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Photo induced multiple fragmentation of atoms and molecules: Dynamics of Coulombic many-particle systems studied with the COLTRIMS reaction microscope

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Abstract

Many-particle dynamics in atomic and molecular physics has been investigated by using the COLTRIMS reaction microscope. The COLTRIMS technique visualizes photon and ion induced many-particle fragmentation processes in the eV and milli-eV regime. It reveals the complete momentum pattern in atomic and molecular many-particle reactions comparable to the bubble chamber in nuclear physics.

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1. Introduction

The photo effect, i.e. the absorption of a photon by an atom or molecule followed by the emission of an electron into the continuum, is a purely kinematical effect. Considering only the energy balance (famous Einstein equation) of photon energy $h\nu = E_B - E_{\text{kin}}$, where E_B is the electron binding energy in the initial-state and E_{kin} the final-state kinetic energy of the electron, fundamental questions concerning the photo effect remain unanswered. An electron at rest cannot absorb a photon. The photon can be only absorbed, if the electron momenta of the initial-state and the final-state match. The electron is projected into the continuum state and does not work against the Coulomb barrier (like in classical physics). Furthermore the electric field of the photon does not accelerate the electron to

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its velocity in the final state. In order to investigate such effects one has to explore in full detail the dynamics of the Coulombic many-particle systems. In particular this applies to multiple ionization processes which are known to depend strongly on the e–e correlation of the initial state.

Correlated many-particle dynamics in Coulombic systems, which is one of the unsolved fundamental problems in physics, can now be experimentally investigated with unprecedented completeness and precision. The recent development of the COLTRIMS reaction microscope (COLd Target Recoil Ion Momentum Spectroscopy) [1,2] provides a coincident multi-fragment imaging technique for eV and sub-eV particle detection. In its completeness it is as powerful as the bubble chamber in high energy physics. Varying the momentum transfer by using different projectiles like photons as well as slow or fast ions the many-particle system can be selectively fragmented. Thus the Coulombic many-particle dynamics can be explored with high resolution.

To reveal either the initial-state correlation of the target system or the final-state interaction of the fragmentation process the energy and momentum transfer from the impacting projectile on the target must be properly chosen. In order to explore the initial-state correlation the momentum transfer between the fragmenting partners should be small compared to the initial-state momenta of the target fragments. In photon absorption the momentum transfer is indeed rather small, however, the photon energy must be high enough so that the electrons are emitted with a high velocity in order to minimize the interaction time between the fragments and thus to reduce the final-state interaction between the fragments.

The interaction of a fast ion with the target at large impact parameters is comparable to the photo effect. The ion interacts with the target mostly via virtual photon transfer. For fast projectiles the momentum transfer from ion to target via virtual photons is rather small too and resembles the one of a real photon. Therefore both cases—photon absorption and fast ion impact—can be used (as long as the energy transfer is properly chosen) to obtain insight into the initial-state correlation.

In case of small energy transfer the emitted electrons are of low energy and post collision interaction becomes relevant. In such cases the final-state interaction dominates the momentum pattern of the final state. Thus for low energetic photon impact or low virtual photon energies one can probe the final-state interaction and the fragmentation dynamics. Particularly if the target is a molecule the molecular structure can be well investigated by analyzing the scattering patterns of the emitted low energy electrons. Due to the high energy and momentum resolution one can determine the orbital, from which the electron was emitted.

We will discuss in this Letter the principle of the COLTRIMS imaging technique and present two selected examples of investigated fragmentation processes: Single photon induced complete 4-body fragmentation of D_2 molecules, i.e. $\gamma + D_2 \rightarrow 2D^+ + 2e^-$ [3,4] and single photon induced 4-body fragmentation of van der Waal bound molecules [5].

In the outlook the technical perspectives of the COLTRIMS technique towards many-particle detection will be discussed.

2. The COLTRIMS imaging technique

In Fig. 1 the principle of the COLTRIMS reaction microscope is presented. In a well designed electric field configuration (static or pulsed) the positively as well as the negatively charged fragments are projected (typically with 4π solid angle) towards two position-sensitive detectors. Measuring the impact position on the detector (typically < 0.25 mm resolution) and the time-of-flight of the fragment (TOF) between the fragmentation and the impact on the detector, the particle trajectory—and thus the particle momentum obtained in the fragmentation—can be determined. To improve its momentum resolution electrostatic lenses can be incorporated into the projection system, such that the influence of the size of the target region, from where the fragments originate, can completely be eliminated [1,2].

