Design and Construction of a Velocity Map Imaging Spectrometer

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Approval Sheet

This thesis entitled **"Design and Construction of a Velocity Map Imaging Spectrometer"** by **Brijesh** is approved for the degree of Master of Science from IIT Hyderabad.

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Abstract

The objective of this Master's thesis was to design and construct a velocity-map imaging spectrometer to measure the electron and ion momentum distribution of a molecular gas beam which is ionized by a pulsed laser beam. Velocity map imaging is technique to study the complete kinematics of a chemical reaction. 3D momentum distribution of ions are recorded on a 2D detector. The project involves designing and simulations. Simulations of electron and ion trajectories in an electric field has been made using a software SIMION and the dimensions of the spectrometer and electrodes has been designed with suitable voltage settings of the electrodes so as to focus the ions onto the 2D detector. And based on this simulation, the entire velocity map imaging setup has been constructed. The method works for both electrons and ions.

Contents

Chapter 1

Introduction

The goal is to study about things happening at atomic and molecular level like ions and electrons their energies, momentum distribution and dynamics involved in a chemical reaction.

One way of understanding about the physics of an electron is knocking it out from its atom and finding out where it goes and what energy it has. This process gives information about the behavior of the electron and the force that knocks it out from the atom.

So, being able to detect and extract information about the emitted electron would be interesting both from a fundamental physics point of view and for examining physical phenomena that can ionize atoms, for instance lasers, x-rays, particle radiation and collisions.

In this thesis, these electrons and ions ionized by a pulsed laser which are studied.

An electron can be detected, for example, by letting it hit a surface where it will transfer is energy and knocks out several more electrons by secondary ionization. These electrons will in their turn hit another surface where they will be multiplied again and so on until a measurable current has been created. This way of detecting will give information about, if and when an electron has hit a surface, but it does not give any information on the energy of the electron. For this purpose an electron spectrometer is needed. Electron spectrometers are an important tool in both atomic and nuclear physics as well as in chemistry and biology.

1.1 Electron Spectrometers

There exists a number of different ways to measure electron energies. A few examples are the Hemispherical Analyzer, the Time-Of-Flight Spectrometer and the Velocity Map Imaging Spectrometer.

1.1.1 Hemispherical Analyser

This spectrometer consists of two stainless steel hemispheres positioned concentrically having different potentials on them. The electrons enter and leave through narrow slits. But only the electrons having right kinetic energy can pass the analyzer at a certain potential difference. If the electrons are traveling very fast they will hit the outer hemisphere and if their kinetic energy is very low they will be attracted to the inner hemisphere. Thus only electrons of a very narrow energy region will be able to pass through the whole analyzer to the detector. So we need to find a better technique.

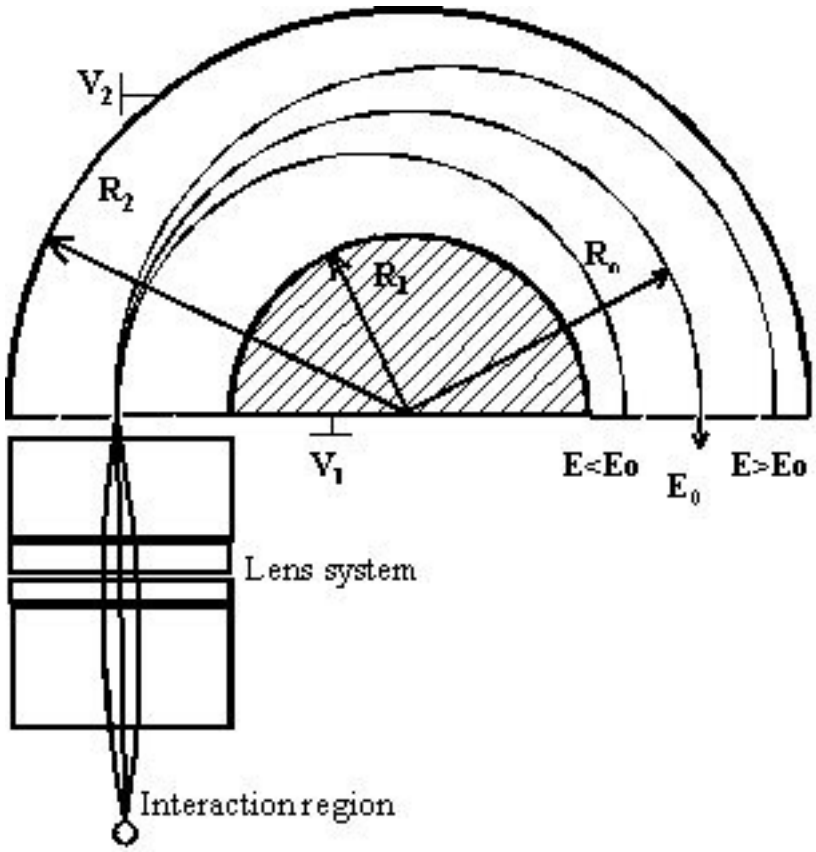


Figure: Schematic of Hemispherical Analyzer

1.1.2 The Time-of-Flight of Spectrometer(TOF)

A TOF spectrometer is another tool for measuring electron energies and their angular distribution. It is a long tube with a detector on one end. On the other end there are two electrodes with different potentials. Sample molecule is ionized in the ionization region and electrons and ions are emitted. According to our measurement whether we want to measure electrons or ions, we will set the polarity of electrodes and electrons or ions will fly toward the detector through a hole in the inner electrode. How long it takes for the the electrons or ions to reach the detector depend on their kinetic energy. By looking at the time of flight with for example an oscilloscope the energy of the electrons can be determined.

One type of TOF spectrometer is The Magnetic Bottle Electron Spectrometer (MBES). In an MBES a magnetic field is created around the TOF tube and the ionization region in the shape of a bottle - hence the name - and the electrons are adiabatically accelerated toward the detector. The MBES is able to collect all electrons with a velocity component towards the detector which give a very high detection efficiency and a much stronger signal compared to the plain TOF.

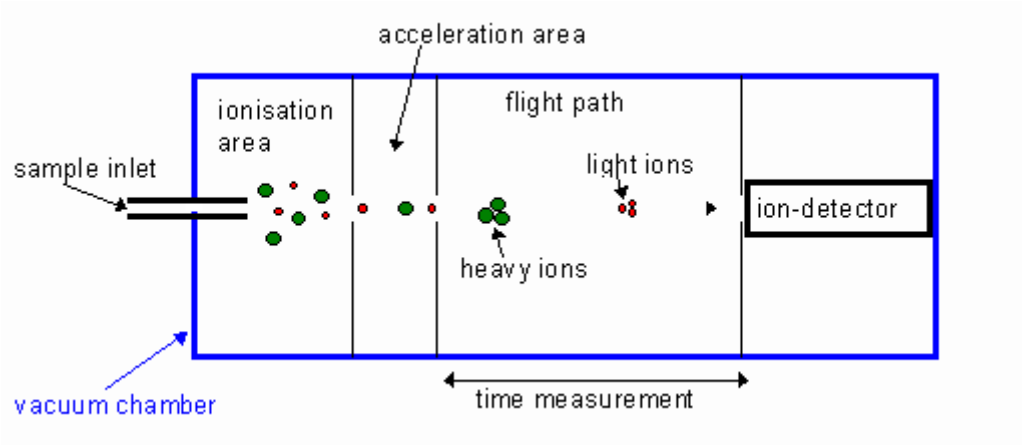


Figure: Schematic of TOF spectrometer

1.1.3 The Velocity Map Imaging Spectrometer(VMIS)

The development of detectors that can register the position where the electron hit, and the use of a position sensitive detector gave rise to a new technique in electron spectrometry called "velocity-map imaging". This technique was first introduced by Eppink and Parker in 1997.

