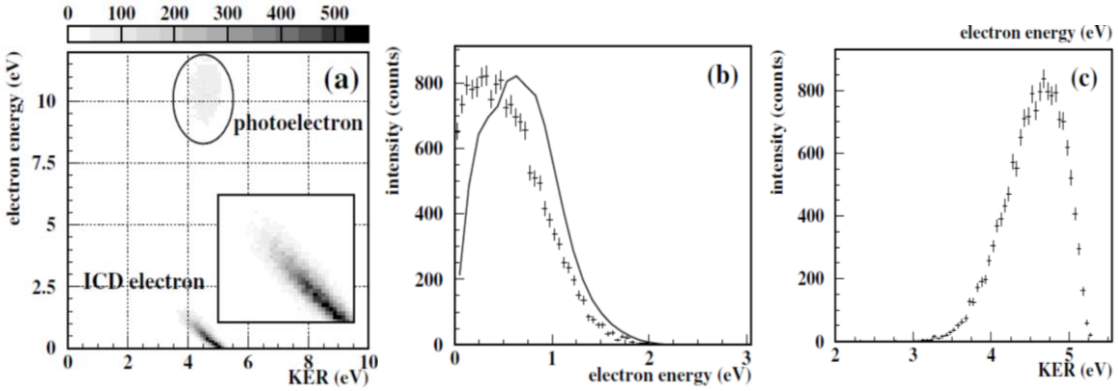

Figure 41: Left: Kinetic energy release KER of the Ne ions versus the energy of photo electron and ICD electron. Right: Projections of electron and KER value distributions [128].

In 2013 Florian Trinter et al. [132] have investigated the ICD process in van der Waals-bound HeNe molecules. B. Najjari and Alexander Voitkiv [134] predicted that in such molecules one of the atoms can act as a very efficient antenna to absorb photons. In case of HeNe, the ionization cross section is strongly enhanced (by a factor of 60) if the photons can first interact with the He atom. It absorbs the photon and in an ICD-process the energy is transferred to the neighboring Ne atom which is then ejecting an electron. In figure 42 (left side) the different steps of this process are shown. The measurement was performed with a C-REMI system detecting the emitted electron and ion in coincidence. Florian Trinter et al. have experimentally verified that a single atom can act as a highly efficient

antenna to absorb energy from a photon field and transfer the energy to a neighboring receiver atom within a few hundreds of femtoseconds. The resolved vibrational states of the resonance provided a benchmark for future calculations of the underlying energy transfer mechanism of ICD.

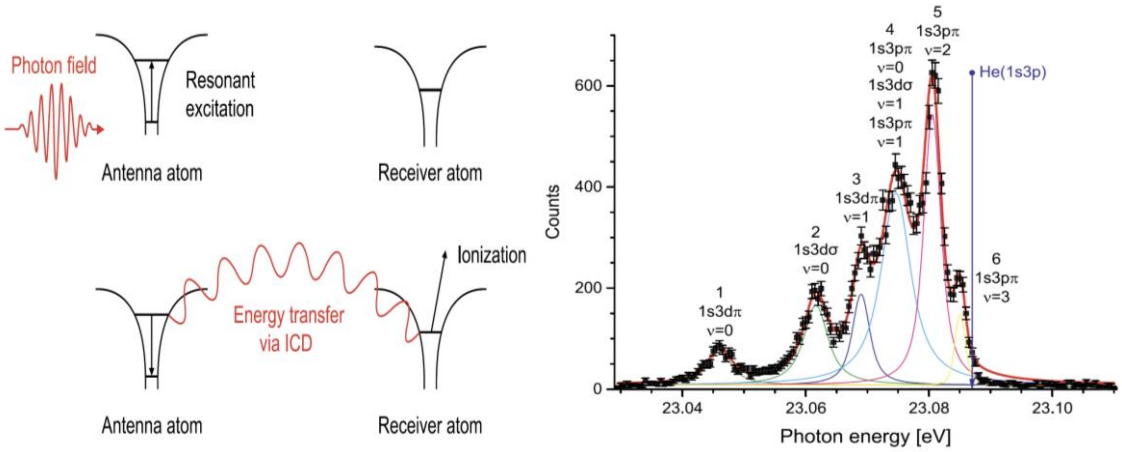

Figure 42: Left: Scheme of absorption and decay steps. The photon coming from left is absorbed by the He atom, which is resonantly excited into the 1s3p state. Before it can decay by photon emission the excitation energy is transferred via resonant ICD to the neutral Ne atom, leading to its ionization. Right: the photon energy was scanned over the range of the He resonance below the actual ionization threshold. The vibrational states of the molecule can be nicely resolved (see theory [136]).

