Differential Investigations of Charge Transfer Processes in Low Energy Ion-Atom/Molecule Collisions

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Abstract. The progress of differential studies of ion-atom/molecule collisions using the reaction microscope was reported. The state-selective and angular differential cross sections were measured for slow ion-atom collisions. In this paper we focus on the two-electron processes, namely double electron capture and transfer ionization, occurring in the collisions of ion impact on two-electron target system of helium and molecular hydrogen, respectively. The molecule Coulomb explosions were investigated by employing the negative ion characteristic. The transfer momentum dependence of two-electron processes for He\(^{2+}\) and H\(^{+}\) impact on He and H\(_2\) were compared, and the kinetic energy release in Coulomb explosions induced by He\(^{2+}\) and H\(^{+}\) was measured.

1. Introduction

The momentum imaging technique known as COLTRIMS (cold target recoil ion momentum spectroscopy) [1,3] has been developed very rapidly during the last decades, as well as the reaction microscope [3,4]. In these techniques complete measurements were performed for the final-state momenta of charged ions and electrons which were produced in ion, electron, photon as well as strong laser interaction with atoms or molecules [5-11]. Many experiments have been carried out on charge transfer concerning state-selective processes and imaging of electron momentum in the final-state momentum space [12], which revealed the dynamics of electron emission at various impact velocities. For example, C. L. Cocke presented a talk at the XXIII ICPEAC focused on the momentum imaging in atomic collisions [13].

In this paper, we will focus on two electron transfer for a bare ion impact of a two-electron target system, namely He and H\(_2\). We will compare the behavior for double electron capture (DC) and transfer ionization (TI) for the following collision systems: He\(^{2+}\) on He, He\(^{2+}\) on H\(_2\), and H\(^{+}\) on H\(_2\).

2. The experimental technique

When a particle (ion, electron, photon) interacts with an atom or molecule excitation, ionization, and/or electron capture can occur individually or simultaneously, and energy and momentum are exchanged in these processes. The initial momentum of the target ion carries rich information about
collision dynamics. The momentum imaging technique, known as COLTRIMS, was for the first time successfully used at the Frankfurt University [1,2]. Later, when it was combined with electron imaging the reaction microscope technique was born [3]. Nowadays, the COLTRIMS/Reaction Microscope techniques are performed worldwide in several groups: Frankfurt University, Max-Planck Institute for Nuclear Physics (MPI-K) in Heidelberg, CAEN, Kansas State University, GSI in Darmstadt, Stockholm University etc. These techniques have broad applications in atomic collision studies. Moreover, by adding magnetic optical trap into the reaction microscope a new technique has been developed, so-called MOTRIM, where the target can be cooled down to μK and even nK by laser cooling. Some pioneering experiments of this kind have been performed for alkali atoms [14-16]. In the following text we restrict ourselves only to describe the setup in ion-atom/molecule collisions

The momentum imaging principle is illustrated in figure 1. For charge exchange reactions the scattered ions with charge change are separated with the down stream electrostatic/magnetic analyzer and detected by the projectile position-sensitive detector (PSD-p), which gives the position \((X_{pk}, Y_{pk})(k = 1,2,\ldots)\) signals and arrival timing signals \(T_p\). The recoil ions and emitted electrons are extracted by a weak electric field \(E\). After passing through the time-of-flight spectrometer the recoil ion is recorded by PSD-r which is producing position \((X_{ri}, Z_{ri})\) and timing \((T_{ri}, i = 1,2,\ldots)\) signals. A uniform magnetic field \(B\) established by Helmholtz coils is set parallel to electric field which improves the electron detection efficiency. The electrons are detected by the detector PSD-e on the opposite direction which is producing \((X_{ej}, Z_{ej})\) and \((T_{ej}, j = 1,2,\ldots)\). In brief, the imaging system should include time-of-flight spectrometers for electrons and recoil ions and the time and position sensitive detectors for electron, recoil ion and scattered ion. The detectors for recoils and electrons are of multi-hit capabilities. For running the experiment, a multi-parameter data taking system working in event-by-event mode is necessary.

Figure 1. Schematic plot for reaction microscope in ion-atom/molecule collisions. PSD-r,e,p are the position sensitive detectors for recoil ion, electron and scattered ion, respectively. \(E\) is the extraction field, \(B\) the uniform magnetic field and \(E_0\) is charge state analyzer. The coordinates defined in the figure are the same throughout the paper.

