Differential energy loss methods for studying ionization and fragmentation by low-energy leptons and ions

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Differential measurements are a powerful tool for providing information about ionization and fragmentation dynamics and channels. This work describes various experimental methods capable of providing highly differential information as functions of the energy deposited by several hundred to a few thousand eV positron, electron, and ion beams. Techniques compatible with conditions where small differential ionization rates must be measured or where the differential rates are small with respect to the overall ionization rates are described and compared. Examples being used at the Missouri University of Science and Technology to generate doubly and triply differential information for positron and electron impact and in low-energy ion impact energy loss measurements are provided.

Keywords: Energy loss; stopping power; electron spectroscopy.

1. Introduction

In atomic collision physics studies of nanoscale structures and interactions have been in progress for many decades. Specifically charged particles and photons are used to excite or ionize particular electrons from individual atoms or molecules and the various ionization or fragmentation channels and/or the interaction dynamics are investigated. For obtaining the maximum information about interactions or how the atoms and molecules respond when particular electrons are excited or ionized, highly differential information of the post-collision particles is required.

Today much emphasis is on using ultrashort intense pulses of photons, either from synchrotrons or lasers, to deposit energy and initiate these processes. Using photons is attractive for two reasons. First, in multiphoton excitation and ionization processes, each photon deposits a precisely known amount of energy. Second, in the case of molecular targets, pulse widths of a few femtoseconds or less are readily achievable and these times are sufficient to freeze the nuclear motion during the time that the energy is deposited. The ultimate goal of such studies is to probe and image the dynamics during the ionization or breakup process by making the pulse short enough such that the bound electrons can also be considered to be frozen in place while the energy is deposited. But, to achieve this will require pulse widths on the order of a few attoseconds and this requires more than an order of magnitude improvement beyond what the leading groups have been able to achieve to date.

On the other hand, as shown in Fig. 1, charged particles can easily achieve subfemtosecond and attosecond interaction times, plus they generate electric field strengths which are comparable in magnitude to intense photon pulses. Unfortunately, charged particles have a major disadvantage with respect to photon induced studies in that the amount of energy deposited during the interaction can be anything between zero and the entire available energy. This is because for charged particle impact the energy deposition varies with the distance between the charged particle and the target atom or molecule, i.e., it depends on the time as the particle passes by, on the impact parameter and, ultimately, also upon the location of the bound electron during the interaction. However, this limitation can be overcome if the energy loss is measured for each ionization or fragmentation event. This approach is currently being used at the Missouri University of Science and Technology (formerly the University of Missouri-Rolla) using sub- and few-keV positron, electron, and ion beams. In this paper we will describe the methods we are using, or have contemplated using, in three particular energy loss measurements. These studies include triply differential single ionization by positron and electron impact, doubly differential single and multiple ionization by positron and electron impact, and ionization and fragmentation of molecules by low-energy ion impact.

In each of these studies, the differential yields of scattered or ejected particles are measured in coincidence with...
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IGURE 1. a) Collision time as a function of impact energy for leptons and protons. b) Electric field felt by a target electron as electrons and protons approach and leave. The vertical double ended arrow shows values typical for intense photon beams. c) Schematic comparison of energy deposited by photons and particles of energy \( E_0 \).

target ions produced in each event. More importantly, they are measured as functions of the energy which is deposited. This is done by measuring the initial and final projectile energies. To obtain this differential information in our particular studies requires using methods compatible with conditions where either the differential signal rates are small or where small signal rates must be separated from much larger overall or background rates. It also requires efficient methods for measuring the energy loss. Finding methods to overcome small signal rates is particularly important for our positron studies while extracting small signal rates from large overall rates is important for our doubly differential and ion impact studies. This paper will describe the methods being used and provide some examples of data in order to demonstrate how successful these methods are.

2. Methods for measuring small differential ionization rates

As stated, one of our criteria involves methods compatible with very small signal rates. This is because for positron-impact ionization studies using radioactive sources, beam intensities that can be generated are a few tens of femtoamps. However, losses associated with transporting and collimating the beams in order to perform differential studies reduces the beam intensity to a few femtoamps or less. (Note that for total cross-section measurements, magnetic confinement and transport of the beam can be used which means little or no loss during transport. But magnetic transport precludes looking at the interaction kinematics of the various particles.) These low-intensity beams mean that in interactions with diffuse gaseous targets the overall signal rates are small and the differential rates are extremely small. Therefore special techniques for working with very small signal rates are required.