The VMIS consists of a long tube with the ionization region in one end and a detector system in the other. Ionization region is in between two electrodes with high voltage which creates an electric field, similar to the TOF spectrometer. But in this case the voltages on the two electrodes are so high so that the electrons initial kinetic energy can be neglected in the time-of-flight to the detector. Instead it is the position where the electrons hit the detector, a multichannel plate and a position-sensitive detector, which give information on the electron or ions energies and their momentum distribution.

1.2 Motivation for VMI

So, why does one want to build a velocity map imaging spectrometer? The difference between the MBES and the VMIS is that the imaging machine gives an actual image of the electrons velocity distribution. If the electrons are emitted from an s-orbital this angular distribution is equal in all directions but if the ionized level is a p-orbital or higher there will be a higher probability for the electrons to be emitted in certain angles .

Angular information can be extracted in both TOF and hemispherical spectrometry by either moving the spectrometer in different angles or by changing the polarization of the laser.

The advantage of the VMIS compared to the TOF spectrometer and the hemispherical analyzer is that all angular information is gathered in one image and nothing needs to be moved or turned.

The angular information given by imaging spectrometry is interesting both from a general scientific point of view, and for a number of applications. The angular distribution of emitted electrons will for example differ for different elements and compounds, different ionized orbitals, number of photons involved in the transition and which electric or magnetic order of the transition (dipole, quadrupole etc.).
Designing and constructing a velocity map imaging spectrometer is the objective of this thesis.

Chapter 2

Basic Principle of VMIS

The principle of the VMIS is quite simple. Electrons and ions are created in the ionization region between two electrodes in one end of the spectrometer by the intersection of a molecular gas beam and a laser beam. Then they are accelerated towards the detector by an electrostatic field that is created by two electrodes. We can add more electrodes so that ions are focused onto the detector. Position of impact of the ions on the detector is recorded. From this position information on both energy and angular distribution can be extracted.

The image will be a number of concentric circles corresponding to electrons emitted with different energies. The angular distribution of the electrons causes the rings to only be visible in certain angles for which the probability for them to be emitted is large.

2.1 Emission of electrons

The first step in the velocity map imaging process is the ionization of atoms. A laser beam incident from outside the chamber is focused onto a collimated gas jet at the centre of the spectrometer.The laser focus is between the lower electrode, called repeller, and the middle electrode, called extractor. And the atoms in the gas are ionized and electrons are emitted. The direction of the electrons depends on what orbital they are emitted from and the energy of the light involved in the ionization process.

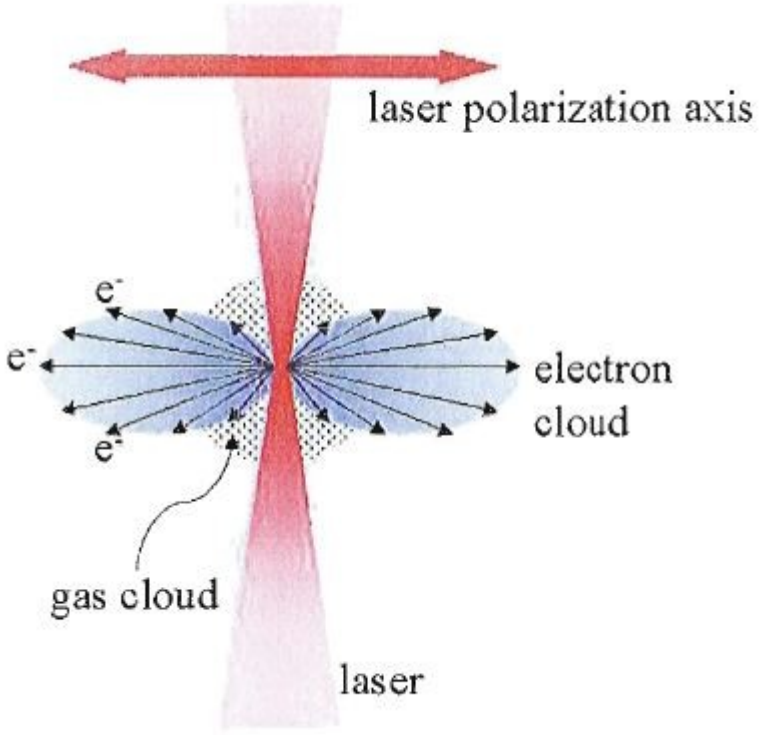


Figure: Ionization of a gas and emission of electrons

2.2 Acceleration of ions towards the detector

In the tube, where the electrons are created, an electrostatic field is introduced. The electrostatic field is created by electrodes in the tube. This electrostatic field will work as an electrostatic lens which will guide the electrons or ions towards the detector. A small hole was drilled in the centre of the repeller electrode to allow ions to pass through when the spectrometer operates in electron imaging mode. This eliminates secondary electron emission which can occur when ions are created in the laser focus and accelerate into the repeller electrode.

An electrostatic lens is a device that assists in the transport of charged particles. For instance, it can guide electrins emitted from a sample to an electron analyzer, analogous to the way an optical lens assists in the transport of light in an optical instrument. Systems of electrostatic lenses can be designed in the same way as optical lenses, so electrostatic lenses easily magnify or converge the electron trajectories. An electrostatic lens can also be used to focus an ion beam.

On the way to the detector the electrons or ions cloud keeps growing radially and when it hits the MCP an image with cm size concentric circles is produced. Electrons with larger kinetic will have a greater radial speed than the ones with lower energy and thus create larger circles.

If the electrons were uniformly emitted in a sphere - as is the case in ionization from an s-orbital - the image would be a complete circle, but because of the angular distributions some angles on the detector will not be hit by an electron at all. The circle will consequently only be visible in other angles.

2.3 Energy and angle resolved spectra

In contrast to time-of-flight spectroscopy, where information of the electron kinetic energy is given by the time it takes for the particle to reach the detector, velocity map imaging gets the velocity information (kinetic energy and angular distribution) from the two-dimensional image.

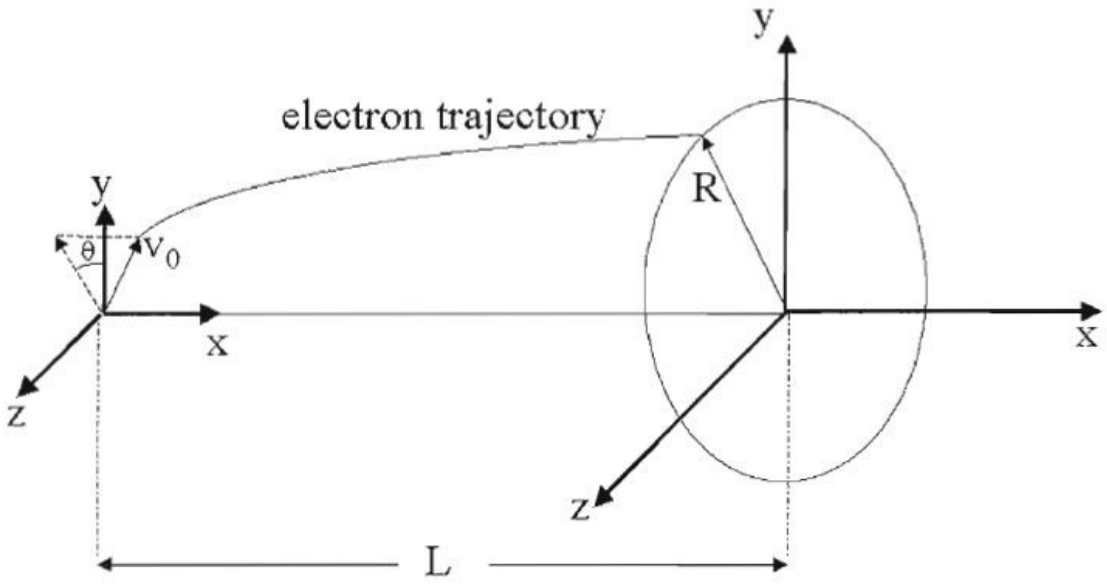


Figure: Schematic view of the geometry of the imaging

The position at which the electron hits the detector is directly proportional to the electrons initial velocity in the yz-direction except for a magnification factor caused by the electric fields component in the yz-direction. This results in a ring on the detector with a certain radius R, related to the expansion speed v_0 of the emitted electron cloud as $R = v_0 t$, where t is the time-of-flight.

