Hot recoils from cold atoms
Turkstra, Jan Willem

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
2001

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

Copyright
Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

Take-down policy
If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Downloaded from the University of Groningen/UMCG research database (Pure): http://www.rug.nl/research/portal. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.
In this section we present the principles of the experimental techniques of relevance to this work: Photon Emission spectroscopy (PES), COLd Target Recoil Ion Momentum Spectroscopy (COLTRIMS) and Translational Energy gain Spectroscopy (TES). PES and COLTRIMS were used in this work and as a complementary technique to COLTRIMS, also TES is briefly discussed.
2.1 Photon Emission Spectroscopy

2.1.1 Introduction

Photon Emission Spectroscopy (PES) makes use of the fact that ion-atom interactions often result in the emission of light. Target electrons can be excited by the interaction with the ion or can be captured into excited states of the ion projectile. The electrons in these excited states will decay to lower lying states by emission of (one or more) photons or electrons. By wavelength selective detection of the photons, the electronic final state population resulting from the ion-atom interaction can (in principle) be reconstructed.

2.1.2 PES: a short historical overview

Spectroscopy, the frequency selective detection of light, can be traced back to Isaack Newton. In 1666 he showed that with the aid of a prism, sunlight could be decomposed into its various colours. It would however take a while before the infrared (1800 Herschel) and ultra violet (1801 Ritter and Wollaston) parts of the spectrum were discovered. A decade later also the absorption lines in the solar spectrum were detected (Wollaston) and a few years later (1814) Fraunhofer extended these spectral measurements mainly due to improved optics to several hundred lines. In 1859 the connection between light emitting gasses in laboratories and the dark lines in the solar spectrum was made by Kirchhoff and Bunsen. Detailed measurements on the solar spectrum would then follow in 1868 by Angström and 1882 by Rowland. Rowland had employed a curved grating for his measurements and this technology is still widely used in many photon spectrometers today.

The atomic physics group in Groningen has always relied strongly on PES as one of the major techniques for studying ion-atom interactions. Collisions of ions like He\(^{2+}\), C\(^{4+}\), C\(^{6+}\) on atomic and molecular hydrogen, helium and lithium were investigated by Hoekstra et al. mainly in the energy range of 50 eV/amu to 12 keV/amu. Experiments on similar collision systems are still performed in our group nowadays (Bliek, Lubinski and Juhasz) however now in the astrophysically relevant energy region of 10 eV/amu to 1 keV/amu.

Another interesting class of collision systems studied in Groningen involves the interaction of ions with laser prepared atoms. By optically pumping the Na(3s→3p) transition a non-isotropic m-state distribution of the Na(3p) level is achieved. By using linearly polarized light the Na(3p) electron cloud can be aligned along the polarization axis. In this way an elongated "dumb-bell" or "peanut" shaped electron density is created and a dependence or anisotropy in electron capture on the parallel or perpendicular alignment of the electron cloud with respect to the ion beam can be measured (e.g. Schlatmann et al. and...
2.1 Photon Emission Spectroscopy

Schippers et al.\textsuperscript{34,35}).

2.1.3 PES: advantages and disadvantages

PES has some distinct advantages. For once PES is highly state selective. Spectrometers can usually resolve lines separated by a few Å and many different final states can normally be distinguished. Even in case of degenerate final states, one can often distinguish them, since they decay via different channels. Moreover, by employing polarization filters between the collision center and the detector, also information on the m-level distribution of the final state can be acquired.

Another advantage is that in general the experimental set-up can be kept relatively simple. The experimental set-up used for the lithium excitation measurements in this work, the stripped down version of ATLAS (the set-up used by Schlatmann and Schippers), was distinctly less complex than the set-up built and used for the COLTRIMS experiments.

Some disadvantages are that the emitted light usually ranges from far infrared to soft x-rays and that a single spectrometer cannot cover this kind of a spectral range. Another limitation of this method is the fact that some final state configurations do not decay via photon but electron emission. The reconstruction of the final state population will therefore usually not be complete. Moreover it turns out that it is very difficult to determine the absolute cross sections of a measured process with high precision due to the large systematic uncertainties associated with photon spectrometers.

The combination of small solid angles of spectrometers and low detector efficiencies mandates intense ion beams and/or dense targets. Last but not least no impact parameter sensitive information (the projectile scattering angle) is obtained with PES.

