Differential Cross Section Measurements in Ion-molecule Collisions

Song Cheng

Department of Physics and Astronomy, University of Toledo, Toledo, Ohio 43606

A 14 m long beam line dedicated to study very small scattering angles in ion-molecule collisions has been set up in the University of Toledo Heavy Ion Accelerator (THIA) Laboratory. Together with position sensitive detectors for both the projectile and the recoil particles' detection, the beam line can be used to measure the projectile forward scattering angles of up to 2.5 milliradians (mrad) with a 0.025 mrad resolution, in coincidence with information on the recoil particles such as recoil charge states, energy, momentum and the molecular orientations.

INTRODUCTION

In studying of collisions of ions with molecular targets, many important quantities need to be measured in order to compare experimental data with theoretical expectations. Two of these quantities for the projectile are the forward scattering angle and the projectile momentum change. As for the recoil, the similar quantities are the recoil energy and the directions, which, in the case of molecular fragmentation, bear signatures of the molecular orientation at the moment of the collision. The more quantities that are measured, the more stringent tests the experimental data provide for the theories. Otherwise, the un-measured quantities need to be integrated over all possible values in theories and thus possibly mask important insights into understanding the collisions. In this sense, differential cross sections are more valuable than total cross sections.

For a heavy projectile with energies in the order of 1.0 atomic unit per mass (e.g. 25 keV proton), the forward scattering angle is typically of order of 1 mrad[1]. To obtain a meaningful differential cross section measurement for the forward scattering angle, a resolution of 0.01 mrad (1% of 1 mrad) or better is needed. Such a small angular resolution requires a long beam line that will allow the scattered projectile to travel a relatively long distance before it hits the detector. An additional requirement is that the projectile needs to be carefully collimated in order to achieve the required high angular resolution. On the recoil side, the main concern is how to effectively collect recoil particles and derive important parameters such as the recoil energy, momentum, and the molecular orientation in the case of the fragmentation. In this paper, we describe a setup which aims to meet these requirements and show an example using the setup to measure the forward scattering angle in coincidence with the target molecular orientations in the collision of 100 keV proton with hydrogen molecules. For a convenience of discussion, this example is used throughout this paper.

In this example, a theoretical calculation[2] predicts that for a fixed hydrogen molecular orientation, the forward scattering angle of the proton projectile shows an oscillatory structure in the differential cross section for the electron capture process (Fig. 1). Thus an experimental setup that can verify this prediction should be able to: (1) determine the hydrogen molecular orientation at the moment of the collision; (2) measure the forward scattering angle with a sufficient resolution to show the structure; and (3) separate the electron capture process

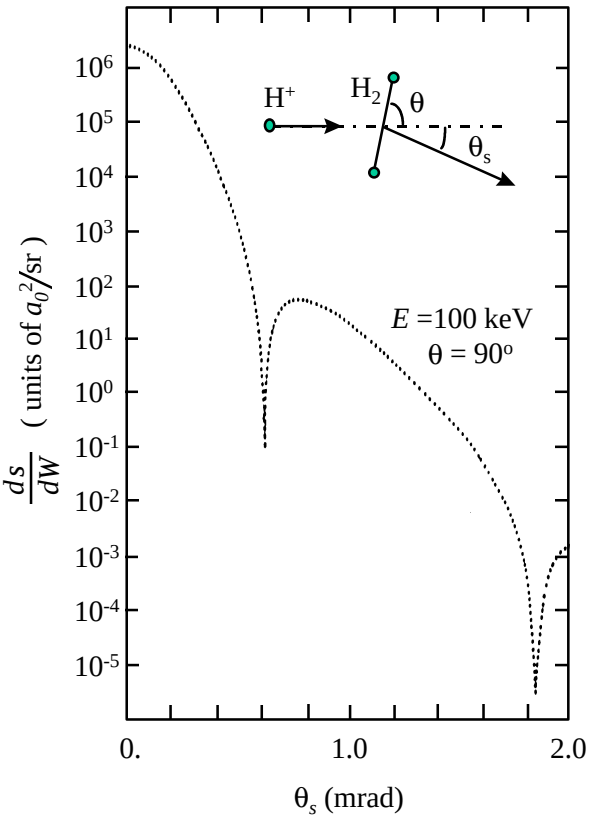

FIGURE 1. Differential cross section for electron capture process for 100 keV proton colliding with hydrogen molecules whose molecular orientation is fixed at θ = 90° (see reference 2).