In the x-direction t.he electron gets the velocity $v_{E,x} = (2K_E/m)^{1/2}$

from the electrostatic field. Here m is the electron mass. The kinetic energy gained by an electron in such a field is

$$K_E = qV$$

where q is the charge of the electron and V is the potential in which it is accelerated.

2.4 Image Focussing

The spectrometer consists of a long tube with the detection system in one end. On the other end of the tube, there are generally three electrodes, first is the repeller, a flat circular electrode, a couple of millimeters in front of the repeller is the extractor, a similar electrode but with a hole in the middle and a third electrode. We can add more electrode to achieve the focusing condition. A positive high voltage is applied to the repeller and extractor which creates an electric field in the electrode tube if we are working in ion imaging mode. The ions are ejected from an area between the repeller and extractor at the crossing of a gas beam and a laser beam.

Main objectives of the spectrometer is to make all electrons with the same initial velocity, energy and angle, to appear on the same point on the detector regardless of where they were created. By changing the voltage on the extractor, or more specifically changing the ratio between the extractor and repeller voltages, one can focus the electrons so that they all end up on the same point on the focal plane. This is due to the fact that the electrons becomes refracted slightly inwards by the electric field and means that the electrodes serve as electrostatic lenses focusing the image sharply on the detector, compensating for the electrons initial area of creation. The focusing is however only effective when the ionizing happens quite near the center of the ionization region.

SIMION, the simulation software I have used, makes use of potential arrays that define the geometry of electrodes as well as the potentials both on the electrodes and in the empty space between the electrodes. The potential energy surface is much like the surface of a golf course. Since ions react in much the same way to potential energy surfaces as golf balls react to hills and valleys.

However it is not just focusing the image that is important for a good resolution. Electrons of different energies and angles need to be focused onto different points on the detector. Increasing energy will result in circles with increasing radius on the 2D image. In order to get maximum resolution the radius of the image of electrons with the highest energy has to be similar to the radius of the detection area as possible. That is, the trajectoris for the electrons with maximum energy have to be as widely spread as possible without hitting the walls of the TOF tube. This is achieved by changing the voltage settings on the electrodes for different maximum energies. Higher voltages are needed for higher electron energies. When the lens configuration is optimal, the resolution of the spectrometer is limited by the detection system.

2.5 Additional Operation Modes of VMI

The VMI spectrometer can be used for imaging both ions and electrons. It can also be used in a time-of-flight mode for just measuring the energy of electrons and ions.

2.5.1 Ions Imaging Mode

Besides emitted electrons, positive ions are created in the ionization of the gas atoms. Studying these ions is another interesting application of the spectrometer. Ion spectrometry can also be used for making sure that the received signal is really from the input gas and not noise like background ionizing. The principle of imaging ions is very similar to imaging electrons. The only difference is that the negative voltages on the repeller and the lenses needs to be switched to positive values. Furthermore the ion imaging is a bit less sensitive to disturbances.

2.5.2 Time-of-Flight Mode

Changing the spectrometer into operating in time-of-flight mode is very easy. An electric circuit that monitors the electron signal is connected to the MCP and the current cased by the electrons or ions is recorded on an oscilloscope. The reason for using the spectrometer this way is that much lower voltages is required for the MCP when you just want to extract the current and not make an image, thereby decreasing the risk of damaging the equipment while aligning the setup.

Chapter 3

Design And Construction

3.1 Designing the electrodes

In order to the set of electrostatic lenses needed to focus the electrons or ions onto the detector I have used a computer program called SIMION. The simulation of the electrons and ions trajectories for different lens configurations are done. Different electrode dimensions and voltages on the electrodes are being simulated so that our main goal that electrons or ions with same initial velocity, energy and angle are focused on the same point on the detector is achieved. Finally the perfect simulated design is used for assembling the spectrometer. Designing and perfecting the electrode dimensions and voltage settings has been one of the most important parts in making this spectrometer.

3.1.1 Desired Properties for the Spectrometer

In order to get a good measurement the following properties are required:

• Focusing: electrons of the same initial velocity need to be focused onto the same point on the detector. This has to work for wide range of electron energies.

• Spread: the maximum energy electrons have to reach the detector with as large radius as possible.

• Free path: the electrons have to get from the repeller to the detector without smashing into the tube walls or the electrode lenses.

• Linearity: the squared ring radius should vary approximately linearly with the kinetic energy.

To achieve these requirements there are a number of different parameters that can be varied:

• Distance between electrostatic lenses, particularly between the point of electron creation and the extractor.

• The number of electrostatic lenses.

• The diameter of the lens openings.

• Voltage settings on the lenses - in particular the extractor/repeller ratio.

• The length of the tube, that is the distance between repeller and detector.
All this has to be taken into account when designing and constructing the velocity map imaging system.

3.1.2 Simulation

The simulation software SIMION is used for drawing electrodes, setting voltages and calculating trajectories for charged particles in an electrostatic field.

SIMION makes use of potential arrays that define the geometry of electrodes as well as the potentials both on the electrodes and in the empty space between the electrodes. Typically, the we define thepotentials on the electrodes and SIMION solves for the potentials in the space between the electrodes (as well as the fields defined by the gradient of those potentials). The potentials between the electrodes are determined by solving the Laplace equation by finite difference methods. In SIMION, this process is called refining the potential array. Refined potential arrays can then be positioned as potential array instances (3D virtual images) into an ion optics workbench volume. Ions can be flown within the workbench volume, with their trajectories calculated from the fields inside the potential array instances they fly through. This basic approach is the foundation for simulating a wide variety of charged particle optics systems, velocity map imaging spectrometer our case. Trajectories for ions of different energy, mass and charge can be calculated and displayed.

The drift tube and electrodes, detector are drawn up by hand in SIMION and are given specific voltages. They are drawn from side view and by choosing cylindrical symmetry and they are rotated around the x-axis. The entire array is then refined, that is the field due to the drawn electrodes is calculated.

Before starting the simulations, mass, energy, angle, charge and start position need to be set for the particles. After this the ion trajectories can be calculated and drawn up. The potentials on the electrodes can be modified by using fast adjusting tool in SIMION so that we can achieve best voltage settings.

At first I chose a lens configuration with repeller (R), extractor (E) and one extra lenses. And we can add more lens as per our focusing requirements. The design of the extra lenses is such that they each consists of circular electrodes with a hole in the middle. Voltage is applied to the electrode in the middle and the two others are grounded. This is einzel lens configuration just to start with the simulation. Then potential is varied in all the electrodes to achieve the focusing condition. Repeller is the first electrode, then there is extractor in the middle, after that an extra lens. For imaging ions, a positive voltage is applied on repeller and less positive potential on extractor and small positive potential on extra electrode, finally a large negative voltage on the detector.

For a specific lens design and length of tube the voltage setting only depend on the maximum electron energy. It is as mentioned earlier the ratio of repeller and extractor voltages that controls the point of focus and this ratio is nearly constant for the different settings.

3.1.3 Setting the voltages of electrodes

The extractor-repeller voltage ratio is about 0.78, but varies slightly for different maximum energies. The spectrometer is made mainly for a maximum electron energy of 10 eV but the voltages are easily changed for lower maximum energies.

Max electron energy	3 eV	5 eV	10 eV
Repeller	1000 V	1800 V	4000 V
Extractor	780 V	1400 V	3170 V
3rd electrode	100 V	150 V	1000 V
V_E/V_R	0.78	0.778	0.79

In practice the focus is never as good as in the simulations and there are very many factors in a real experiment that affect the outcome of the imaging. I have learned that it is sufficient to use only the repeller and extractor since the extra focusing the two lenses provide is often blurred out for various reasons in the actual image. However the extra lenses could still be useful for fine tuning in a very precise measurement, and they are necessary when the image need to be magnified. When the lenses aren't used, the first one of them (closest to the extractor) is set to 0V, in order to make the last bit of the electrode tube field free. Here are some pictures of VMI chamber designed in SIMION with different kinetic energies. Particles which are flying are proton and are focused onto the detector.