A reaction microscope was built on the atomic beamline of electron resonance ion source at the Institute of Modern Physics (IMP). The atomic/molecular target beam is provided by one-stage supersonic gas-jet. The target gas is cooled to liquid nitrogen temperature and expanded through a 0.03mm nozzle and then collimated by a skimmer of 0.4 mm diameter. The background pressure in collision chamber is maintained to \(6.5\times10^{-7}\) mbar when target gas is supplied. The target density is estimated to be about \(10^{14}-10^{15}\) atom/cm\(^3\) at the collision zone [17]. Three dimensional focusing design is applied to recoil ion optics in order to reduce the influence on the recoil time and position resolution due to the extended collision zone. The delay-line microchannel plate detectors with an effective diameter of 75 mm are used for recoil ion, projectile and electron detection [18]. All the timing and position signals are handled by front-end electronics, then fed into the PC-CAMAC data acquisition system and recorded and stored in event-by-event mode. In the setup the electrons are constrained by a magnetic field with a homogeneity better than 0.6%.
3. Double electron processes in two electron systems

Helium atom and molecular hydrogen both have two electrons with opposite spins in their ground states. The former one is a one center system while the later has two centers. Helium has a spherical symmetry, while H₂ has a rotational symmetry around the molecular axis. The question arises: When an ion impacts on the two-electron system, He and H₂, do they respond differently in charge transfer processes? Does the H₂ molecule axis orientation influence the charge exchange process? The molecules H₂/D₂ were already used as targets in some experiments for studying Coulomb explosion. For slow highly charged ion impacts, post collision effects were discussed in [19, 20] by Ali and Dubois. For MeV impact energies, Cheng analyzed angular distribution of dissociated deuterons [21], Abdalah et al. [8] measured the electron emission pattern in transfer ionization process while Afaneh et al studied the saddle point electron mechanism [22].

3.1. He²⁺ on helium targets

The He²⁺ on He has been a showcase system studied for many decades, and was also explored in detail by recoil momentum imaging technique in recent years [23]. Here the main results will be described in order to have a comparison with molecular hydrogen H₂ target in the next sections. Figure 2 shows the two-dimensional plots for longitudinal momentum versus transverse momentum for double electron capture and transfer ionization at an incident energy of 30 keV (a velocity about 0.55 a.u.). The dominant transverse momentum range for DC and TI processes are 1-5 a.u. and 5-12 a.u., respectively. Generally, the bigger transverse momentum is, the smaller impact parameter in ion-atom collisions is. The above results reveal that the DC occurs at a larger distance than TI in He²⁺ on He collisions. Here double electron capture into ground state of projectile is the dominant channel because it is a symmetric resonant process. It was found that the contribution due to DC into doubly excited states in TI process was not important. Fricke et al. [21] performed completely unified theoretical description by considering the excitation, transfer and ionization channels simultaneously, and excellent agreement was found for the integral probabilities for all the reaction channels in the above collisions.

![Figure 2](image_url)  
Figure 2. (Colour online) (a) Two-dimensional plot for longitudinal momentum distributions versus transverse momentum for double electron capture and (b) transfer ionization in He²⁺ impact on He at 30 keV incident energy.

3.2. He²⁺ on H₂ Targets

When the target is changed to molecular hydrogen the question arises: Where are the two electrons which are loosely bounded compared to the electrons in helium, and how these two electrons will be transferred to the projectile? The DC and TI processes were both observed in the present
experiments. However, here one more point needs to be considered. Two protons will subject to Coulomb explosion by removal of the two bonding electrons. Also, more reaction channels are opened in the collisions: single electron capture (SC) and SC induced dissociation of the molecular ions (SCD), double electron capture and transfer ionization accompanied by the Coulomb explosion (CE).

At this low incident energy the direct ionization reaction and related dissociation channels can be neglected. The reaction channels are summarized as follows:

SC: $He^{2+} + H_2 \rightarrow He^+(nl) + H_2^+$ (1)

SCD: $He^{2+} + H_2 \rightarrow He^+(nl) + H^+ + H$ (2)

DC-CE: $He^{2+} + H_2 \rightarrow He^+(nl) + H_2^{2+} \Rightarrow He + H^+ + H^+$ (3)

TI-CE: $He^{2+} + H_2 \rightarrow He^+ + H^+ + H^+ + e$ (4)

In order to compare the momentum exchange in the collisions, the momentum of $H_2^{2+}$ ion needs to be reconstructed, namely the momentum transfer to the center of the mass of the system (CM). The two exploding protons have equal momentum with opposite direction in the CM system. As shown in figure 3, for the measured momentum $p_1$ and $p_2$ of the two protons in the laboratory system, by the momentum vector addition operation, one can easily obtain the CM momentum $P_{CM}$, and then the momentum due to Coulomb explosion can be deduced ($p_{ICE}$ and $p_{TICE}$). In the case of transfer ionization, the momentum of the continuum electron is neglected for reconstruction of the proton momentum.

![Figure 3](image1.png)

In the LAB system

Vector addition

In the CM system

Figure 3. (Colour online) The momentum reconstruction scheme for Coulomb explosion reactions.

![Figure 4](image2.png)

DC: $He^{2+} + H_2 \rightarrow He + H_2^{2+}$

TI: $He^{2+} + H_2 \rightarrow He^+ + H_2^{2+} + e$

Figure 4. (Colour online) Two-Dimensional plot for longitudinal momentum distributions versus transverse momentum for double electron capture (a) and transfer ionization (b) in $He^{2+}$ impact on $H_2$ at 30 keV incident energy.
The results of longitudinal versus transverse momentum for H\textsubscript{2} targets are shown in figure 4 for DC and TI, respectively. In transfer ionization, the recoil longitudinal momentum peak around -0.7 a.u., indicates that there are two main contributions. In one case the two electrons are captured into doubly excited states of the projectile and one electron is then emitted into continuum by Auger decay. In the other case the capture of one electron is accompanied by promotion into the continuum. In double electron capture the two captured electrons are mainly populated in the singly excited states of the projectile (one in ground state and one in excited state). The TI process has a much smaller transverse momentum compared to that of DC process. This may indicate that the TI occurs at larger impact parameter than DC. It is in clear contrast to the case of atomic helium target where DC occurs at a slightly larger distance than TI, and the transverse momentum extends to larger values.