CP475, *Applications of Accelerators in Research and Industry*,
edited by J. L. Duggan and I. L. Morgan

from the other scattering processes. All three requirements are generally needed in order to provide a stringent test of theories in ion-molecule studies.

BEAM LINE DESCRIPTION

The projectile ion beam is generated as follows: positive ions generated in a conventional ion source are extracted by a potential in the range of 6 to 20 kV applied to the ion source, mass analyzed by a bending magnet, and are accelerated to energies up to 250 keV. Then the ion beam is directed down a long beam line by an electric field deflector system. A schematic diagram of the long beam line is shown in Fig. 2. The beam is first collimated by the slits S1 to define a reference point for the rest of the beam line. The beam current is reduced from a couple of μA down to the nA scale after passing the slits S1 with a slit width in the order of 100 μm. Before traveling down the long beam line, the beam is slightly steered horizontally to the right by the deflector DP1 and if necessary, vertically by the deflector DP2. Steering the beam slightly in the horizontal direction directs the beam a little off the center to hit the slits S2 that is 6.5 meters downstream. Due to this long flight path, some projectile ions will interact with the background gas and capture or lose electrons. For example, for the proton beam, there will be H^0 and H^- impurities near the slits S2. With a help from two additional horizontal deflectors DP3 and DP5 near the slits S2, the charged beam is bent twice and only the H^+ is directed through a collimator of 0.1-mm diameter (the AP1 inside the collision chamber) to enter the collision chamber. The H^0 impurity beam is not affected by the deflections and is stopped on the right side of the slits S2. This pre-collision separation of the various charge state beams in the incident projectile beam is very important for this collision system since the charge exchange processes have a large cross sections in this energy range[3]. The pure charge state incident beam then enters the collision chamber to interact with the target. The combination of deflectors DP2 and DP4 provides a vertical steering to accommodate a possible minute misalignment that the collimator AP1 may have.

Inside the collision chamber, target molecules are introduced through a jet at 90 degrees with respect to the beam from the side opposite to the position sensitive detector (PSD2). The jet is made of a glass capillary array from the Galileo Electro-optics Corporation[4]. The pore diameter of an individual capillary tube is 25 μm and the length is 2 mm long. The size of the glass capillary array is about 1.25 mm. After interactions with the projectile ions, the molecular ions are extracted and accelerated to the PSD2 by an electric field set up by two plates G_1 and G_2 that are separated apart by 2.4 mm (see the exploded view of the extraction region in Fig. 3). The voltages on the plates, V_1 and V_2, are chosen in such a way that the electric field between the plates is strong enough to accelerate all molecular ions and molecular fragments in all directions after the collision towards the PSD2. Too strong electric field in this region is avoided since it would also deflect the projectile beam considerably. For hydrogen molecules, the maximum initial energy of the molecular fragments are about 10 eV. So at least 20 V potential difference should be maintained across the two plates, assuming the projectile interacts with the target right in the middle of the plates. In reality, the voltage difference should be larger to accommodate the beam size of the projectile and the case in which the projectile is a little off the center of the two plates.