5.5. Core-hole localization

Each atom or molecule represents one unified dynamical quantum state with a well-defined total energy, where all electrons together with the nuclei form by spin-orbit coupling one state with well-defined angular momentum and exactly ZERO total momentum in its own center-of-mass system - strictly conserved over varying time. Each atom or molecule is not a sum of single particle states, the experimenter cannot number and distinguish each electron e.g. as a specific K-shell or L-shell electron, which can be knocked-off to the continuum thus allowing an initial state localization of the ejected electron. One can only create an ionized atom/molecule in an excited new energy state with a K-shell or L-shell vacancy. In case of an inner-shell hole this vacancy may be localized for an extremely short time near the nucleus of one atom. In 2008 Markus Schöffler et al. [135] have been able to explore this open problem by investigating the symmetry in the angular emission distributions of photo- and Auger electrons emitted from molecular N_2. In their experiment, they measured the photo- and the Auger electron, as well as the emitted ionic fragments in coincidence. The emitted electrons yielded de-facto an ultra-fast probe of the shortly existing possible asymmetry of the electronic potential near both nuclei. Early theoretical calculations [136] suggested that even fully symmetric molecules consisted of asymmetric contributions in their ground-state in case of core-hole localization. This work resolved a decade of debate on possible core-hole localization with several experiments proving its existence and others concluding that core-holes are fully delocalized. The C-REMI work by Markus Schöffler et al. demonstrated, that the question of core-hole localization or delocalization is ill-posed.

It is not only the core-hole (or the corresponding photoelectron) that needs to be considered, but the whole molecule as such. The emitted photoelectron (and thus the core-hole) forms an entangled state with the Auger electron and the fragment ions of the molecule. Depending on the properties of the entangled partners, the properties of the core-hole changes, as well. It was shown in [135] that fingerprints of a localized core-hole can be observed, if the Auger electron resides in a superposition of *gerade* and *ungerade* states,

and inversely, the core-hole is delocalized if the corresponding Auger electron can be attributed to a distinct *gerade* or *ungerade* configuration. Figure 43 depicts the photoelectron angular emission distribution in the molecular frame. Panel A shows a symmetric distribution averaging over all emitted Auger electrons. The distribution in Panel B becomes asymmetric (depicting localization) as a gate on distinct Auger electron emission directions is applied (thus selecting a *gerade*/*ungerade*-superposition).

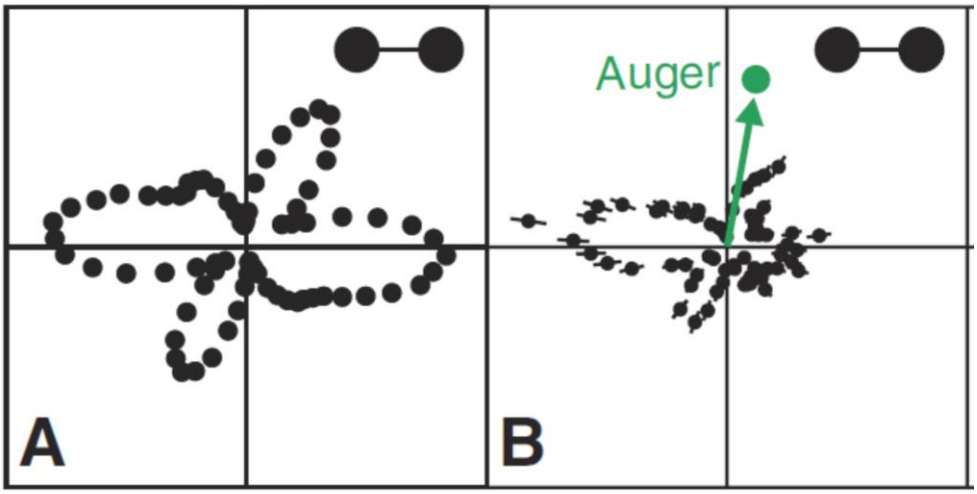

Figure 43: Angular distribution for of 9 eV photo-electrons emitted from the K shell N_2. The circular polarized photons had an energy of 419 eV. The propagation of the photons is perpendicular to the plotted distributions. The molecule orientation is indicated by the bar-bell. A: Integrated over all Auger-electrons, B: Photon electron distribution coincident with a specific Auger electron emission direction (green arrow) [135].

5.6. Efimov state of the He trimer

Since more than hundred years, long-range van der Waal forces have attracted great interest in molecular physics. In their origin they differ from the Coulombic and the covalent bonding force and are created by dynamical correlation (or better dynamical entanglement), Van der Waal forces can create bonding at huge inter-nuclear distance. V. N. Efimov [137] predicted in the late 60-ties of the last century a universal three-body state which exists as any dominating two-body force vanishes. Such bound three-body states have been termed since then "Efimov-states". It has been predicted that at very low temperature an excited He trimer molecule may form an Efimov state with several hundred Angström inter-nuclear distance between its atomic constituents. Already the He dimer is one of the largest, naturally occurring system (exceeding by far 100 Å) with a binding energy of only a few hundred neV. It was discovered in 1994 by Wieland Schöllkopf and Jan Peter Toennies [138] by matter-wave-diffraction and analysis of the observed interference structures.