To detect also high energetic electrons one can apply magnetic fields, superimposed on the electric field [1], as well as pulsed electric fields. If particle detectors based on fast delay-line position read-out are used multi-hit detection is possible. Even two particles impacting on the detector at the “same” instant ($\Delta t < 1$ ns) can simultaneously be detected—as long as they are separated in space by at least 10 mm. The data are acquired and stored in list mode. The number

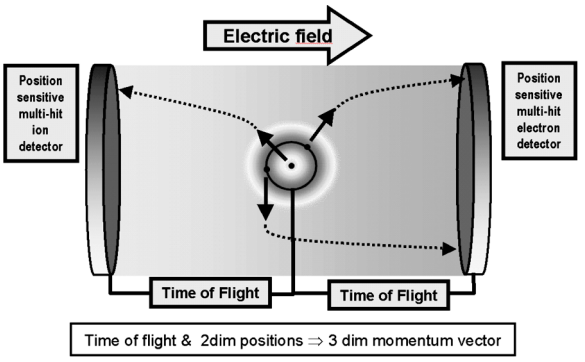


Fig. 1. Scheme of the COLTRIMS imaging system [2].

of detected multi-hits is only limited by the electronics needed to store the information. Thus, for low energy particles (milli-eV to hundreds of eV) the COLTRIMS method is indeed as powerful as advanced bubble chamber systems for high energetic (MeV) particles. It is even comparable with modern Time-Projection Chamber systems used in high energy physics. Furthermore the rate of fragmentation processes per second can exceed 100 kHz.

To obtain 4π solid detection angle one has to ensure that the transverse velocities of the fragments multiplied with the time-of-flight from target location to detector yields a transverse position which falls within the detector area. To yield good momentum resolution a long time-of-flight is required. Thus for high energetic electrons (above 100 eV) a magnetic field (parallel to the spectrometer axis) must be applied in order to bend the electrons onto spiral trajectories leading towards the detector. Since x and y components are decoupled from the z component (time-of-flight direction) the initial momentum of the electron can still be deduced from the electron time-of-flight and position on the detector. Details of this technique are given in [1].

To yield good momentum resolution for the ionic fragments the target object has to be at rest in the laboratory system. Thus target cooling by super-sonic jet expansion or even laser cooling in a trap is crucial. Since cooling to sub-Kelvin temperatures of heavy atoms and molecules is difficult, a cold helium jet can be used as carrier medium. If all fragments of a molecular breakup are detected it is possible to calculate the initial momentum of the molecule before the fragmentation. This allows for the elimination of the momentum uncertainty caused by the internal temperature of the target.

Other important components of the imaging device are the position-sensitive detectors. In some cases several fragments can impact on the detector within a few nanoseconds. Therefore the detectors must produce short signals widths below 10 ns. Thus fast delay-line techniques for position recording instead of charge-dividing techniques are required to provide this kind of multi-hit capability. Using standard channel plates MCP typical pulse width's are in the 10 ns regime. On the position-sensitive delay-line anode these MCP pulses create signals of less than 10 ns half width (typically about 5 ns FWHM). The signal propagates with a velocity of about 20 cm/ns to both ends of the anode plane. The arriving time can be measured with a precision of about 100 ps on both ends of the delay line. Since the delay-line anode can have two or even three crossed wire planes for each particle 5 or even 7 time signals are recorded. This redundancy allows for a precise position measurement. Due to this multiple time recording the multi-hit dead time for a three-layer anode (hexagonal delay-line structure) is in the order of less than one nanosecond.

This multi-parameter recording is only manageable if list-mode data recording is used. List-mode recording stores all information of the fragmentation process (e.g. each parameter of an event is recorded as a number in a list event by event), thus at any time later the entire measurement can virtually be replayed again and again in the computer analysis.