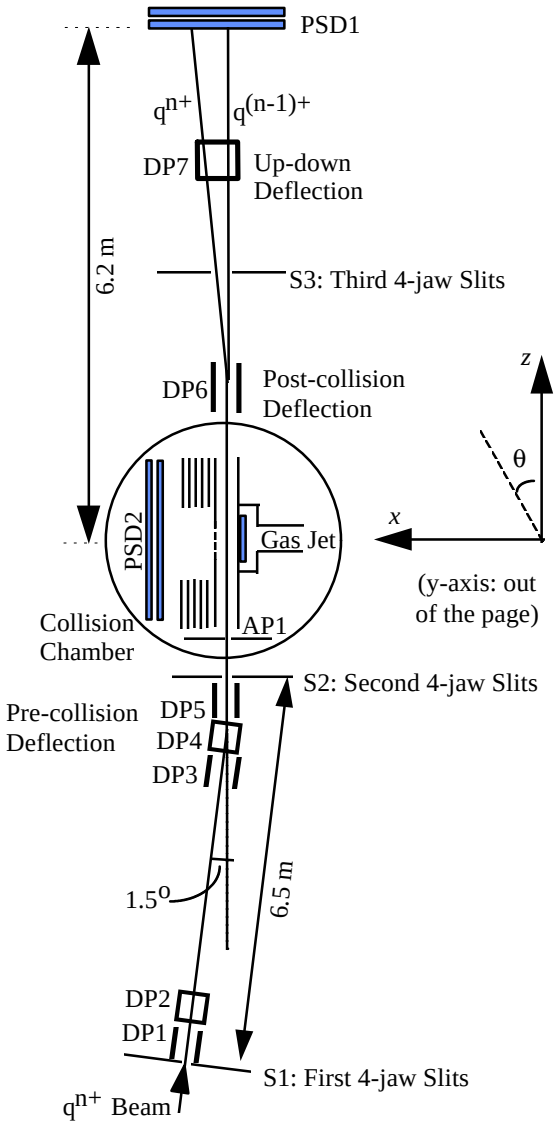

FIGURE 2. A schematic of the long beam line setup. DP1, DP3, DP5, and DP6 are horizontal deflectors. DP2, DP4, and DP7 are vertical deflectors. AP1: a 0.1-mm diameter aperture. PSD1 and PSD2: two-dimensional position sensitive detectors.

There is a 6.3 mm diameter aperture with a high transmission grid on the plate G_2 to limit the PSD2 solid angle so that it can detect only recoil fragments from collisions that happen in the vicinity of the gas jet. A trade-off exists when deciding the diameter of the aperture and it will be discussed further later in this paper. The extracted molecular ions or fragments after passing the aperture are accelerated by a uniform electric field (l_2 in Fig. 3) and then are allowed to drift in a field-free region (l_3 in Fig. 3) before hitting the two-dimensional position sensitive detector. A measurement of the position (y,z) that ions hit the PSD2 and the time-of-flight that ions take from the jet to the PSD2 is sufficient to allow a calculation of the molecular orientation of the target molecule at the moment of the collision.

A second position sensitive detector PSD1 is placed about 6.2 meters downstream in the forward direction at the end of the beam line to detect the scattered projectile beam. The beams of different charge states are separated by the post-collision deflector DP6 so that they hit the PSD1 at different locations horizontally. The relative counts at these horizontally separated groups represents the percentage of beam that changes charge states after beam interacts with the target gas jet. The position spread within each group with respect to the center of the group itself is proportionally related to the forward scattering angle θ_s as defined in the Fig. 1. At about the midpoint between the post-collision deflector DP6 and the PSD1, there is a vertical deflector DP7 which alters the beam trajectories slightly in the vertical direction. The purpose

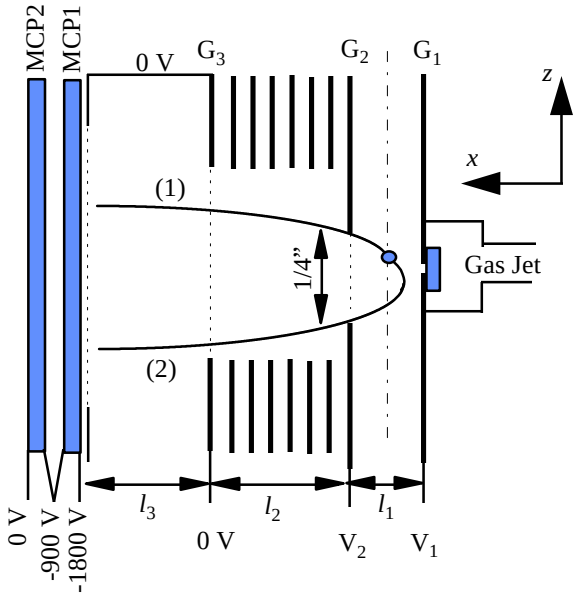

FIGURE 3. An exploded view of the extraction region and the detector PSD2. MCP: micro-channel plates. l_1: the scope of the extraction field set up by the V_1 and the V_2. l_2: the scope of the acceleration field set up by the V_2 and the ground. l_3: the scope of field-free drift region. A 1/4" aperture is on the plate G_2.