To compare various methods that could be used for measuring the differential ionization, let us assume that \( N_B \) beam particles per second interact with a target of density \( N_T \) and length \( l \) and that a detector having efficiency \( \eta \) detects all reaction products emitted into solid angle \( \Delta \Omega \). For simplicity, let us assume that the product emission is isotropic. The measured signal rate \( N \) is then given by

\[
N = \sigma N_B N_T l \Delta \Omega \eta T D. \tag{1}
\]

Here \( \sigma \) is the overall ionization or fragmentation cross section, \( T \) accounts for any reduction in transmission due to grids between the interaction region and the detector, and \( D \), the duty cycle, accounts for the percentage of time data are collected.

At intermediate positron energies, the cross section is approximately \( 10^{-16} \) cm\(^2\). Assuming a target pressure and length of 1mbar and 0.2 cm respectively and solid angle \( \Delta \Omega \), this gives an expected signal rate for ALL reaction products into this solid angle of approximately

\[
N(Hz) \sim 5 \Delta \Omega \eta T D \text{ per fA of beam} \quad \tag{2}
\]

If a spectrometer is used to select particles within a certain energy range \( \Delta E \), then the expected differential signal rate, \( dN \) is

\[
dN(Hz) \sim 5 \Delta \Omega \Delta E \eta T D \text{ per fA of beam} \quad \tag{3}
\]

This equation applies for doubly differential single ionization studies where one either measures the energy and angles of the ejected electron or the post-interaction projectile.
energy and scattering angle. In triply differential single ionization studies, the angles of both and energy of one must be measured and Eq. 3 would be modified by including $\Delta \Omega \Delta \varepsilon \eta T$ values for another detector, which would reduce the rate even further.

One very powerful method of overcoming this problem is to use the recoil ion momentum spectroscopic (RIMS) method to measure the ejected electron and recoil ion energies and angles. See Fig. 2. Here, time- and position-sensitive detectors are used to measure the electron and ion positions and flight times from which the energies and angles, or momentum components, are calculated. The reader is referred to reference 1 or other articles in the literature for detailed descriptions of this technique and various studies that have been performed using this technique. The advantages of the RIMS method are that the detection solid angle can be $4\pi$ and that many energies and angles are probed simultaneously. The disadvantages are that the resolution depends on the interaction volume and on the target temperature and that measuring the flight time requires knowing when the interaction took place. Producing a localized interaction volume means using a highly collimated beam and supersonic jet targets. However, supersonic jets typically have densities that are an order of magnitude smaller than the density used in Eqs. (2) and (3). Also, reducing the positron beam diameter in order to generate a small interaction volume causes a drastic reduction in beam intensity. In addition, the only viable method for measuring the interaction time in lepton impact is to use a pulsed beam. For the necessary time resolution, the duty cycle must be on the order of $\sim 0.001$, i.e., a few ns wide pulse every few $\mu$s. As can be seen from Eqs. (2) and (3), these combined factors lead to extremely small signal rates. Because of these factors, the RIMS method is not compatible with positron beam intensities which are available today.

Other methods to measure the energies and angles are shown schematically in Fig. 3. Here, various types of spectrometers measure the ejected electron, or scattered projectile, energies at a specific angle. The advantage of these methods is that the energy and angle are measured directly; the disadvantages are that using traditional spectrometers as shown schematically in Fig. 3a limits the measurement to one energy and angle at a time. Plus the solid angle tends to be small and generally the energy resolution, defined by $k = \Delta E/E$, is coupled to the solid angle. This limits the ability to significantly increase the solid angle while maintaining a reasonable energy resolution. Using typical values for the various parameters which are given in the figure and Eq. 3 means that the differential signal rates for a $1 \text{ fA}$ beam are $\sim 0.007 E^{-1}$ s$^{-1}$. Note that the energy window increases with the detected energy, hence the signal rate does also. We are employing this method in our doubly differential electron impact studies for small scattering angles where the signal rates are sufficiently large and where the beam intensity can be increased to hundreds of femtoamps. We are also using this method to measure the scattered positron energy (which gives the energy loss) and scattering angle in our positron triply differential studies. However, for our positron studies where the rates are small a specially designed spectrometer equipped with a position sensitive detector is used. The parameters for

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**Figure 2.** Schematic of recoil ion momentum spectrometer showing the intersecting beam and supersonic gas jet and collision fragments.