3.3. \textsuperscript{H}^+ on H\textsubscript{2}

When \textsuperscript{H}^+ is used as a projectile, for double electron capture, the two captured electrons have to be in the ground state because \textsuperscript{H}^+ ion can only have one stable bound state. Here the Q-value of the reaction can be determined uniquely which is a big advantage in studying ion molecule interaction by recoil momentum imaging because it reduces one degree of the freedom of projectile. The negative ions production process has been employed by Chen et al. [25] for studying biomolecule fragmentation by ion impact. In the longitudinal momentum spectrum in figure 5, the DC peak is located almost at zero position which is in accordance with the prediction. The longitudinal momentum spectrum for TI is overlapped by DC spectrum in figure 5 for direct comparison. It is found that the TI peak almost sits at the same position as DC. The explanation is that the TI is a capture process where one electron is into the ground state of atomic hydrogen and the other is captured in the continuum of the projectile (ECC). Because the \textsuperscript{H} has a very shallow bound potential (0.75 eV), the position of ECC peak and the DC peak will have very small difference (0.036 a.u.) which is not distinguishable in our present experiment. Another case would be the excitation plus capture into ground state, but this could result in a forward peak of recoil ion (0.5 a.u.). It would be very interesting to measure the emitted electron in the TI process. This experiment is under way.

![Figure 5. (Colour online) Longitudinal momentum distributions for double electron capture and transfer ionization in \textsuperscript{H}^+ impact on H\textsubscript{2} at 15 keV incident energy.](image)

In figure 6, the transverse momentum versus longitudinal momentum are plotted for double electron capture and transfer ionization in the collisions of \textsuperscript{H}^+ on H\textsubscript{2} target. It can be seen that the DC process occurs at small impact parameters compared to the TI process (large transverse momentum). This is similar to the situation in the collision of \textsuperscript{He}^{2+} on H\textsubscript{2}. Though the absolute values of transverse momentum are different for the two systems, the transverse momentum for DC and TI are clearly in separated ranges.

Finally, we examined the proton energy in the CM system due to Coulomb explosion (CE) for molecular H\textsubscript{2} target. In contrast to the experiments with highly charged ions impact, in the case of \textsuperscript{He}^{2+} as projectile where post collision effects should be negligibly small, the scattered ions are either neutral (DC) or singly charged (TI). The proton CE energies are shown in figure 7 for \textsuperscript{He}^{2+} on H\textsubscript{2} (a)
and H⁺ on H₂ (b) collisions. It can be seen that for H⁺ impact, the peak of proton energy shifts to a larger value (11.8 eV) compared to the He²⁺ on H₂ system (where peak is around 10 eV). The width of the proton CE peak is broader for H⁺ impact than for He²⁺ impact. This will be checked more carefully in the coming experiment.

![Two-dimensional plots for longitudinal momentum distributions versus transverse momentum for double electron capture and transfer ionization in H⁺ impact on H₂ at 15 keV incident energy.](image1)

Figure 6. (Colour online) Two-dimensional plots for longitudinal momentum distributions versus transverse momentum for double electron capture (a) and transfer ionization (b) in H⁺ impact on H₂ at 15 keV incident energy.

![The proton kinetic energy in the CM system for double electron capture and transfer ionization for collision systems of He²⁺ + H₂ (a) and H⁺ + H₂ (b).](image2)

Fig 7. (Colour online) The proton kinetic energy in the CM system for double electron capture and transfer ionization for collision systems of He²⁺ + H₂ (a) and H⁺ + H₂ (b).

4. Summary

The progress of ion-atom/molecule collisions mainly performed using the reaction microscope at the Institute of Modern Physics was reviewed in this paper. State-selective and angular differential cross sections were measured for slow ion-atom collisions. The electron transfer in the two-electron target system, He and H₂, which has similar strength of electron-electron interaction, was experimentally studied by He²⁺ and H⁺ impact. The results for removal of two electrons from the targets are compared for transverse momentum exchange. It was found that for atomic He target, double electron capture occurs at a distance more larger than transfer ionization. On the contrary, the DC process occurs at a closer distance for H₂ target. When He²⁺ ion impact on H₂ molecule, one of the electrons prefers to go to the ground state of projectile while the other goes to higher lying states, which is different from He target where the two electrons almost exclusively go into the ground state of the projectile at the incident energy range of the present studies. The formation of a negative ion in double electron capture has the advantage to determine the Q-value more simply. This characteristic
can be further used to study the many-electron molecules where excitation cannot be avoided due to double charge transfer.

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