3. Data and discussion

The experimental as well as the theoretical investigation of the complete fragmentation of the H_2 or D_2 induced by a single photon has been one of the great unsolved challenges in molecular physics [6–15]. Only very recently Weber et al. [3,4] measured the complete momentum distribution of the 4-body fragmentation process $\gamma + D_2 \rightarrow 2D^+ + 2e^-$ and Vanroose et al. [16,17] could successfully calculate the corresponding differential cross sections. This investigation is a breakthrough in molecular physics. The two-center potential of the D_2 molecule influences the final-state momentum pattern of the two outgoing electrons. Even though the molecular axis is randomly

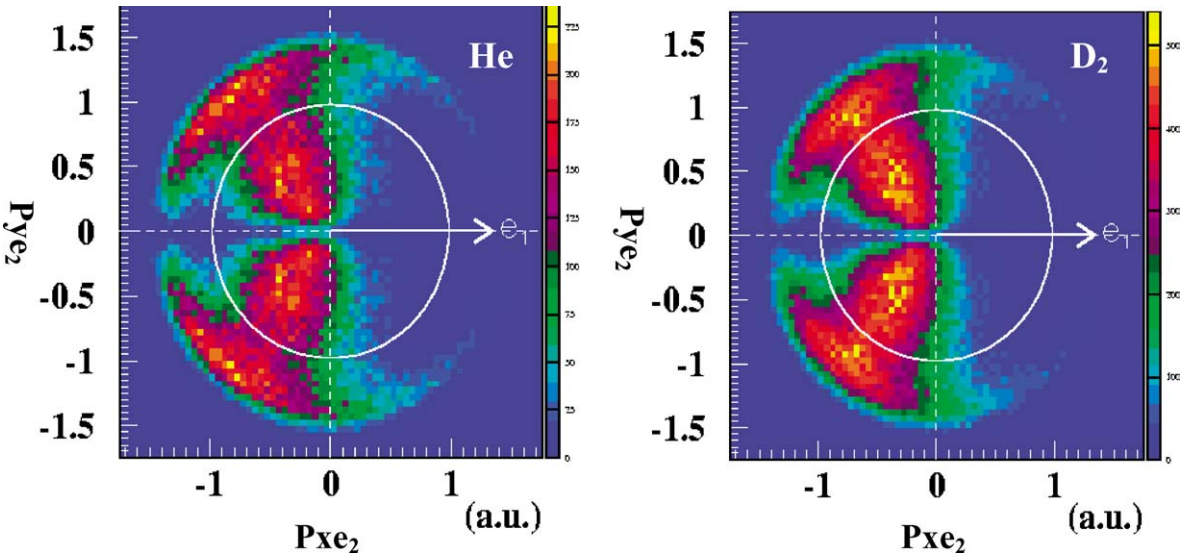


Fig. 2. Correlated momentum distributions in the plane which is defined by the momentum vectors of the two electrons for photon induced double ionization of He and D_2. Electron 1 is emitted in the direction of the arrow (integrated over all momenta Pe_1). The polarization vector of the linearly polarized photon is oriented horizontally. The circle marks the region of equal energy sharing. For D_2 the molecular axis is randomly oriented.

aligned before the collision it is possible to deduce the alignment of the axis from the measured final-state momentum vectors of the D^+ ions. These ionic momentum vectors are always parallel to the orientation of the molecular axis at the instant of the ionization. The correlated momentum distributions of the emitted electrons for He and D_2 are shown in Fig. 2 (the labeling of the two electrons is arbitrary).

In case of randomly aligned molecules in other words if we integrate over all molecular orientations we observe strong similarities to the double ionization of He. Due to Coulomb repulsion electron 2 is emitted into the left half plane opposite of electron 1. Angular momentum selection rules result in a node at 180° [8,10–15].

In Fig. 3 the same differential cross sections are displayed. But now we have selected only those events in which the molecular axis was aligned. The chosen alignment of the molecule is indicated in Fig. 3. In this case electron 2 is emitted preferably along one side of the molecular axis. It is evident from these figures, that now the emission of electron 2 depends strongly on the alignment of the molecular axis with respect to the photon polarization and to the emission angle of electron 1 (see Fig. 4).

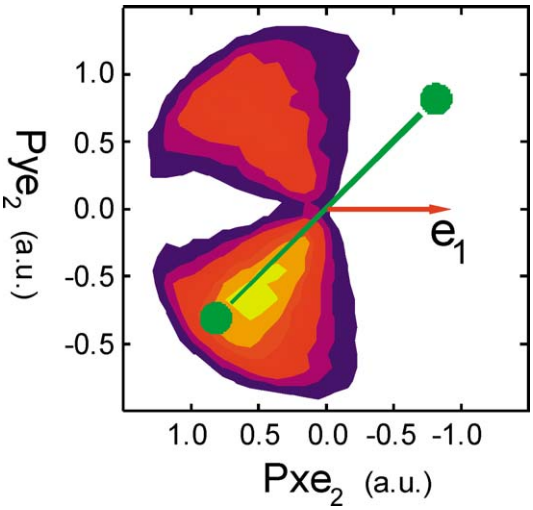


Fig. 3. The same momentum distribution (contour plot) as in the right part of Fig. 2, but now for one given molecular alignment (green dumb bell).