**Figure 3.** Various types of spectrometers for measuring differential electron emission. Typical parameters associated with each are provided. See text for additional details.

**Figure 4.** Two-dimensional spectra showing scattering and energy loss when 500 eV positrons singly ionize argon atoms. Higher intensities are indicated by brighter colors. The yellow circle indicates the location and size of the positron beam. Its vertical position defines the zero scattering angle.
this spectrometer are \( \Delta \Omega \sim 0.03 \), \( T = 0.8 \), and \( \Delta E \) is totally adjustable because of the position sensitive detector. As a result, for scattering angles less than approximately 7 degrees, the signal rates are large enough to use, as shown below in Fig. 4. In general, however, pico- or nano-amp, or larger, beam intensities are required to perform studies using this general type of spectrometer.

This method is improved even more by using a retarding potential spectrometer, as shown in Fig. 3b. Here, a larger solid angle can be obtained while still maintaining a good energy resolution. Energies are resolved by using a biased grid between two grounded grids to remove all energies less than a certain value. By shifting the bias back and forth between two values and subtracting the signals, the differential signal for energies between the two values is obtained. Hence, the energy resolution is totally adjustable while the solid angle is only restricted to be such that the emission angle remains well defined. The negative aspects are that the grids introduce transmission factors and because a subtraction process is required, the duty cycle is 1/2 and statistics become important. Again, typical values for the various parameters are given in the figure. With respect to the “simple” spectrometer, a gain of approximately 7 in signal rate is obtained. We are using two spectrometers of this type in our doubly differential electron impact measurements to obtain data at larger scattering angles.

The solid angle can be maximized by placing a grid as close as possible to the interaction region and applying a strong field between the grid and a position sensitive detector, as shown in Fig. 3c. Using position information, many angular bins can be measured simultaneously. This method is used in our positron triply differential studies. See Ref. 2. In order to determine the emitted electron energies, coincidences with scattered projectiles where the energy loss is measured are used. Again, parameters for this type of detector are provided and show that the expected overall signal rate is approximately 0.4 Hz for a 1 fA beam. In our positron studies, we divide this into approximately 12 angular bins and measure 12 differential electron emission signals simultaneously. As stated, the electron energies can be determined via coincidences with scattered projectiles and doubly differential yields for electron emission can be measured with this detector.

For our triply differential information, two options exist. One is to select specific projectile scattering angles and energy losses using the projectile spectrometer of type 3a mentioned above and perform coincidence measurements of the differential electron emission using method 3c. The other is to select specific electron emission angles using method 3c and measure the projectile scattering angles and energy loss using method 3a equipped with a position sensitive detector. Fig. 4 shows an example for single ionization of argon by 500 eV positron impact. Here, when coincidences with all electrons are selected, we see a well defined ridge of ionization as a function of energy loss and scattering angle. Ionization below “zero” scattering angle (with respect to the primary beam location which is shown as a yellow circle) is due to binary interactions where the target nucleus simply acts as a spectator and the scattered and ejected particles leave in opposite hemispheres. Ionization above “zero” is due to recoil interactions where the ejected electron is turned around by the target nucleus which means the scattered and ejected particles leave in the same hemisphere. The data shown in Fig. 4 clearly demonstrate that highly differential data can be obtained even when the overall signal rates are extremely small.

3. Methods for separating small signal rates from large background rates

Each of our studies is based upon detecting the target ion in order to determine the degree of ionization or fragmentation pattern. However, except for our positron-impact studies, to measure the differential yields as a function of energy loss only a very small portion of the recoil target ions are in coincidence with the post-collision projectiles of interest since most recoil ions are produced for small energy losses and scattering angles. Much rarer are interactions where the scattering angle and/or energy loss is large. Thus the

![Figure 5](image5.jpg)  
**Figure 5.** Time of flight spectra for coincidences between scattered electrons and target ions for 500 eV electron impact on krypton. The observation angle was 45° and the energy loss was approximately 40 eV. The left figure is for DC extraction of the recoil ions and a data collection time of 1 day. The right figure is for pulsed extraction using a higher beam intensity and a data collection time of 7 minutes.