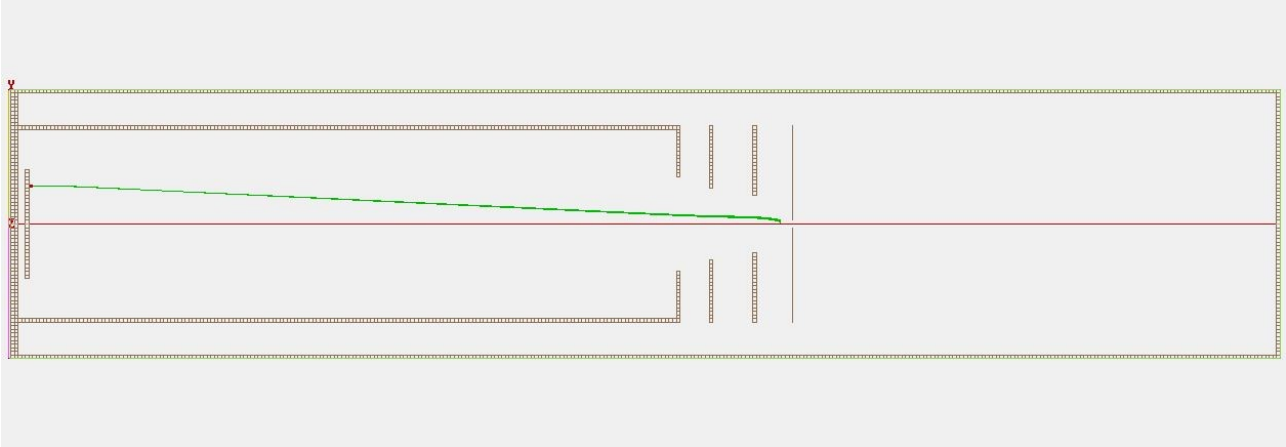


Figure: VMI chamber designed in Simion with maximum kinetic energy of 5 eV

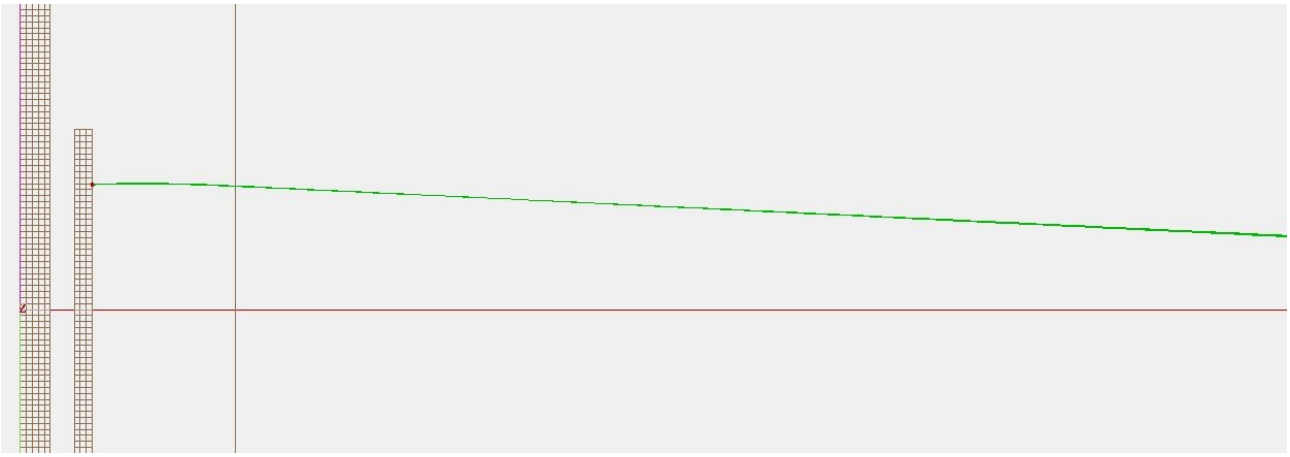


Figure: previous figure in magnified mode

As the maximum kinetic energy is increased to 10 eV, the protons are focusing on different position as compared to 5 eV and we can change the voltage on the electrodes to adjust the focusing position.

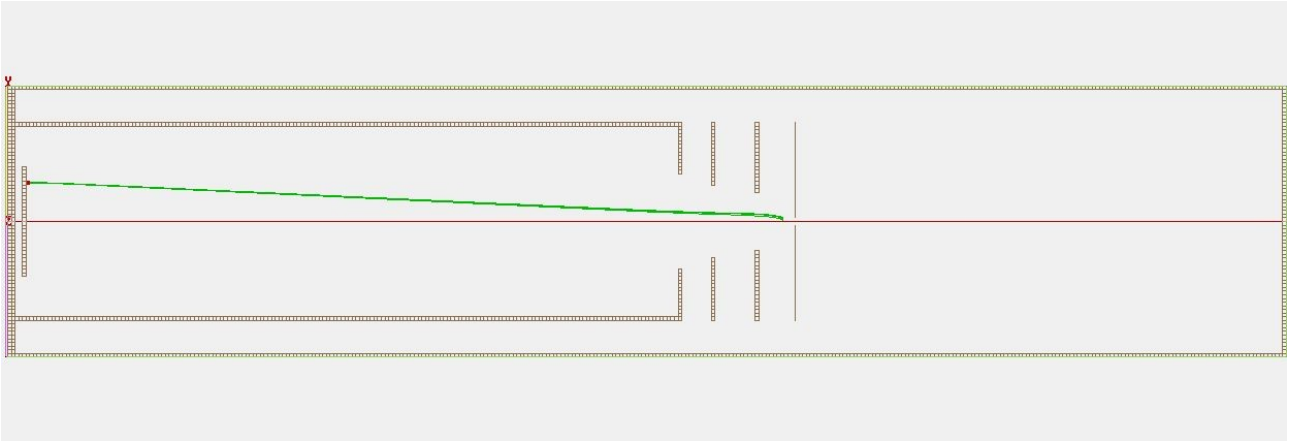


Figure: VMI chamber with maximum kinetic energy of 10 eV

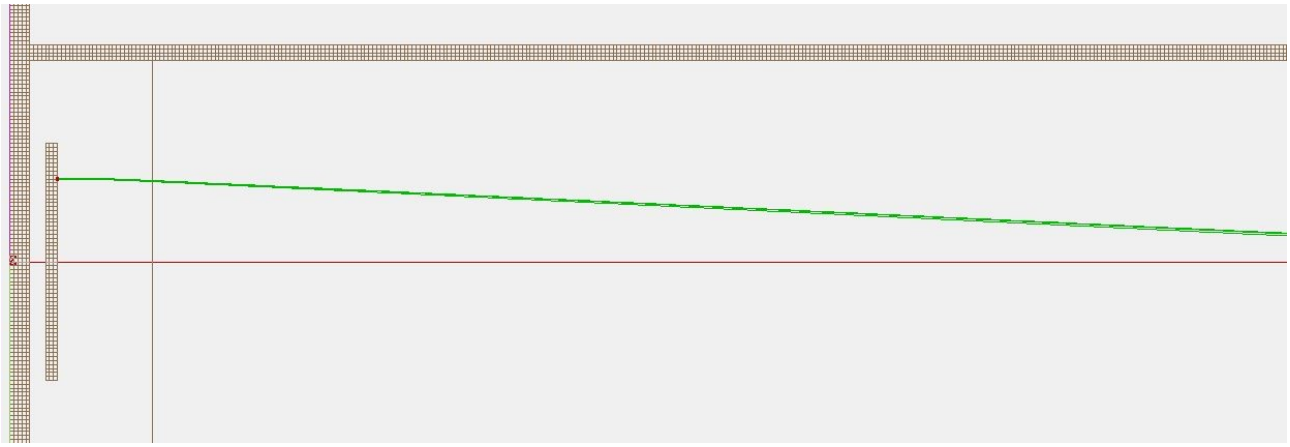


Figure: previous figure in magnified mode

3.2 Vacuum

A long mean free path is needed for the electrons inside the spectrometer. The number of collisions on the way to the detector has to be as low as possible. This means a high vacuum is required in the chamber. Also, the MCP needs high vacuum to not be damaged when high voltage is applied to it. To produce high vacuum the chamber has to be tightly sealed and equipped with a good vacuum pump system.