2.1.4 Conclusion

Photon emission spectroscopy has proven over the years to be a very effective tool for the study of single electron capture and target excitation in ion-atom collisions. Its main advantage, which can outweigh all the disadvantages, is that it is highly state selective, and that information about the (n,l,m) population of the final states in a collision system is obtained. Two disadvantages are that i) the final state population determination will usually not be complete because of gaps the spectral information and ii) no impact parameter sensitive information is obtained.
2.2 Cold Target Recoil Ion Momentum Spectroscopy

2.2.1 Introduction

In this section we introduce COLTRIMS (COld Target Recoil Ion Momentum Spectroscopy). COLTRIMS implies measuring the complete recoil momentum and subsequently reconstructing the final state and scattering angle distribution of an ion-atom collision. Review articles on COLTRIMS, containing many more references than the following brief overview, are available from Ullrich et al., Dörner et al. and Abdallah et al. Technical details our COLTRIMS experiment are discussed separately in Chapter 6.

2.2.2 COLTRIMS: a brief overview

The concept and techniques of COLTRIMS were introduced by the group of Prof. H. Schmidt-Böcking (Frankfurt) just before the 1990’s and in particular with the work of J. Ullrich and R. Dörner. By using static 30 K ($\Delta E = 4$ meV) gas targets they demonstrated that transverse recoil momenta could be measured corresponding to $\mu$Rad projectile scattering angles. In the 1990’s however, the real breakthrough for COLTRIMS came with the development of the ultra-cold supersonic gas jet (Mergel et al.) and also with sophisticated recoil ion extraction and detection techniques by using electrostatic lenses (Ali et al., Frohne et al.). These two improvements pushed the resolution of helium recoils to 1.2 $\mu eV$ (Mergel et al.). Moreover the solid angle for recoil detection increased to 4$\pi$.

With spectrometers installed at Kansas, Caen, Frankfurt, GSI, RIKEN, Berkeley and Rolla major advances in the field of scattering angle dependent, state selective single and double electron capture were made (Mergel et al., Abdallah et al., Cassimi et al.). Kinematically complete experiments on ionization (Dörner et al.), capture+ionization or transfer-ionization (Mergel et al.) and photo-ionization (Spielberger et al.) were performed. In order to obtain the kinematically complete information also the momenta of one or more electrons were measured with very high precision. This last achievement was another major breakthrough, and the apparatuses capable of detecting recoil and electron momenta are nowadays known as the reaction microscopes.

The field of COLTRIMS with photo-ionization saw already the introduction of Magneto-Optically trapped targets (Wolf and Helm) and the field of COLTRIMS with ion impact is nowadays pursued by three groups: Copenhagen (Li$^+$ +Na), Groningen (A$^+$+Na) and Kansas (Cs$^+$+Rb).
2.2.3 COLTRIMS: kinematics

In order to understand the basic principles of COLTRIMS (and TES, next section) we look at the kinematics of an ion-atom collision. We start by writing down the energy balance of a collision:

\[ E_i = \sum_j \epsilon_{j,i} + E_{p,i} + E_{t,i} = E_{p,f} + E_{t,f} + \sum_j \epsilon_{j,f} + \sum_j E_{j,f} = E_f \]  

(2.1)

- The labels p,t denote projectile and target,
- the labels i,f denote initial and final,
- E is the kinetic energy,
- \( \epsilon_j \) is the binding energy of active electron j and \( E_j \) is the energy of the (ionized) electron in the continuum.

In eq. 2.2 we define the momentum balance of a collision:

\[ \mathbf{P}_i = P_{p,f} + P_{t,f} + \sum_j P_{el,j,f} = \mathbf{P}_f \]  

(2.2)

and here we used:

- \( \mathbf{P}_p \) and \( \mathbf{P}_t \) for the momentum vectors of projectile and target, respectively a longitudinal (\( \mathbf{P}_{\text{long}} \)) and a transversal component (\( \mathbf{P}_{\text{trans}} \)), both with respect to the ion-beam direction.
- \( \sum_j P_{j,el,f} \) : the vectorial sum of all the ionized electrons’ momenta.