![Figure 6](image6.jpg)  
**Figure 6.** Schematic of pulsed target ion extraction being used in our lab. The bottom (near) extraction plate is biased slightly positive. Normally the top (far) extraction plate is grounded but when a scattered electron which has suffered energy loss \( \Delta E \) is detected, a large positive pulse is applied. See text for additional details.
The experimental problem is to isolate the small signal rate of interest from large "background" rates, i.e., to isolate the real coincidences involving the few recoil ions of interest from the random coincidences resulting from the vast number of recoil ions that are produced overall. As an example, Fig. 5a shows a time of flight spectrum for 500 eV electron impact on krypton. Coincidences between recoil ions and electrons which have lost approximately 40 eV and scattered through 45 degrees were measured for one day using a recoil ion rate of approximately 20 kHz. As seen, because of the small scattered electron detection rate, only about 6 coincidences per time channel were recorded. However, because of the large recoil ion rate, random coincidences dominate and there is no sign of any peaks to indicate real events.

In sharp contrast is the time of flight spectrum shown in Fig. 5b where a peak is readily apparent after only a few minutes of measurement. Here the random coincidences were drastically reduced by using a pulsed recoil ion extraction field as illustrated in Fig. 6. Our pulsed extraction technique normally applies a small positive voltage to the near extraction plate. This avoids detecting any "false" coincidences associated with any previously produced ions because they are expelled from the extraction region before the interaction of interest takes place. The time required to expel these ions places an upper limit on the beam current that can be used. Once a scattered projectile is detected, a large positive voltage pulse is applied to the far extraction plate. This extracts the ion associated with the detected projectile, i.e., with a projectile that has scattered through a particular angle and which has lost a specified amount of energy. As a result, random backgrounds are dramatically reduced. In addition, higher beam currents can be used (subject to the limitation mentioned above) which significantly improves the data collection rate. Another advantage of the pulsed extraction method is that the interaction occurs in a field free, or very low field, environment. Thus, studies using much lower beam energies can be performed and distortions to the trajectories of the outgoing electrons are minimized.

However, when using pulsed extraction care must be taken to shield the various detector signals from the extraction pulse. Good shielding is required since the extraction pulses can be many tens of volts or higher whereas the recoil ion signals are on the millivolt level. In addition, tests are required to ensure that the pulse width is sufficient to remove the desired ions from the interaction region. The minimum pulse width depends on the ion charge and mass as well, on the pulse magnitude, and on the dimensions of the interaction region and exit aperture. If the width is too small, the time of flight spectrum can be distorted or disappear; if it is too large the number of random coincidences increases. Overall, however, it enables very weak differential signals to be measured. Using this method we were even able to measure triple ionization of krypton for scattering angles up to 90 degrees and energy losses up to half of the initial kinetic energy. [3]

4. Methods for measuring the projectile energy loss

As stated, each of our studies requires knowing how much energy is deposited during each event and this is determined from measurements of the initial and final projectile energies,
i.e., from measuring the projectile energy loss. We have already described how this is done in our positron and electron impact studies where both the energy loss and scattering angle were measured. In our low-energy ion impact studies, because the scattering angles are quite small the concentration is simply on measuring the energy loss. Various options capable of separating the different energy losses include electrostatic or magnetic deflection, a Wien filter, or time of flight techniques. These are shown schematically in Fig. 7.

In the electrostatic deflection method, shown in the upper left portion of the figure, the post-collision beam of charge \( q \) and energy \( E \) is deflected as it passes through a uniform electric field produced by parallel plates of length \( L \), separation \( d \), and voltage difference \( \Delta V \). At a distance \( D \) downstream, the beam is deflected a distance \( x \) given by

\[
x = \left( \frac{q \Delta V}{E} \right) \left( \frac{DL}{2d} \right).
\]

(4)

If a detector is placed behind a slit of width \( \Delta x \) at this position, or if a position sensitive detector having a position resolution of \( \Delta x \) is used, energies between \( E \) and \( E + \Delta E \) will be sampled. Here, where \( \Delta E = E \Delta x / x \). Typical values for the various parameters are given. As seen, unless the slit is very narrow, only modest energy resolution can be achieved with this approach. The other possibility to achieve higher resolution is to make \( x \) larger. But this introduces construction problems and makes it more difficult to use. The advantage of the electrostatic method is that it is extremely simply to construct.