Starting from initial work on He dimers [139], Maxim Kunitski et al. [140] succeeded in 2015 to produce, identify He trimers in an Efimov state and measure the vibrational wave-function of the ^{4}He Efimov-trimer. They prepared the excited state by employing the matter-wave diffraction technique of W. Schöllkopf and J. P. Toennies [138] and multiply-ionized the trimers with a short, highly intense Laser pulse. The rapid ionization yielded a Coulomb explosion of the trimer and - using the C-REMI approach - the momenta of the ionic fragments were measured in coincidence. From the measured momenta the spatial structure was determined, which is shown in Figure 44. The agreement between experiment and theory is very good. The Efimov trimer consists in principle of a He dimer with the third He atom orbiting at even further distance. This experiment has proven that C-REMI is able to clearly identify even very rare events in the presence of other hugely dominating

processes or background, due to the coincident detection of all fragments with the precise measurement of momenta.

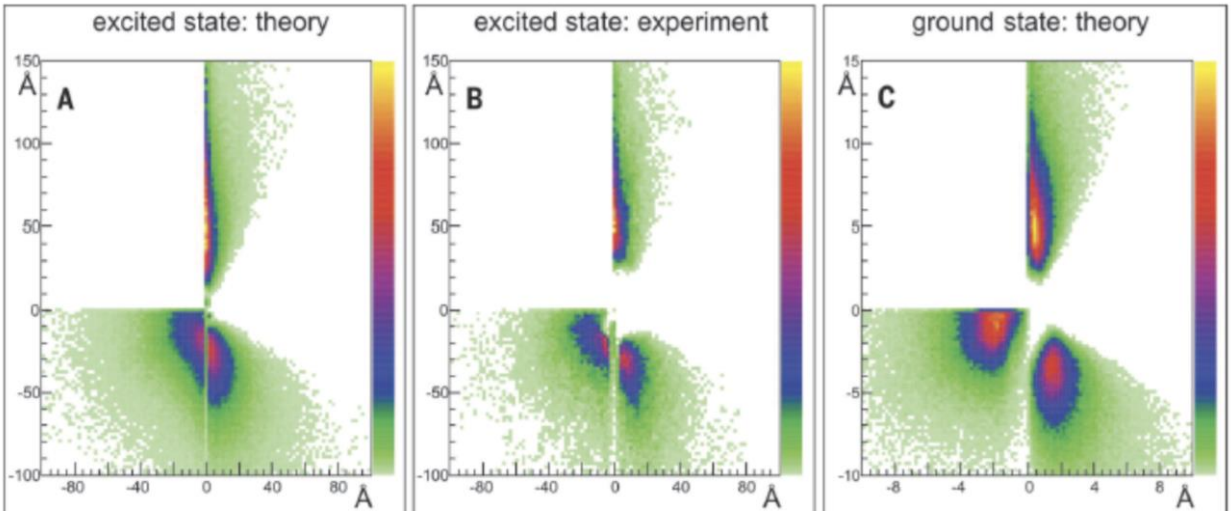

Figure 44: Structure of the He trimer. A: The structure predicted by theory and B: the measured one for the excited Efimov state [140+141]. C: for comparison, the ground state structure as predicted by theory. Notice the factor 10 difference in the size.

5.7. Imaging of structural chirality

Many pharmaceutical drugs have a chiral structure. Since the "Contergan" case [142] in 1961 it became clear that the purity of drugs is a crucial condition for their application. One handedness is constitutional and the opposite handedness can be noxious. Even a very small impurity of the wrong handedness can be very dangerous. Thus it would be of great help if one can recognize for each molecule whether it has the proper chirality. A C-REMI can analyze molecules in the gas phase (and in the future eventually drugs) and decide practically with 100% certainty which handedness is present. Martin Pitzer from the Frankfurt group together with the chemistry group of Robert Berger in Marburg investigated the single-photon (710 eV) and strong-field induced complete fragmentation process of chiral molecules as, for example, CHBrClF and detected the five ionic fragments in coincidence [143+144]. The molecules are randomly oriented in the gas phase, but as pointed out before, the coincident detection of ionic fragments allows for a determination of their orientation on a single molecule basis. Moreover, when investigating larger

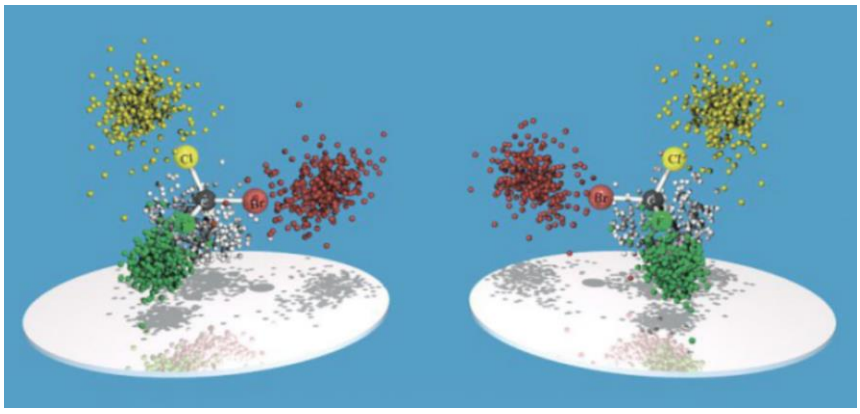

Figure 45: Momentum vector distribution of ionic fragments of the chiral CHBrClF molecule after Laser ionization. Left: Left handedness, right: right handedness [143] (see text above).