of the DP7 is to estimate the contributions to the beam impurities developed *after* the projectile beam passes the slits S2 and interacts with the background gas before it interacts with the gas jet. Since for the beam that changes charge states during the path from the deflector DP6 to the deflector DP7, it will be separated vertically on the PSD1. The relative counts at these vertically separated groups represents the percentage of beam that changes charge states when the beam travels through the background gas of "target thickness", which is equal to the background gas density multiplied by the separation from the deflector DP6 to the deflector DP7. The background gas densities in locations *before*, *in*, and *after* the collision chamber are monitored or estimated so that the background signal can be accounted for in the data. The coincidence between timings from the PSD1 and the PSD2 relates the forward scattering angle with the molecular orientation of the target molecules at the moment of the collision.

Placing the PSD1 at about 6.2 meters downstream from the target gas jet is to gain a sufficient angular resolution needed to observe the narrow interference structure as shown in the Fig. 1. For the PSD1, that has a linear resolution of about 0.15 mm, the angular resolution that can be achieved by this arrangement is about 0.024 milliradian, or about 0.0014°, which should be sufficient to resolve the oscillatory structure shown in the figure. The size of the position sensitive detector PSD1 is about 20 mm in radius. That means it can cover the forward scattering angle up to 3.2 milliradians if the beam is let to hit in the middle of the detector. In reality, a reliable coverage is about 80 % of 3.2 milliradians, to be 2.5 milliradians.

In order not to deteriorate the 0.024 milliradian angular resolution achieved on the PSD1, the incoming projectile beam has to be collimated at least in the same order of magnitude, or about 0.012 milliradian. To satisfy this condition, the projectile is collimated by two slits S1 and S2 that are separated by 6.5 meters upstream. In order to close these slits down to a zero width, the opposite slits are designed in such a way that they will not touch each other when closed down to a zero width and will overlap about one millimeter if they are continued to close down. No absolute calibration or alignment is needed for these two precision-micrometer-driven slits and the zero slit width is obtained by watching the projectile counting rates to go to zero as the slits are closed down. A 0.012 milliradian divergence of projectile calls for slit openings of 80 microns for both the slits S1 and the slits S2. The real openings of the slits are actually dictated by the beam current that the forward PSD1 can handle. For example, the THIA accelerator routinely delivers about 5 microamperes of proton beam onto the slit S1 with a beam diameter of about 5 mm. However, a position sensitive detector can normally only handle about 10,000 particles per second. So the beam current has to be reduced down

to less than a few femto-amperes when the beam reaches the PSD1. Therefore, very narrow slit widths are needed to cut the beam intensity down to an acceptable range, resulting in slit openings of approximately 50-60 microns.

APPLICATION TO p + H_2 COLLISION

We would like to use this setup to measure the forward scattering angle θ_s in coincidence with the molecular orientation for electron capture process in p + H_2 collisions. The proton energy is chosen to be 25 keV and 100 keV in order to directly test the theoretical predictions by Deb *et. al.*[2]. Their predictions can be summarized as: (1) for a fixed molecular orientation, the forward scattering angle q_s shows an oscillatory structure; (2) as the projectile energy changes, or the fixed orientation θ changed to other than 90°, the oscillatory structure changes accordingly.