Another method is to use magnetic deflection, as shown in the upper right. In this case the angular deflection and resolution can be related to the positions and energies and we have

\[
\frac{\Delta \theta}{\theta} \sim \frac{\Delta x}{x} = \frac{1}{2} \frac{\Delta E}{E}.
\]

(5)

Thus, the resolution is a factor of two better than for electrostatic deflection. Again, higher resolution is possible simply by using a larger deflection angle. However, this introduces difficulties in aligning the permanent or electro magnets such that the beam hits the slit or detector. Also, large deflection angles can lead to large backgrounds because of reflections of other energy particles. This is particularly troublesome when working with weak intensities associated with large energy losses.

To obtain much better resolution, a Wien filter can be used. This, as shown in the lower left portion, consists of crossed electric and magnetic fields such that the electric and magnetic forces cancel for a particular velocity ion. Thus, downstream a distance \( D \), an ion having velocity \( v_o \) will have zero deflection if the values of the electric and magnetic fields are such that \( v_o = \varepsilon / B \). If the ion has a lower velocity because it has lost energy \( \Delta E \), it will be deflected a distance \( x \) given by

\[
x = D \left( \frac{q \Delta V}{E} \right) \left( \frac{L}{2d} \right) \left( \frac{\Delta E}{E} \right)^{1/2}.
\]

(6)

In this case, typical values that can be achieved for the various parameters show that a good energy resolution is possible. Note also, that by using larger electric and magnetic fields it is possible to achieve higher resolution or the same resolution at much higher energies. The disadvantages of this method are that it requires a good alignment between the electric and magnetic fields. Plus, it requires a fairly strong magnetic field which may require a permanent magnet. This can make it particularly difficult to find the correct alignment parameters for a particular beam.

For low-energy ion beams, time-of-flight techniques can be used in order to measure many energy losses simultaneously. This method is quite simple to set up, only requiring a pulsed beam. It is shown in the lower right portion of Fig. 7. As seen, the energy resolution is given by

\[
\Delta E = \frac{E \Delta t}{t}
\]

(7)

where \( \Delta t \) is the pulse width and \( t \) is the flight time. Typical parameters for a low-energy beam show that good energy resolution can be achieved, subject, of course, to the pulse width that can be achieved. To obtain better energy resolution, either a longer flight path is required or the beam energy must be lower.

One of the disadvantages of the time of flight technique is that narrow pulses tend to mean a low duty cycle. Another is that the main beam also is detected. This limits the beam intensity that can be used and drastically reduces the rates for larger energy losses. A method that we are currently using to overcome this is to combine time of flight with electrostatic deflection. The electrostatic deflection is used to separate the main beam from any charge exchanged components, plus to spread out the energy loss components. By using a large slit width [see Eq. (4)], a wide range of energy losses can be sampled. The time of flight techniques can then be used to

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**Figure 8.** Time of flight spectra for 500 eV H$_2^+$ ions obtained using time of flight plus electrostatic energy loss techniques. \( \Delta E \) gives the approximate energy loss for each spectrum.
improve the energy resolution. In addition, this technique allows us to increase the overall beam current so we can measure larger energy losses.

Figure 8 shows various time of flight spectra measured by this technique using a 500 eV H$_2^+$ beam. The bottom spectrum was taken using no electrostatic deflection. It shows that for energy losses of 100 eV or larger the rates have dropped by three orders of magnitude and are no longer measurable with any degree of accuracy. However, by using electrostatic deflection and repositioning the detector, the top spectrum shows that in the same energy loss range only a small elastically scattered main beam is visible. Plus, there is a drastic reduction in the signal for small energy losses. Hence, by eliminating the main beam and small energy losses, the beam intensity can be increased and reliable data can be obtained at larger energy losses. We are currently trying to improve this technique in order to study molecular fragmentation as a function of the energy deposited in the molecule.

5. Summary

Various experimental techniques which can be used to provide highly differential information about ionization and fragmentation as a function of the energy deposited by leptons and low-energy ions have been presented. These techniques are particularly appropriate for working with very small signal rates or conditions where small signal rates have to be extracted from much larger overall rates. Typical parameters for each were presented and the advantages and disadvantages of each method discussed. A few examples of data they generate were provided in order to demonstrate their effectiveness.

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