molecules, even the molecular structure can be reconstructed from the momentum measurement. As an example, the distribution of the measured momentum vectors is shown (after multiple ionization of CHBrClF using a fs-Laser) in figure 45. The Carbon ion is marked by the black sphere, the H ion by the white dots, the F ion by the green dots, the Cl ion by the yellow dots and the Bromine ions by the red dots. The multiple coincidence condition of 4 or five fragments reduces the background nearly to zero and allows to distinguish molecules of different handedness from a racemat, i.e., the experimenter can extract for each ionization event the handedness of the molecule. In figure 46 this unambiguous identification of the handedness by using C-REMI becomes obvious. Here the data are plotted as function of the chirality parameter

$$\cos \Theta_{F(ClxBr)} = \boldsymbol{p_F} \bullet (\boldsymbol{p_{CL}} \times \boldsymbol{p_{Br}}) / (\left| \boldsymbol{p_F} \right| \bullet \left| \boldsymbol{p_{CL}} \times \boldsymbol{p_{Br}} \right|) \text{ [137].}$$

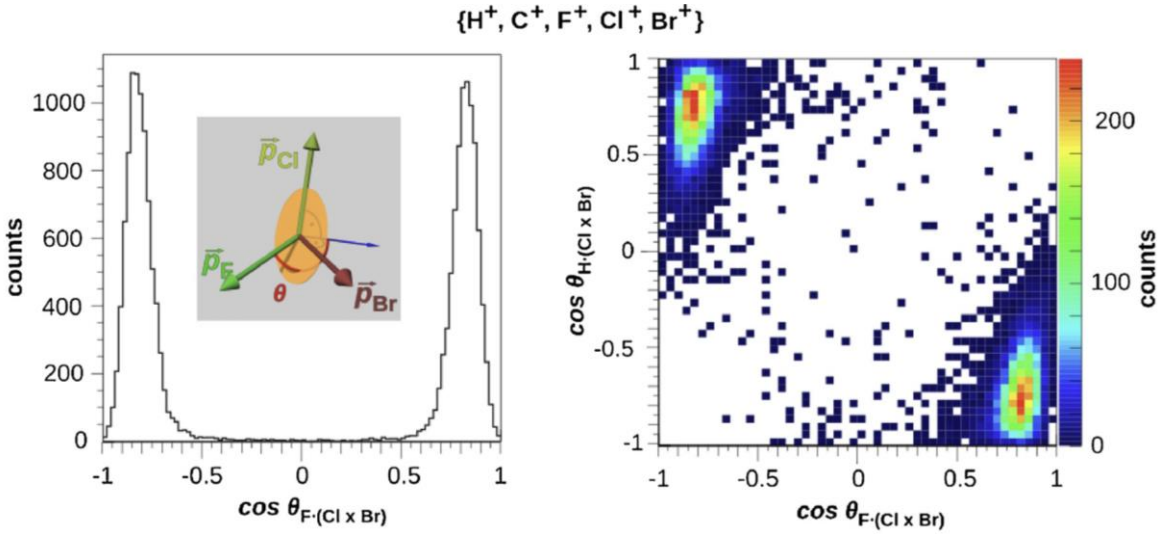

Figure 46: Measured handedness distribution as function of the chirality parameter $\cos \Theta_{F(ClxBr)} = \boldsymbol{p_F} \bullet (\boldsymbol{p_{CL}} \times \boldsymbol{p_{Br}}) / (\left| \boldsymbol{p_F} \right| \bullet \left| \boldsymbol{p_{CL}} \times \boldsymbol{p_{Br}} \right|)$ [143].

5.8. Spatial imaging of the H_2 vibrational wave function

Dependent on the gas temperature, molecules in the gas phase undergo repetitive collisions with neighboring molecules. This leads to excitation of vibrational or rotational states. The exploration of this intra-molecular motion with traditional x-ray or electron diffraction methods is complicated, as the method is not very sensitive to such features and yields the mean averaged spatial structure. Using Coulomb explosion imaging methods [144] with subsequent coincident measurement of all momenta of the ejected fragments, however, can yield information on this intra-molecular motion (i.e., the vibrational wave function of the nuclei) with high resolution. In Frankfurt Lothar Schmidt et al. [145] have investigated the vibrational states of excited H_2 molecules. 2.5 keV H_2^+ ions produced in a Penning ion source collided with a very cold super-sonic jet He beam and were neutralized by capturing one electron into the different vibrational states. Using the C-REMI approach the two neutralized H fragments were detected in forward direction by a multi-hit capable time- and position-sensitive detector and the He^+ ion was detected perpendicularly to the ion beam with a C-REMI system. Since the momenta of all three fragments were measured with high resolution (< 0.04 a.u. => 3 micro eV) the kinetic energy release KER and the electronic excitation energy (different vibrational states) could be cleanly determined. From the measured H momenta, the H_2^+ inter-nuclear distance was inferred using the reflection approximation (see figure 47). The reflection methods yield slightly different results in case of approximating the nuclei as "frozen" or “moving”. This difference becomes obvious from figure 49, when the data are analyzed for both reflection methods

(green dots: frozen nuclei; red circles: moving nuclei). The solid line represents a mean value of both reflection methods.