3.2.1 Vacuum Chamber

The chamber is made of stainless steel components with Conflat flanges. The Conflat flange is a seal fitting with a knife edge that gets pressed into a copper gasket when screwed on to another flange. This ensures that the chamber is tight enough to contain high vacuum.

3.2.2 Vacuum Pump

Under the main chamber a turbomolecular pump is mounted which is backed by a scroll pump. In a turbomolecular pump, a rapidly spinning fan rotor hits gas molecules from the inlet of the pump

towards the exhaust in order to create or maintain a vacuum. The turbopump can not operate if the pressure is too high. So before turning it on, the pressure in the chamber needs to be reduced by the scroll pump. The turbomolecular pump which we are using has a pumping speed of 355 litres/sec by which we achieved a vacuum pressure of 10^{-7} torr.

3.3 Gas system

To start with the experiment, the detection gas used in the spectrometer is a noble gas and then we moved to nitrogen and it needs to be present in the ionization region of the spectrometer during an experiment. At the moment a needle valve is installed where small amounts of gas can be let directly in to the main chamber. But in order to get a good result from the imaging the gas-laser crossing has to be quite small and the amount of gas let in to the chamber needs to be as small as possible.

3.4 Detection System

The detection system consists of a multichannel plate (MCP) behind which a delay line position sensitive anode is kept.

The micro-channel plate detector consists of an array of very fine tubes (micro-channels) made from a highly resistive material with diameters of 10 μm and lengths of 1 mm.
The particles hitting the MCP detector lead to the emission of secondary electrons in the micro-channels, which are accelerated towards the back of the MCP by applying a voltage on the front and on the back of the channel plates. As the accelerated electrons collide with the walls of the micro-channels they produce avalanches of secondary electrons. This process amplifies the original signal by several orders of magnitude.

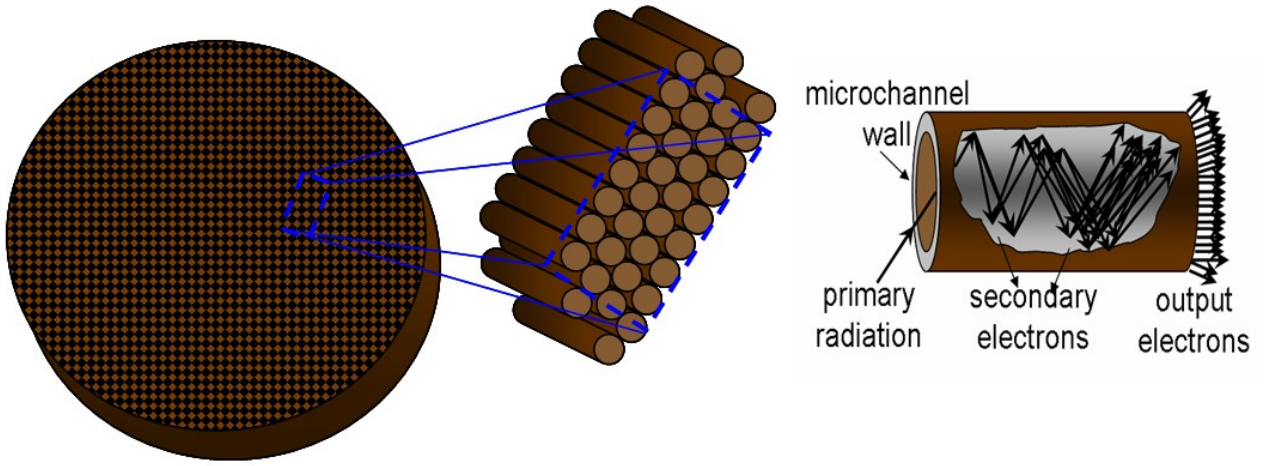


Figure: Schematics of a micro-channel plate detector

The electrons formed through secondary emission in the micro-channels are accelerated after the output from the MCP towards the delay line anode by applying a voltage of about 300 − 500 V on the anode.

The delay line anode consists of two (a quadratic anode) or three (a hexagonal anode) pairs of thin copper wires which are disposed at 90 degrees (60 degrees , respectively) with respect to one another. The advantage of the third pair of wires is a better time and position resolution .

Each one of the two (three, respectively) layers consists of a pair of wires that are tightly wound such that the distance between two adjacent wires is 0.5 mm and that a gap of approximately 1 mm is left between the different layers. By setting the voltage on one of the wires (from here on referred to as "the signal wire") in a pair to a slightly higher voltage (30 − 50 V) than the other one (henceforth addressed to as "the reference wire") one ensures that the electrons are collected on the signal wire only.

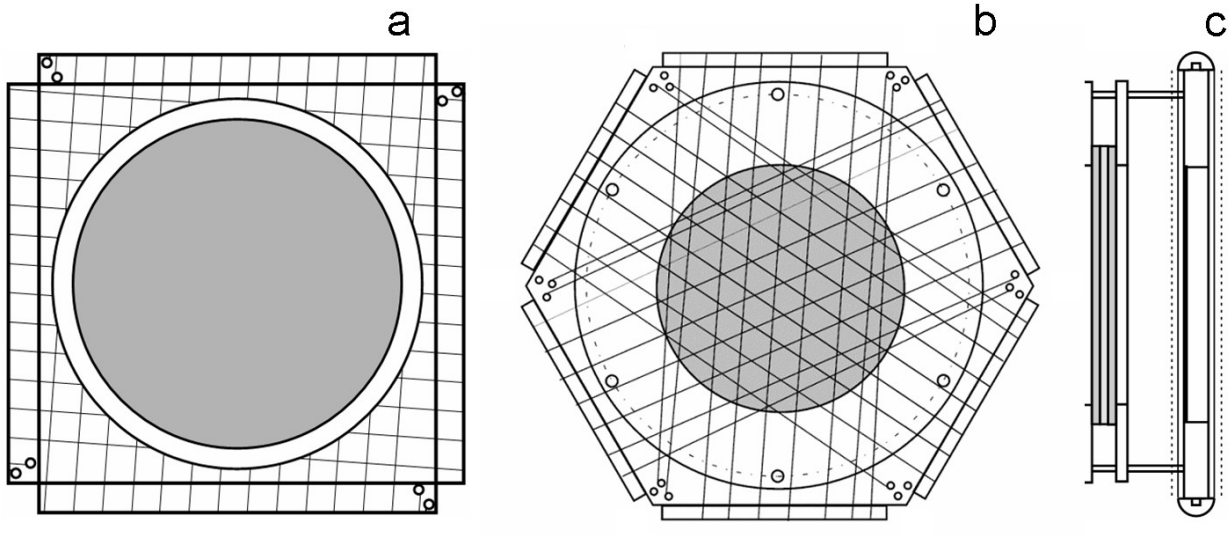


Figure: Schematics of a quadratic (a), hexagonal (b) and side view (c) of a delay line anode

Upon hitting the anode, the electron cloud will induce a signal in the wire, which will propagate towards both ends of the wire where it is picked up for further processing.
From determining the difference in the propagation time of the signal to both of the ends of the wires one can derive the position (X, Y) where the signal was picked up, as described by (for a quadratic delay line anode):

$X = (X_1 - X_2) \times v_\perp$
$Y = (Y_1 - Y_2) \times v_\perp$

where X_1, X_2 , Y_1 , Y_2 represent the measured signal arriving times of the signals to the four ends of the of the delay lines. As a start signal for determining the four arrival times, one can use either the signal from the front or from the back of the MCP stack. Here $v_\perp$ stands for the transverse propagation velocity of the signal on the delay line anode, and is typically of the order of 1 mm/s. The time sums $X_1 + X_2$ and $Y_1 + Y_2$ are constant and irrespective of the hit position and can be used as a time of flight mark if the start signal corresponds to an external trigger (a laser pulse or a synchrotron bunch marker). The time sums also allow for consistency checks of the validity of the recorded position information.