The measured data set is a nine-fold differential cross sections of a four-fold fragmentation process. In the analysis procedure the multi-dimensional data are typically projected down to three or less dimensions. Often the projection plane is given by the photon po-

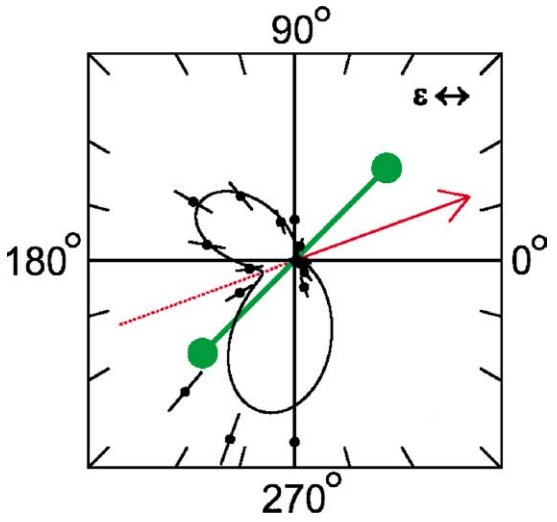


Fig. 4. The angular distribution of electron 2 (similar as in Fig. 3) in case of equal energy sharing (We04). The polarization vector is horizontally aligned. Electron 1 is emitted in the direction of the arrow. The selected alignment of the molecule is indicated by the green solid circles. The solid line shows calculations by Feagin [8,10].

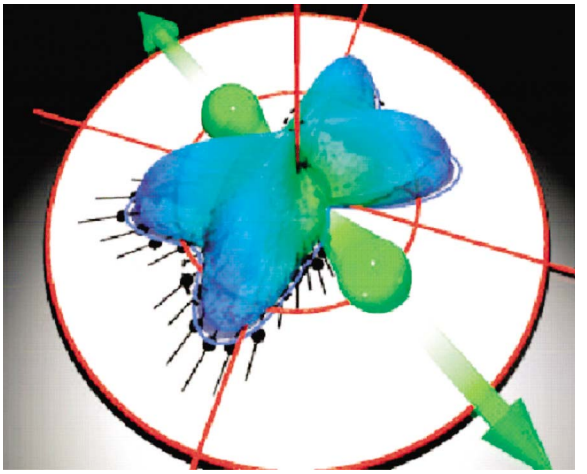
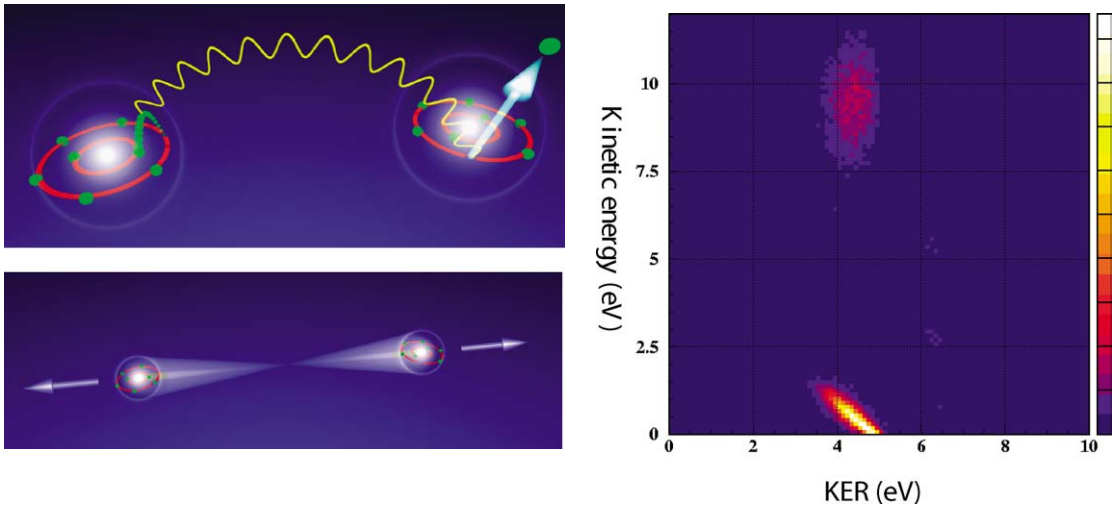