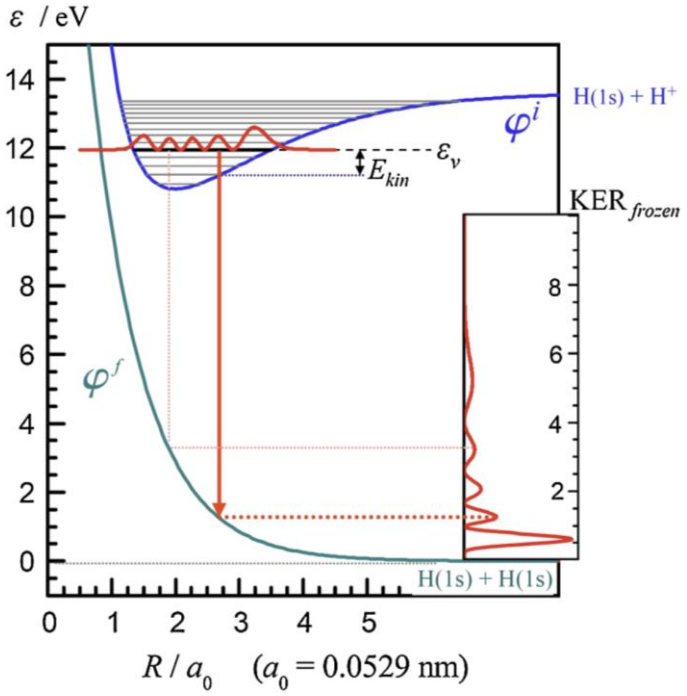

Figure 47: Calculated energy levels as a function of the inter-nuclear distance R for H_2 molecules depicting the concept of the "reflection approximation".

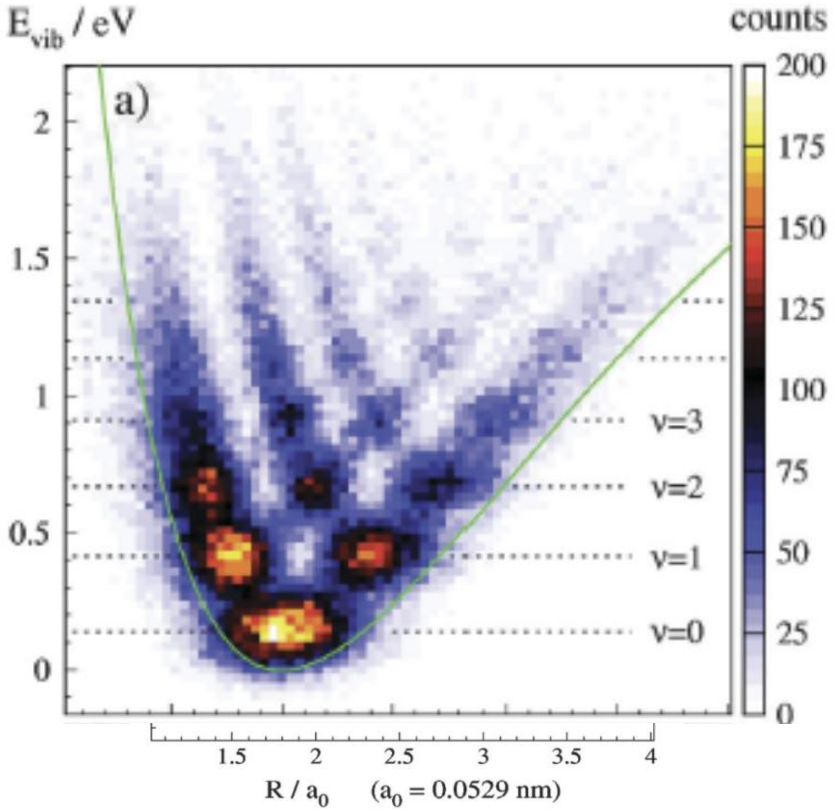

Figure 48: Experimental density plot of vibrational states as function of the inter-nuclear distance and electronic excitation energy E_{vib}. The green line is the potential energy curve ($H_2^+(1s\sigma_g)$) calculated in the Born-Oppenheimer approximation [145].