If we omit ionization and assume \( \Delta P/P_i \ll 1 \) (i.e. the momentum change is small compared to the total momentum in the collision) the momentum and energy conservation laws (eq. 2.1 and 2.2) can be rewritten into two elegant equations:

\[ P_{\text{long}} \equiv P_{\text{long},t,f} = Q/v - 0.5 \cdot (N_e) \cdot v \]  

(2.3)

\[ P_{\text{trans}} \equiv P_{\text{trans},t,f} = M_p \cdot v \cdot \Theta \]  

(2.4)

where we used:

- \( Q = \sum(\epsilon_{j,f} - \epsilon_{j,i}) \) is the change in binding energy of electrons due to the ion-atom collision. A negative Q-value implies an energy gain.
- \( v \) is the velocity and \( M_p \) is the mass of the projectile.
• $\Theta$ is the scattering angle of the projectile

The following conditions should be met in order for eq. 2.3 and 2.4 to be valid:

• $E_{t,f} \ll Q$ and $E_{t,f} + Q \ll E_{p,i}$.

• $P_{\text{trans},p,f} \ll P_{\text{long},p,f}$.

• $m_{el} N_c \ll M_p$ ($m_{el}$ = mass of the electron).

These criteria are basically specific cases of the already mentioned $\Delta P/P_i \ll 1$ condition. The conditions were found to be always valid in the ion-atom collisions studied in this work. Should this condition not be valid, then extra terms have to be added to eq. 2.3 and the longitudinal momentum can no longer be considered independent from the transversal momentum. This will be illustrated in chapter 3.2.2 with figure 3.3.

Also assumed are the initial conditions: $P_{p,\text{trans},i} \approx 0$ (the incident ion beam has a negligible divergence) and $P_{t,i} \approx 0$ (the initial momentum spread of the target is negligible). The first condition can be met reasonably easy in an experiment by using small diaphragms to collimate the beam, but the second condition ($P_{t,i} \approx 0$) demands a (ultra) cold target.

We now know that the longitudinal recoil momentum (eq. 2.3) is directly linked to the collision’s Q value i.e. the change in binding energy of the active electrons $\sum (\epsilon_{j,f} - \epsilon_{j,i})$. Capture of electron(s) into different final states (n,l) of the projectile will result in different Q’s and thus, as seen in eq. 2.3, in different $P_{\text{long}}$’s. Target excitation will also contribute to the Q and thus to $P_{\text{long}}$. We can illustrate all this with a very basic example: H$^+$ on He (figure 2.1):
Figure 2.1: Two examples illustrating the link between the electronic final state and the longitudinal recoil momentum. (A) single electron capture resulting in a small backward recoil momentum. (B) single electron capture + target excitation resulting in a large forward recoil momentum. $U(*)$ denotes the binding energy (in atomic units) of the active electron.

This example illustrates that $P_{long}$ can be positive or negative depending on the energy gain or loss in the reaction. Charge transfer collisions will usually result in negative $P_{long}$'s due to the transfer of an electron and the energy gain caused by the Coulomb interaction of the charged collision partners (see section 3.2.2) but capture into highly excited states, ionization and target excitation will as a rule create positive $P_{long}$'s.

Not yet discussed is the transversal momentum, a quantity directly linked to the impact parameter. A model for the ion-atom interaction should however be used to make this link. In section 3.2.3 we will show how the Over-the-Barrier (OtB) model can be employed. With the aid of the OtB model, the effective charge state of ion and projectile can be calculated at any moment during the collision. With this knowledge Rutherford scattering between projectile and target can be performed and an impact parameter→ transversal momentum conversion is obtained. Also obtained are transversal momentum distributions of target recoil ions. Also the Classical Trajectory Monte Carlo (CTMC) model will provide us with transversal momentum distributions of Na$^q+$ recoils.
2.2.4 COLTRIMS: advantages and disadvantages

A distinct advantage of COLTRIMS over PES is that the ion-atom interaction is measured in a direct and complete way. Since one does not measure the decay of the final state population but the population itself, there are usually no gaps in the measured distribution and even processes like ionization are accessible to the experimentalist. Moreover the scattering angles involved in the various ion-atom reaction channels can be measured and a direct link to the impact parameter range can be made. In the case of the reaction microscopes even the ionizing collisions can be reconstructed.