Fig. 5. Three-dimensional momentum distribution for the second electron with the condition that electron 1 was emitted perpendicular to the plane of molecular and polarization axis. The alignment of the molecule is indicated by the green lobes and arrows. Since the kinetic energies of the D^+-ions are known (KER) it is possible to calculate the internuclear distance at the moment of the ionization. For this picture we have chosen reactions with very small internuclear separations. It turned out that the shape of this distribution strongly depends on the internuclear distance.

larization or the molecular alignment. However, which cuts and projections are best suited to separate the correlations in the initial-state from the correlations in the final-state cannot be answered by the experimentalists alone. For this difficult task the support from theory is essential. Recently Vanroose et al. [16,17] have calculated the many-fold differential cross sections. They show that by viewing the cross sections in the proper chosen plane mostly the initial-state or (vice versa) final-state correlations determine the resulting momentum patterns. Thus only with the theoretical assistance the fundamental aspects of the four-body correlation can be studied. According to Vanroose et al. [16,17] the dynamical correlations in the initial-state have a strong impact on the final-state momentum pattern in particular if electron 1 is emitted perpendicular to the plane of the molecular axis and the photon polarization. In Fig. 5 the three-dimensional momentum distribution is shown for the second electron under the condition that electron 1 is emitted perpendicular to the plane which is defined be the molecular and the polarization axis (small internuclear distance). Since the kinetic energies of the D^+-ions are measured too it is possible to deduce the internuclear distance at the moment of the ionization. The joint experimental and theoretical investigations show that the shape of this distribution strongly depends on the internuclear distance.

In addition to the above described experiment a second measurement is presented in the following section: To demonstrate the multi-coincident imaging capabilities of COLTRIMS the photon induced 4 particle fragmentation of Ne dimers into two Ne^+ and two electrons is presented here [5]. At a photon energy of 10 eV above the ionization threshold this fragmentation channel can only occur if the excitation energy in one atom is transferred through the intermolecular barrier to the other atom and results in an autoionization of the neighboring atom. This process is called Inter-Atomic Coulombic decay. Cederbaum et al. [18] have predicted that this process is the dominant decay channel and that it proceeds via a fast energy transport between the two atoms via virtual photon exchange.

To investigate this decay channel one 2s electron of a Ne dimer is ionized by absorption of a 59 eV photon (Fig. 6(a)). This 2s hole cannot decay directly via Auger decay in that ion because the resulting energy is not sufficient for the detachment of another electron from the Ne^+-ion. Thus the 2s hole in the Ne^+ ion can only decay by photon emission or by a newly



(a) (b)

Fig. 6. (a) Scheme of the ICD process: First a 2s-hole is created in the left Ne-atom. The hole decays and the excess energy is transferred to the neighboring atom via exchange of a virtual photon. This energy is sufficient to ionize the other neutral Ne atom. This leads to the dissociation of the dimer. (b) Electron energy vs. KER (sum energy of both Ne^+-ions). In this representation the ICD-electrons (lower section) can be clearly separated from the photo electrons of the primary 2s ionization (upper section).

discovered decay channel: The excitation energy is transferred to the neutral molecular partner in 6.7 a.u. internuclear distance by a virtual photon. The energy is sufficient to ionize the neutral neighbor atom. This new deexcitation process is called ICD—interatomic Coulombic decay [18]. The energy transfer can occur via Auger-like electron–electron interaction through the barrier (tunneling of an electron through the barrier) or via van der Waal-like virtual photon exchange between the molecular partners. These two processes can experimentally only be distinguished if the decay time is measured. The Auger-like process with electron tunneling through the barrier would have a decay time longer than one nanosecond, the virtual photon exchange process according to Cederbaum et al. occurs on the time scale of 100 fs which is even much faster than the radiative decay of a 2s hole in Ne^+-ion. In order to identify this decay channel the momenta of the two Ne^+ fragments, the primary 2s photo electron and the low energetic ICD-electron from the neutral Ne atom have to be measured in coincidence. In Fig. 6(b) the energy distribution of the fragmentation process is presented: on the horizontal axis the sum energy (Kinetic Energy Release—KER) of both ionic fragments against the electron energy (vertical axis) is plotted.