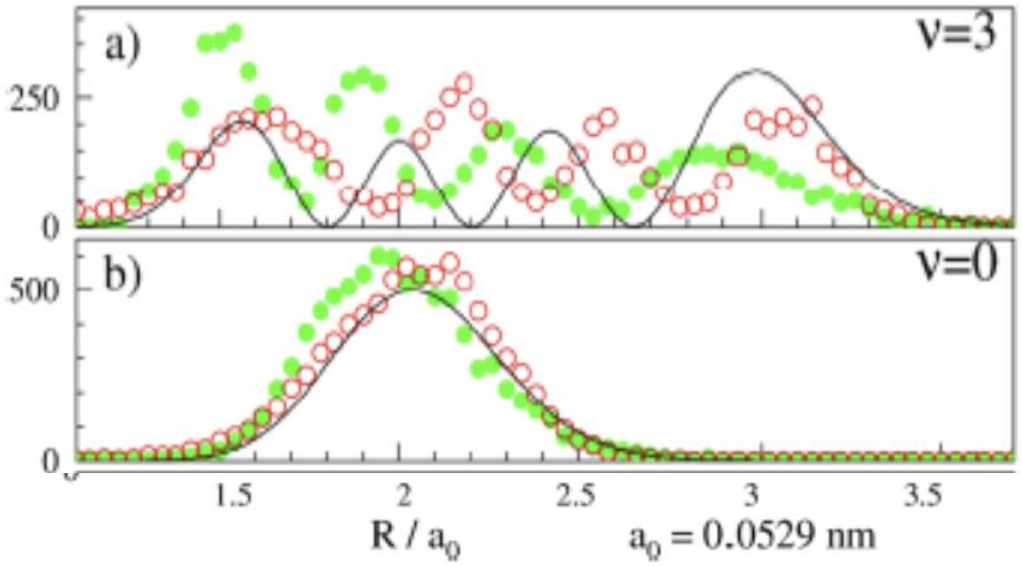

Figure 49: Distribution of vibrational states as function of the inter-nuclear distance R, where R is calculated for the frozen and moving nuclei reflection methods (green dots: frozen nuclei; red circles: moving nuclei). The solid line represents a mean value of both reflection methods [145].

5.9. Visualization of directional quantization of quasi-molecular orbitals in slow ion-atom collisions

"Space quantization" or more appropriate "Directional quantization" (Richtungs-quantelung) of atomic states in the presence of an outer magnetic field is known since 1916 when it was proposed by Sommerfeld and Debye [146] and its verification in 1922 in the

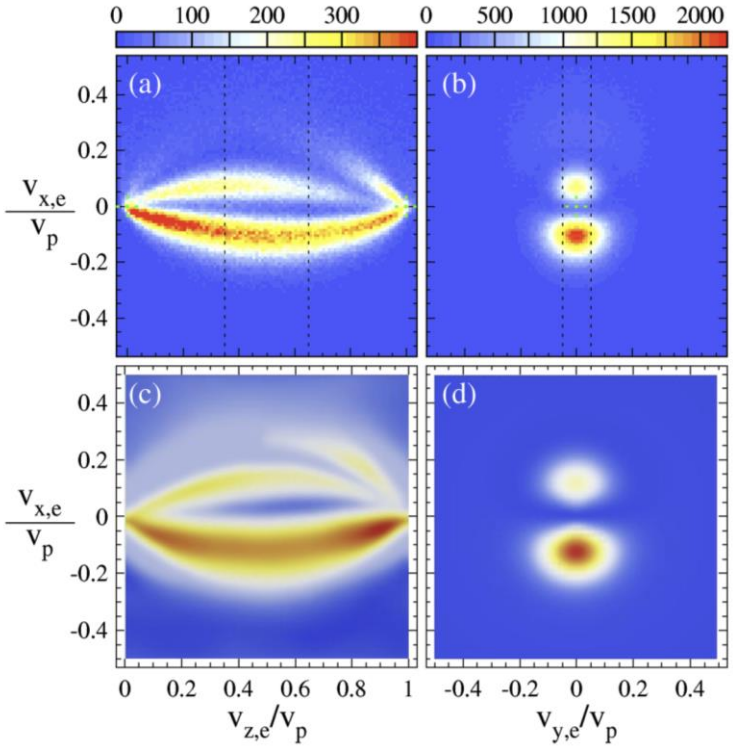

Figure 50: Electron momentum plots in 10 keV He^{2+} + He → He^{+} + He^{2+} + e transfer ionization processes [147]. The abscissa and the ordinate are in units of the projectile velocity v_p, i.e. the electron momenta, panels a) and b) are experimental data and c) and d) theoretical predictions. Panels a) and d) depict the projections on the nuclear scattering plane an b) and d) perpendicular to it.

Stern-Gerlach experiment [14]. The existence of such a directional quantization also in electric field was already indirectly seen in the Stark-effect. The existence of a directional quantization of electronic quasi-molecular states was recently nicely explored by Lothar Schmidt in Frankfurt [147] where he measured the electron emission in slow 10 keV He^{2+} + He → He^{+} + He^{2+} + e transfer ionization processes. By measuring all three emitted charged fragments in coincidence, the electron emission pattern with respect to the nuclear scattering plane were visualized. In this slow collision process inner-shell quasi-molecular orbitals are formed which are oriented in angular momentum (directional quantization) with respect to the nuclear collision system.

Averaging over all orientations of the nuclear scattering plane, the electron emission pattern does not show any sign of directional quantization, only when for each event the orientation of the nuclear plane is measured. In Figure 50 the distribution of the emitted electron projected on the nuclear collision plane is shown (a experiment, c theory). The discrete structure corresponds to discrete angular momentum states. The abscissa and ordinate are given in units of the ion velocity v_p. A detailed discussion of this structure is given in [147]. In Figure 50 b and d the projections perpendicular to the nuclear scattering plane are presented. The comparison between experiment and theory shows perfect agreement and proves that in any quantum measurement where the experimenter is sensitive to angular momentum the quantum system reveals the principle existence of directional quantization, i.e. the ordering concept of dynamics in quantum systems.