A distinct advantage of COLTRIMS is that no elaborate ion beam preparation is needed. The ion beam allowed to have an energy spread in the order of 1% without influencing the experimental resolution. Moreover COLTRIMS can be used in multiple-user ion beam facilities like storage rings, quite unlike TES.

The main disadvantage is that the typical recoil energies are very small (meV in the worst case), which makes the experiments very complicated (when compared to basic PES set-ups) and the desired momentum resolution difficult to obtain. This disadvantage is becoming however less of an issue since some of the most difficult parts of the experiment (like the ultra-cold gas jet and the 2D detectors) are nowadays commercially available. Another disadvantage is that the experiment is limited to those target species which can be prepared as cold gas or vapour targets, either by supersonic jet techniques (especially rare gases) or by laser cooling (alkali, earth-alkali and metastable rare gases).

2.2.5 Conclusion

In this section we discussed the COLTRIMS experimental technique. We discussed the relation between the longitudinal recoil momentum and the collisional energy gain and the relation between the transverse recoil momentum and the impact parameter. The main advantage of COLTRIMS is that one directly measures the physical quantities of interest (final state distributions and scattering angles), the disadvantage is that COLTRIMS experiments are (as a rule) a major technological challenge.

Although we concentrated in this section on the recoil momentum, it should be clear that the same information can be obtained from the momentum change of the projectile. To measure the very small projectile momentum changes requires the experimental approach called Translational Energy Spectroscopy (TES) and this is the topic of the next section.
2.3 Translational Energy Spectroscopy

2.3.1 Introduction

In this section we will discuss briefly the Translational Energy Spectroscopy (TES) experimental technique. TES consists of the precise detection of the projectiles energy (momentum) change e.g. due to electron capture. In this way one can measure, just like COLTRIMS, directly the final state distribution of an ion-atom collision. An overview of the TES experimental technique and a more complete list of references is given in a review by Janev and Winter.\(^6\)

2.3.2 TES: a short historic overview

Projectile energy loss spectroscopy has been extensively used for studying both elastic and inelastic collisions of singly charged ions with neutrals (Ziemba et al.\(^5\) et al. (1960), Aberth and Lorents\(^5\)). The energy spectroscopy is in these cases performed by a time-of-flight method. Measurements with doubly charged ions, which allowed an electrostatic analysis of the charge changed projectile, were performed by Siegel et al.\(^5\), Kamber and Hasted,\(^5\) Huber et al.\(^5\) and McCullough et al.\(^5\). State selective electron transfer, even with sub eV (Huber et al.\(^5\) 150 meV, Kobayashi et al.\(^5\) :100 meV) resolution was eventually possible. With slow, singly charged projectiles eventually an impressive 10 meV resolution (Itoh et al.\(^6\)) was achieved. The TES experimental method was also used with highly charged ions (references in Liljeby\(^6\)) and just like PES, used for to studying the electron capture and projectile scattering anisotropy from laser aligned and oriented targets (Aumayr et al.\(^6\), Dowek et al.\(^6\)).

Another important application of TES can be found in the field of ion-solid interaction where TES is known as Low Energy Ion Scattering (LEIS). LEIS is an important and still developing experimental tool, but an overview on LEIS would be beyond the scope of this work.

2.3.3 TES: advantages and disadvantages

The advantages are somewhat similar to the COLTRIMS experimental technique. TES gives direct and in principle complete information about the final state populations of an ion-atom collision system, which is an advantage over PES. Also TES is not (within reason) restricted in the choice of target atoms in the way COLTRIMS experiments are, and therefore also molecular targets are accessible (CO\(_2\) was for instance used by Itoh\(^6\) et al.). TES is however not well applicable to collisions with high energy projectiles because small energy changes of high-energy projectiles are difficult to identify. A major difference between TES and
COLTRIMS is that TES can be applied in collisions of ions with surfaces and COLTRIMS of course not.

TES usually does not provide, and this is a major disadvantage with respect to COLTRIMS, the full kinematic information (i.e. Q-value and scattering angle) of the various ion-atom reaction channels. TES can provide the Q-value distribution and sometimes scattering angle distributions, but only if a time-of-flight technique is used. TES also does not provide the \((n,l,m)\) final state resolution possible with PES and when compared to normal PES set-ups (as used in this work), TES can be a quite difficult experiment.