The electrons in Fig. 6(b) at about 10 eV are the primary 2s photo electrons, the ones between 0 and 2 eV are the Auger electrons resulting from ICD and the continuum distribution between both (not visible in Fig. 6(b)) originates from intermolecular electron-electron scattering processes. The diagonal structure of the low energetic electrons in Fig. 6(b) is an unambiguous characteristic of the ICD-electrons. No other processes could produce such a structure. With the help of theoretical models it is possible to determine the life time of the interatomic Coulombic decay. In order to do this one must analyze the intensity distribution along the diagonal structure. The analysis of the data yields a decay time of approximately 100 fs [19].

This measurement of the ICD-process in Ne dimers and the results of the analysis presented here demonstrate the capabilities of the COLTRIMS technique. It is a powerful tool for the investigation of the dynamics of many-particle fragmentation processes.

4. Outlook

The COLTRIMS reaction microscope has indeed opened a window into the world of quantum dynamics

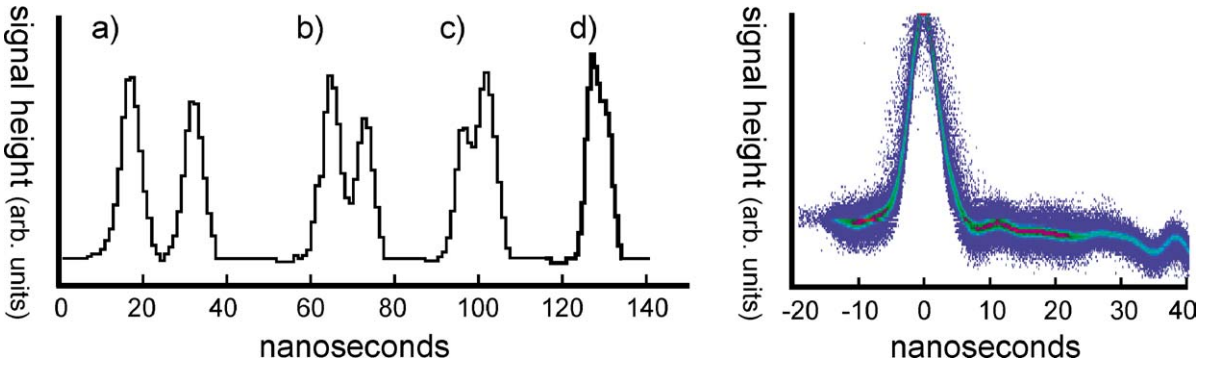


Fig. 7. Left part: Double peaks with different spacing. Even case (d) can be resolved as two peaks using pulse fitting methods. Conventional TDC-systems combined with constant-fraction discriminators do not reach this performance. Right part: Several hundred normalized pulses are plotted in the same histogram. The shape of the signal is apparently independent from the pulse height which is a prerequisite for the fit algorithms.

of many-particle Coulombic systems. The technique is widely applied for single but also multiple photon absorption processes. More and more complex fragmentation systems are being investigated. Therefore the improvement of multi-hit capability with better timing characteristics is one of the experimental challenges to be solved in near future. The multi-channel plate detectors with delay-line anode read out are presently not the limit in multi-hit detection. These devices can handle even local detection rates beyond GHz rates (for a short fraction of time). However, the high-rate electronic read-out and data storing is presently the bottle neck of the system. Fast transient recorders (ADCs), which digitize the incoming signals on a sub-nanosecond scale, can be used to overcome this problem. In Fig. 7 examples of such recorded signals are shown (1 ns/channel). It can be seen, that multi-hit events can even be recognized as two pulses if the two signals are separated by only about three nanoseconds. If two particles are impacting within this dead time the double-hit can still be identified and analyzed, if both particles are separated in detector position by a few mm. The test experiments performed showed that the internal detector and electronic timing can thus be improved to yield a timing resolution of well below 100 ps. Furthermore the integrals and the amplitudes of the signals are measured too. This information helps to match anode signals with each other even if the timing information itself is ambiguous. This yields an important improvement for the analysis of multi-hit events, e.g. when three or more particles hit the detector simultaneously.

In combination with improved readout electronics the COLTRIMS imaging technique is prepared for the next step—multiple fragmentation processes of large molecules. Furthermore the imaging technique is not restricted to pure gas phase experiments. Experiments on fragmentation or electron-emission processes on surfaces are already in progress. The COLTRIMS technique opened a window into the dynamics of the quantum world. In its significance to reveal dynamics on the sub-atomic scale it is comparable to electron or scanning tunneling microscopes which revealed static structures with sub-atomic resolution.

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