5.10. Time-resolving studies employing coincidence detection techniques

In the recent past it has been demonstrated, that time-resolving experiments are possible without having a projectile source with corresponding timing properties, as, for example, in a laser pump-probe scheme. In some cases, the temporal evolution on atomic or molecular time scales can be deduced from other information obtained from the coincident detection of ions and electrons. This subsection will provide three recent examples of such studies.

Interatomic Coulombic Decay (ICD) has been a subject of large interest, as described in section V.d. Its efficiency (and thus the lifetime of IC-decaying states) is strongly linked to the inter-nuclear distance between the participating entities. As typical ICD lifetimes are in the range of a few tens of femtoseconds to picoseconds, the excited compound, that will undergo ICD, will exhibit changes of its geometry prior to the decay. These nuclear dynamics triggered strong interest in performing time-resolved measurements of ICD during the last decade, because – as mentioned above – the nuclear motion alters dynamically the electronic decay probability, making ICD a prototype process for distinct non-exponential decay behavior. A molecular movie of the nuclear motion during ICD in helium dimers has been obtained in 2013 by Trinter and coworkers [148]. They used a synchrotron source for triggering ICD in He_2, which has obviously no timing properties, that allow for a direction determination of single event ICD lifetimes (typical synchrotron light pulses have a duration of approx. 100 ps). Accordingly, Trinter et al. introduced a novel approach to extract the decay time of single ICD events from their coincidence measurement. By the so-called “PCI-streaking” the decay time is encoded in the photoelectron kinetic energy. PCI (Post Collision Interaction) is an effect studied in detail already since the 1970-ties [149]. Adopted to the scheme of ICD, the following process takes place: a low energy photoelectron is emitted from a dimer creating the IC-decaying state. As ICD occurs, an ICD electron (in the case of He_2 of approx. 10 eV kinetic energy) is released. If the photoelectron has been chosen sufficiently slow (by selecting an appropriate photon energy from the synchrotron light source) the ICD electron will

overtake the photoelectron which causes a change of the effective potential the photoelectron is emerging from, i.e., the potential changes from effectively singly charged to doubly charged. The more attractive potential will decelerate the photoelectron, and the amount of deceleration depends on the emission time of the ICD electron. Thus, by performing a high resolution measurement of the photoelectron momenta and the two ions created in process, the decay time can be inferred from the photoelectron energy and the inter-nuclear distance of the two atoms of the dimer from the ions' kinetic energy release. Employing this approach, Trinter et al. were able to create snap shots of the nuclear motion during ICD covering the first picosecond after the excitation.

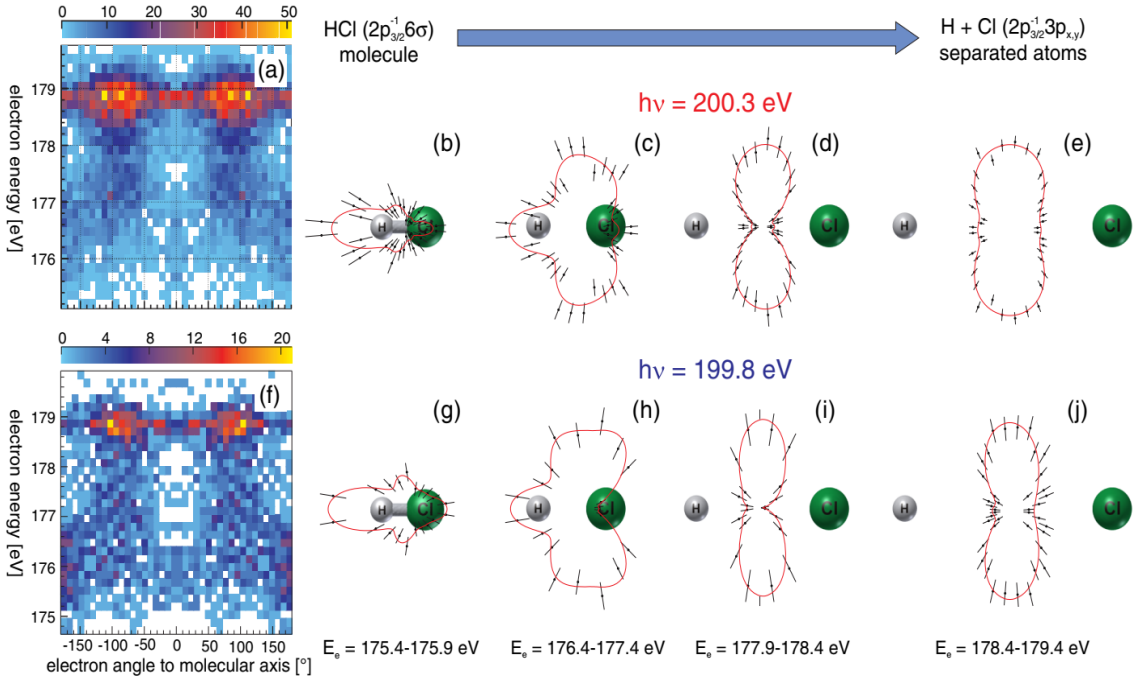

Figure 51: b)-e) and g)-j) show the transformation of the molecular frame angular distributions of Auger electrons emitted during ultrafast dissociation of HCl. The MFPAD shows initially molecular features (left) and becomes atomic for larger inter-nuclear separations (right). The figure has been taken from [150].