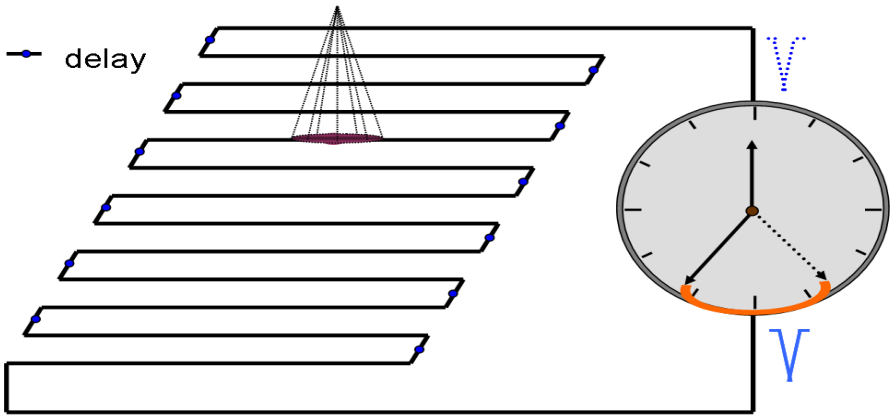


Figure: Schematics of the delay line principle

3.5 Abel Inversion

The Abel transform is an integral transform often used in the analysis of spherically symmetric or axially symmetric functions.

The image from the spectrometer is a two dimensional projection of the three dimensional velocity distribution. If the image contains an axis of symmetry, as is the case in the velocity mapping of electrons, inverse Abel transform can be performed to retrieve the full 3D information.

The Abel transform of a function *f*(*r*) is given by:

$$F(y) = 2\int_y^\infty \frac{f(r)r\,dr}{\sqrt{r^2 - y^2}}$$

where F(y) represent the 2D radial distribution obtained on the detector.

Assuming *f*(*r*) drops to zero more quickly than 1/*r*, the inverse Abel transform is given by

$$f(r) = -\frac{1}{\pi}\int_r^\infty \frac{dF}{dy}\frac{dy}{\sqrt{y^2 - r^2}}$$

f(r) is reconstruct the 3D velocity distribution.

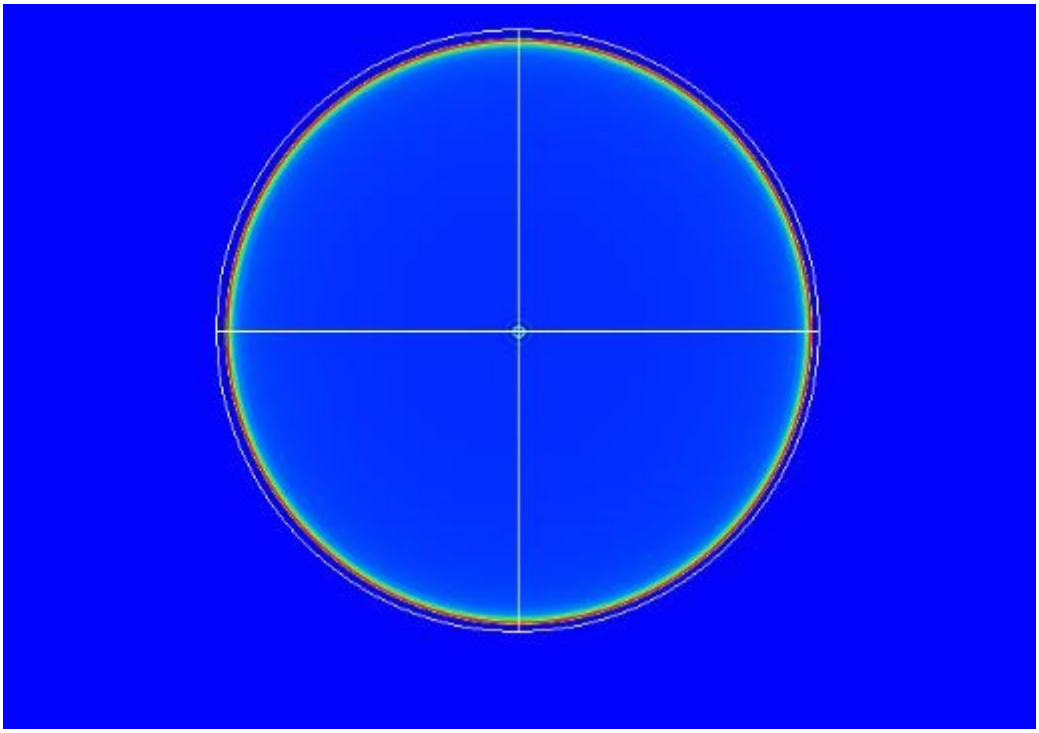


Figure: abel tranform of image recorded in simion at 5 eV

Chapter 4

Experimenting with VMIS

4.1 Experiment

After getting all the ideas about size of the VMI chamber, size of the drift tube, all the electrodes and lens, distances between the electrodes, voltages on the electrodes and lens, the entire VMI set up is assembled at TIFR Hyderabad and the experiment is done.

length of the VMI chamber	700 mm
length of the drift tube	370 mm

All electrodes, lens and drift tube have cylindrical geometry with diameter of 110 mm. Repeller has a hole diameter of 1 mm whereas extractor and lens has hole diameter of 25 mm and 40 mm respectively.

An ultrashort pulsed laser beam is focused onto a molecular gas beam from an effusive gas jet in the ionization region between repeller and extractor. After the ionization, emitted ions are guided towards the detector by electrostatic lens configuration. Images are taken for both perpendicular and paraller polarization of the laser.