The main difficulty in this application is how to determine the molecular orientation at the moment of the collision. For a sufficiently high energy proton impact, there is a certain probability[5,6] that, after the electron capture event, hydrogen molecular ions are left in one of the dissociative states. For example, for 25 keV proton impact on H_2, the total capture cross section s_C is about 5 $\times 10^{-16}$ cm^2 (ref. 7), and the cross section for transfer ionization s_{TI} ($p + H_2 \Rightarrow H^0 + H_2^{++} + e$) is about 1.0×10^{-17} cm^2 (ref. 5), and the cross section for transfer excitation s_{TE} ($p + H_2 \Rightarrow H^0 + (H_2^+)^*$) is about 1.0×10^{-16} cm^2 (ref. 5). Therefore, when electron capture events happen, 22% of the recoiling hydrogen molecular ions will be left on the dissociative states which will dissociate into H^+ and H^0 in the case of $(H_2^+)^*$, or dissociate into two H^+ ions in the case of H_2^{++}.

When the molecule ions undergo dissociation, the momenta of the two partners are equal in magnitude and opposite in direction (the thermal energy of the hydrogen molecules can be neglected when compared with the dissociation energy carried away by these two partners, which in the order of a few eV to 10 eV each), and the directions of the momenta determine the molecular orientation. Since the collision time is much shorter than the rotation time of hydrogen molecules (which is in the order of 10^{-12} second), the molecules can be considered 'frozen' at the moment of the collision. The post-collision interaction between the projectile and the dissociated ions has little effect on the trajectory of the dissociated ions[8]. Therefore, the probability of finding the dissociated particles in a neighborhood of a fixed orientation is proportional to the solid angle dW associated with the orientation. For example, in the case of θ = 90° and Δθ = ±2.5°, the probability is about 4.3%, assuming that molecular fragments in all directions are collected and the azimuthal symmetry is valid.

In order for the PSD2 to collect the dissociated particles of kinetic energy of a few eV (up to about 10 eV) and moving in all directions, V_1 is set to 491 V, and V_2 to 453 V. The lengths l_1, l_2, and l_3 are set to 2.4 mm, 19.6 mm, and 20.8 mm respectively to maximize the time-of-flight resolution for the fragments[1]. Measurements of the time-of-flight of the fragments and the position (y,z) where the fragments hit the PSD2, theoretically, are enough to determine the molecular orientation. However, the determination of the orientation gets complicated due to the fact that the gas jet is not an ideal point source but rather has an extended spread along the beam axis. In this case, the molecular fragmentation can happen anywhere along the beam seen by the PSD2. Therefore, the origin of the collision can not be defined and the molecular orientation can not be calculated. There is no solution to this for a given jet. To cure the problem to some extent, an aperture is placed on the plate G_2 to restrict the view of the PSD2 to the region near the jet. The influence of the aperture on the fragments to be detected depends on which channel the H^+ comes from: the $(H_2^+)^* \Rightarrow H^+ + H^0$ channel or the $H_2^{++} \Rightarrow H^+ + H^+$ channel. For the $H^+ + H^0$ channel, only the H^+ is detected. The origin of H^+ needs to be known in order to calculate the molecular orientations. In this sense, an aperture with a smaller diameter is preferred. However, a smaller diameter aperture needs a stronger electric field in the extraction region l_1 to ensure that all fragments pass the aperture without being blocked. A stronger electric field would deteriorate the time-of-flight resolution and in turn deteriorate the orientation measurement. For the $H^+ + H^+$ channel, it does not matter where the origin is, since the momentum conservation dictates that if one H^+ comes with an angle θ, then another must come with 180°− θ. In this sense, one would prefer to have no aperture at all. Based on the initial velocity of the H^+ fragments, the geometry of the extraction region, and the diameter of the PSD2 (40 mm), a compromise of 6.36 mm diameter is used for the aperture in this setup.