Similarly, but using a different approach in detail, Sann et al. showed, how an electronic orbital transforms from being *molecular* to *atomic* upon dissociation of a molecule [150]. A resonant excitation of a HCl molecule triggered its (ultrafast) dissociation [151]. During the dissociation an Auger electron is emitted. Depending on the emission time, the Auger electron is either emitted from the still intact HCl molecule, an intermediate state or – at later times – from the Cl atom. Sann and coworkers investigated the molecular frame angular distributions of the Auger electron for different inter-nuclear distances during the dissociation. The inter-nuclear distance has been inferred from the energy of the ion measured in coincidence with the Auger electron, providing (as the overall dissociation and decay process typically occurs within 5 fs) information on the timing, as well. The molecular frame angular distributions changed during the dissociation from showing signatures of a molecular orbital to an atomic distribution as shown in figure 51.

Very recently, Grundmann et al. investigated the following question employing a multi-particle coincidence approach [13]: Is an electron emitted simultaneously from all across a molecular orbital as it is released by photoionization, or is it first released from that portion of its orbital that is "illuminated first" by the photon? A H_2 molecule has been used as prototype test bench to answer this question. Photoemission from a homo-nuclear molecule can be - due to the two-center nature of the molecule - intuitively regarded as a microscopic

analog to scattering at a classical double slit. The photoelectron wave is emitted as a superposition from the “left” and the “right” atom of the molecule, which, indeed, causes Young-type interference patterns in the molecular frame angular emission distribution of the electron. Grundmann and coworkers showed, that the molecular frame angular emission distribution changes subtly if the molecule is oriented along the photon propagation direction or perpendicular to it during the photoionization process. Within the double slit picture these changes are understandable: if the molecule is oriented perpendicular to the photon direction, the photon arrives at both nuclei of the molecule at the same time. However, if it oriented in parallel to the propagation direction, one of the atom is hit prior to the second one. This delay in the arrival time can be modelled as a phase shift of the one of the emerging photoelectron waves, or, in the double slit picture, a phase shift in one of the two slits, which causes a measureable displacement of the double slit interference pattern. From this displacement a birth time delay of approx. 250 zeptoseconds was resolved in the experiment, which nicely corresponds to the travel time of the photon along the molecule. The sensitivity of this approach is below a few 10 zeptoseconds, despite employing synchrotron light pulses of >100 ps duration.

5.11. Proposed experiments in Neutrino Physics:

In an article published in 1994 in Comments on Atomic and Molecular Physics Ullrich et al. [152] presented future perspectives of the C-REMI technique. One exotic one is worth mentioning here: With the C-REMI approach, i.e. measuring with ultra-high resolution the momenta, one can determine in principle from one single event also the mass of a particle. Therefor it was thoroughly discussed whether one could use a C-REMI to measure, in the decay process of Tritium, the neutrino momentum by performing a He^{1+} recoil ion-electron coincidence. To be sensitive to a very small neutrino mass of about one eV in this decay event the electron kinetic energy must be very close to the Q-value of the tritium decay. These events are extremely rare. Thus because of the huge number of random coincidences the required time of measurement would nearly approach the life-time of an experimenter. May be somebody will discover a way to handle such a high random rate?

6. Conclusion

The C-REMI technique can be considered as the "Bubble Chamber" or "Time Projection Chamber" in atomic and molecular physics. Using the multi-coincidence concept initially developed in nuclear and high energy particle physics, a C-REMI can image the whole momentum space in a single-event quantum process. Using ultra-cold targets in the gas phase and electro-magnetic spectrometer designs with focusing conditions an excellent sub-atomic momentum resolution and a large multi-hit coincidence efficiency are obtained. Thus visualizing the complete dynamics in a single event the dynamical entanglement in many particle systems can be explored. The C-REMI is now a standard detection system in many fields of physics and chemistry and is used by many groups around the world.

7. Acknowledgement

Many of the scientists contributing to the C-REMI development are co-authors of this paper. But we are indebted to many other colleagues: Reinhold Schuch, Amin Cassimi, Nora Berrah, Andre Staudte, Paul Corkum, Kiyoshi Ueda, Tadashi Kambara, Yasu Yamazaki, Paul Mokler, Siegbert Hagmann, Klaus Blaum, Erhard Salzborn, Alfred Müller, Karl Ontjes Groeneveld, Hans Joachim Specht, Bernd Sonntag, Jochen Schneider, Berthold Krässig, Timor Osipov, etc. for a long close cooperation and many theorists providing ideas for measurements like John Briggs, Burkhard Fricke, Hans Jürgen Lüdde, Jan Michael Rost, Steve Manson, etc.. Additionally we want to thank BMBF (Dietrich

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The early team laying the foundations of C-REMI. Painting by Jürgen Jaumann 2011

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