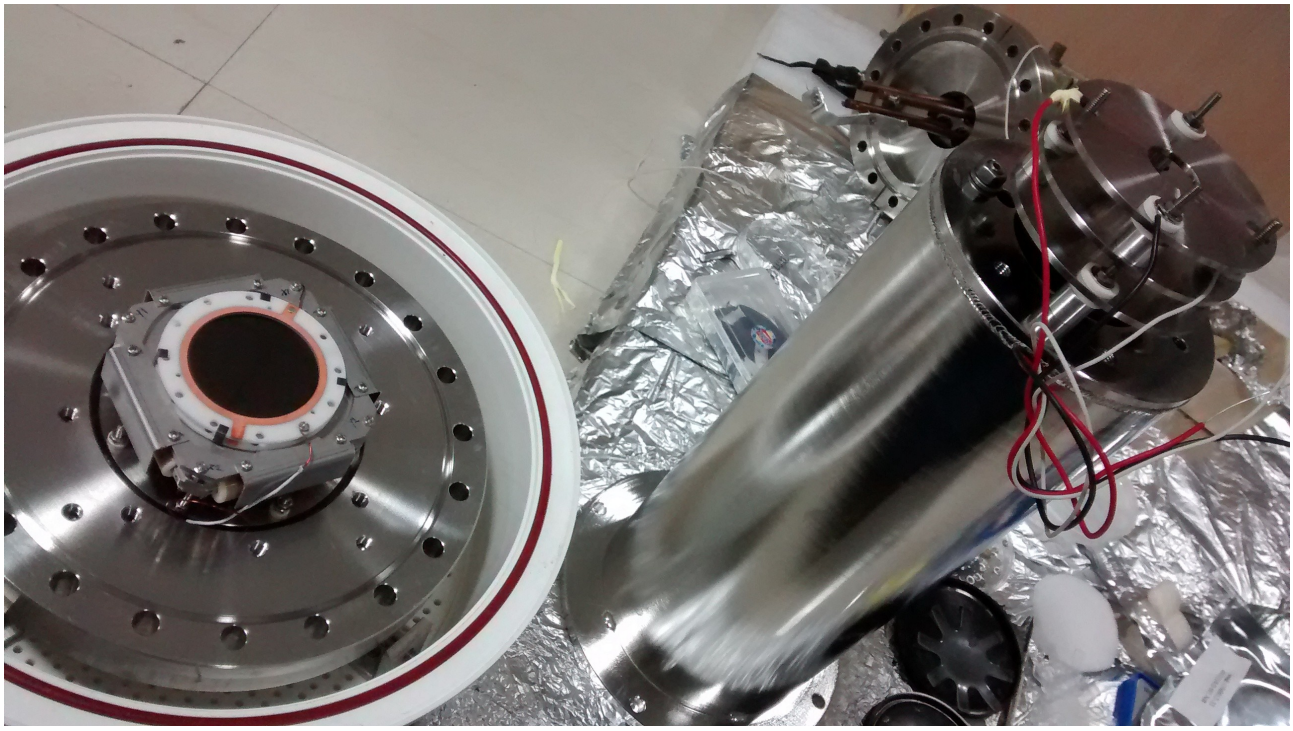


Figure: picture of detector and vmi drift tube with electrodes

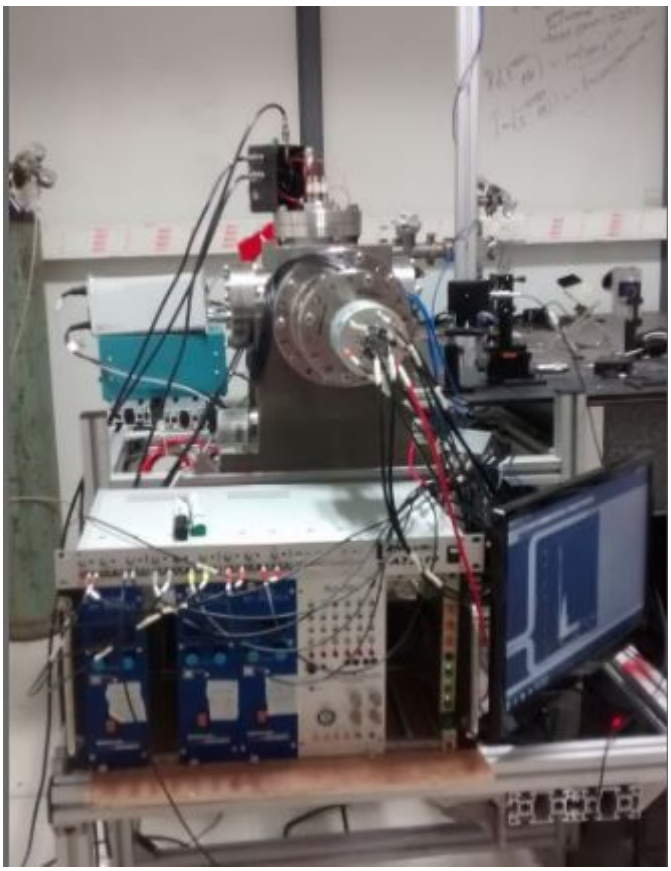
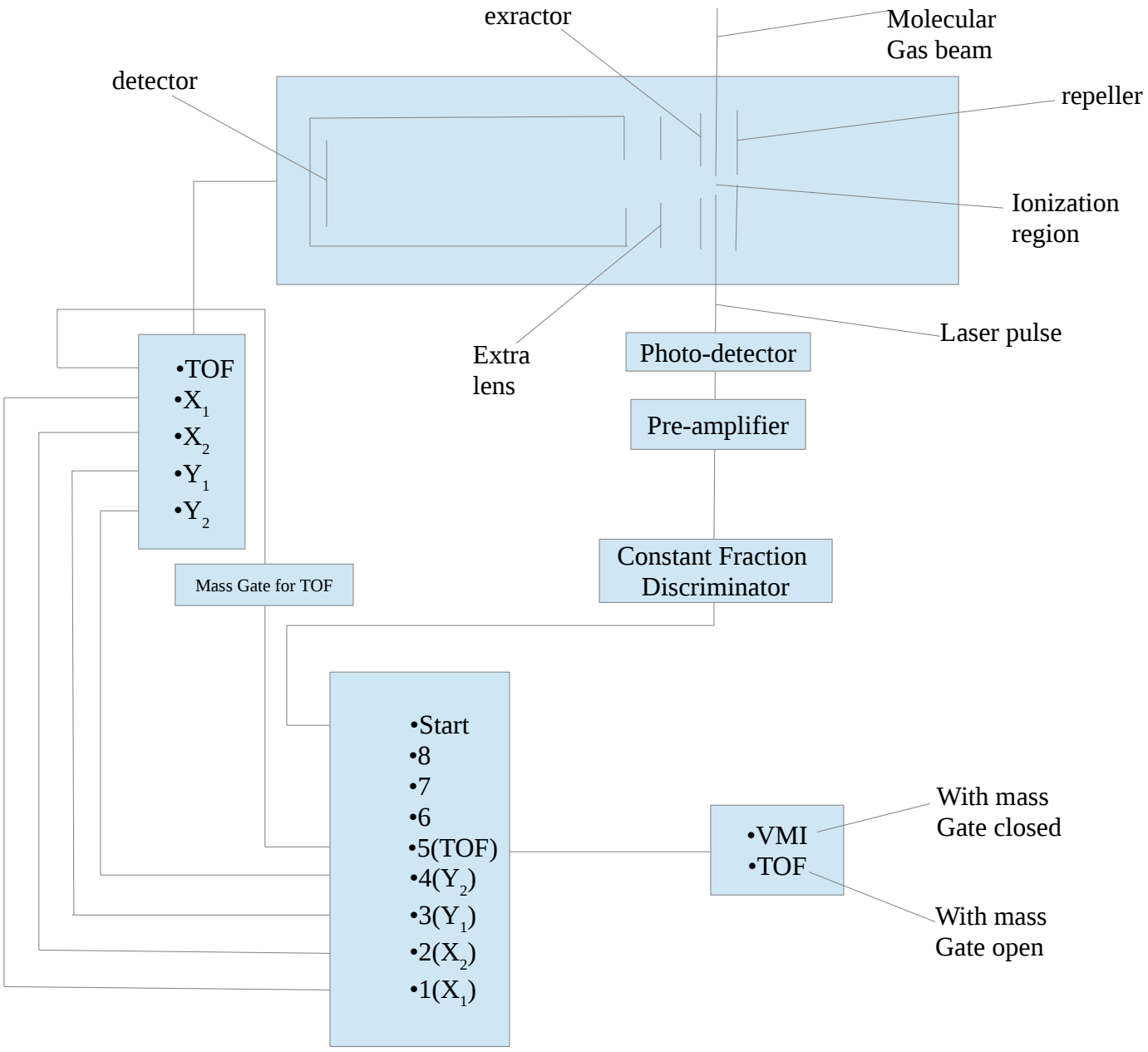


Figure: picture of velocity map imaging experimental set-up

Below is the block diagram of VMI experimental set-up. Initially the experiment is done without mass gate to obtain the time-of-flight for the masses which are present. Then the VMI images for different masses are obtained by using a mass gate. TOF mass gate wiil select the mass to be imaged and rest of all will not be considered. This method makes VMI a faster technique as compared to others.



This is time-of-flight graph for different mass/charge ratio from ionization of Argon gas.