An estimate of the coincidence rate can be made based on the known cross sections. As stated earlier, for 25 keV proton impact on H_2, the total capture cross section $s_C = 5 \times 10^{-16}$ cm^2, $s_{TI} = 1.0 \times 10^{-17}$ cm^2 for the transfer ionization, and $s_{TE} = 1.0 \times 10^{-16}$ cm^2 for the transfer excitation. From the relation $Y = s(nL)N$, where N is the projectile rate; (nL), the product of the target density and the scattering length; is the target thickness; s, the cross section for a specific process; and Y, the rate of the specific process, it is reasonable to assume the (nL) to be $2 \times 10^{14}/cm^2$ to keep the single collision condition valid. Under this condition, the total capture rate is $Y_c = 0.1N$ and the rate for transfer ionization is $Y_{TI} = 0.002N$, and for transfer excitation, $Y_{TE} = 0.02N$. Since the projectile detector PSD1 can handle about 10,000 particles per second, and we are only interested in the charge exchange processes, the projectiles that are not involved in charge exchange process (H^+ in

this case) are blocked from hitting the projectile detector by the slits S3, and only the H^0 ions are allowed to hit the detector. Assuming that, after interaction with the target, the projectile H^0 rate is 10,000/s, then about 2,000/s are from the transfer excitations (of cross section 1.0×10^{-16} cm^2) and 200/s are from the transfer ionization (of cross section 1.0×10^{-17} cm^2). All these transfer excitation and transfer ionization events lead to the dissociation of hydrogen molecules. The events whose molecular orientations are at $\theta = 90° \pm 2.5°$ regardless the azimuthal angle are about 86/s (4.3% of the 2000/s) and 8.6/s (4.3% of the 200/s) respectively for the transfer excitation and transfer ionization.

The coincident events can be dramatically increased by the following consideration. When we examine the differential cross section shown in the Fig. 1, we realize that the oscillatory structures lie totally beyond the scattering angle $\theta_s > 0.4$ milliradian and only account for about 1% of the total cross section. That is to say that 99% of the electron capture events have forward scattering angles $\theta_s < 0.4$ milliradian and do not contribute to the oscillatory structures. Therefore if we block the events that hit the projectile detector PSD1 in the center within the radius of $0.4 \times 10^{-3} \times 6.2$ m = 2.4 mm, we can reduce the projectile counting rate by a factor of 100 without throwing away any events which contribute to the oscillatory structures. In this way, the beam intensity can be increased 100 fold, which in turn increases the molecular fragmentation rates by 100 fold without saturating the detector. An easy solution to block the beam from hitting the center of the detector is to use a channel plate that has a hole in the center, which is commercially available from the channel plate manufacturer, Galileo Electro-optics Corporation[4].

The molecular orientation of $\theta = 0°$ which is parallel to the beam direction can not be measured by the above discussed procedures for the $H^+ + H^+$ channel. In this case, the time-of-flights for the both H^+ are the same and will be counted as one. However, the use of the isotopic HD molecule will overcome this difficulty. Though each fragment still has the same initial momenta, their initial velocities are quite different, which means their time-of-flights are quite different also.

ACKNOWLEDGMENTS

The author would like to thank Dr. Cocke for discussions on the feasibility of this experiment and Dr. Kamber for encouragement which stimulated me to present this paper in this meeting.

REFERENCES

1. Cocke, C. L. and Olson, R. E., *Physics Reports* **205**, 153-219 (1991).
2. Deb, N. C., Jain, A., and McGuire, J. H., *Phys. Rev. A* **38**, 3769-3772 (1988).
3. Stier, P. M. and Barnett, C. F., *Phys. Rev.***103**, 896-907 (1956).
4. Galileo Electro-optics Corporation, Glass Capillary Array Data Sheet No. 7100, and Microchannel Plate Data Sheet No. 9000.
5. Lindsay, B. G., Yousif, F. B., Simpson, F. R., and Latimer, C. J., *J. Phys. B: At. Mol. Phys.* **20**, 2759-2771 (1987).
6. Cheng, S., Cocke, C. L., Kamber, E. Y., Hsu, C. C., and Varghese, S. L., *Phys. Rev. A* **42**, 214-222 (1990).
7. Tawara, H., Kato, T., Nakai, Y., *Atomic Data and Nuclear Data Tables* **32**,235-303 (1985).
8. Dunn, G. H. and Kieffer, L. J., *Phys. Rev*. **132**, 2109-2117 (1963).