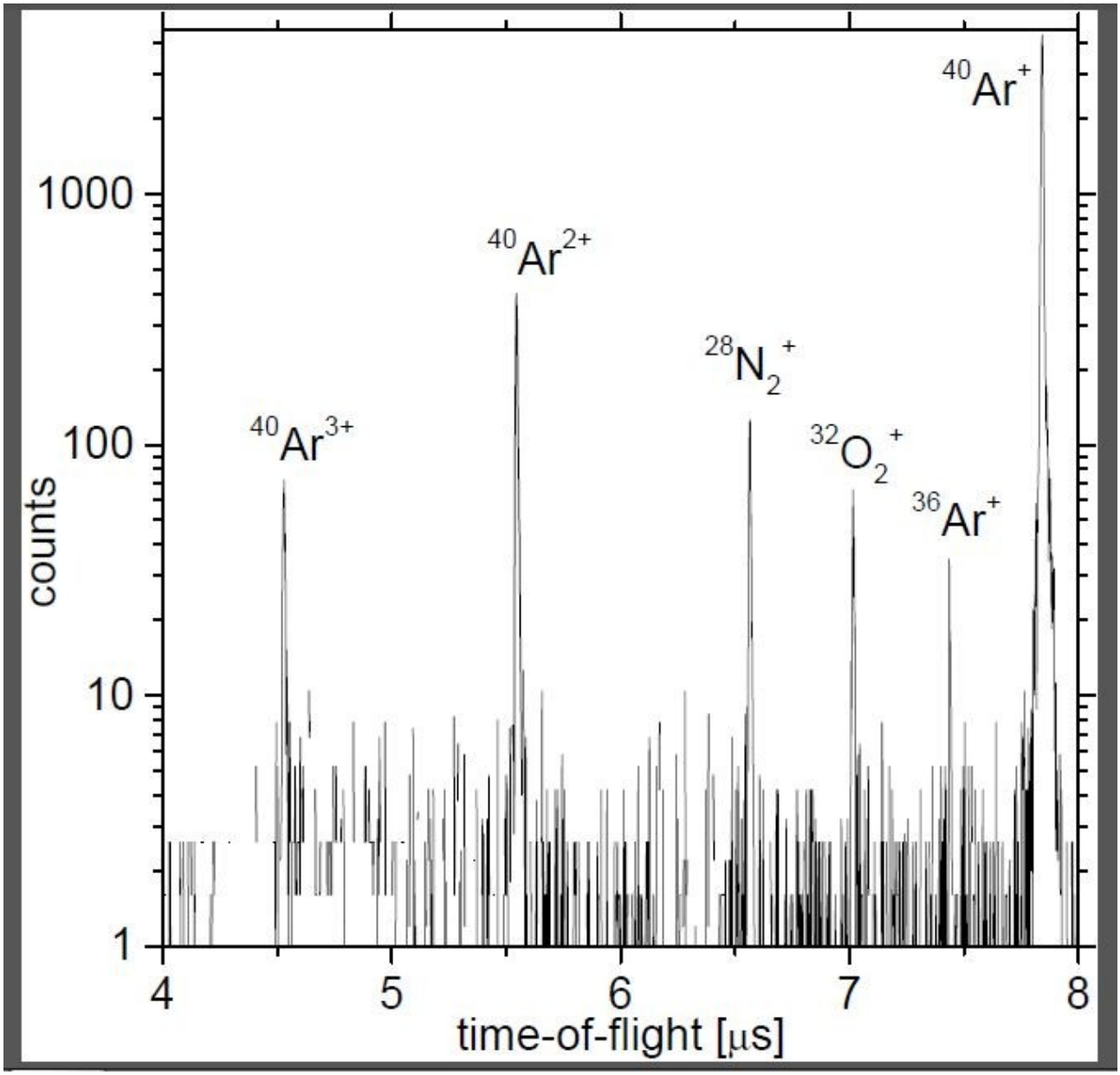


Figure: time of flight

Below are the images obtained at TIFR Hyderabad, an experiment done with N_2 . First picture shows velocity map image for nitrogen ions when laser polarization is kept perpendicular to time-of -flight axis and next pictures is velocity map image when laser polarization is kept parallel to time-of-flight axis.

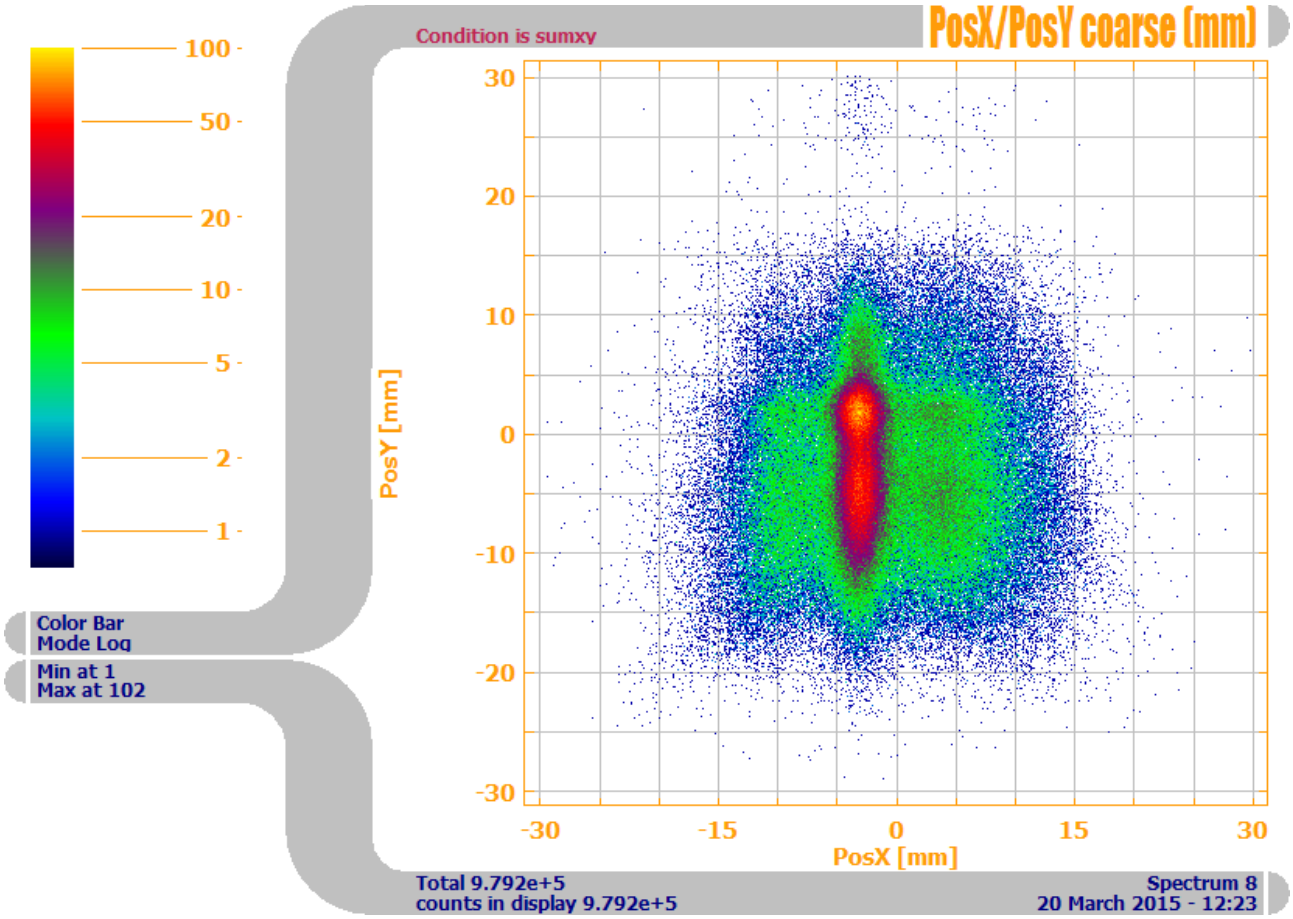


Figure: velocity map image for nitrogen ions with laser polarization perpendicular to TOF axis

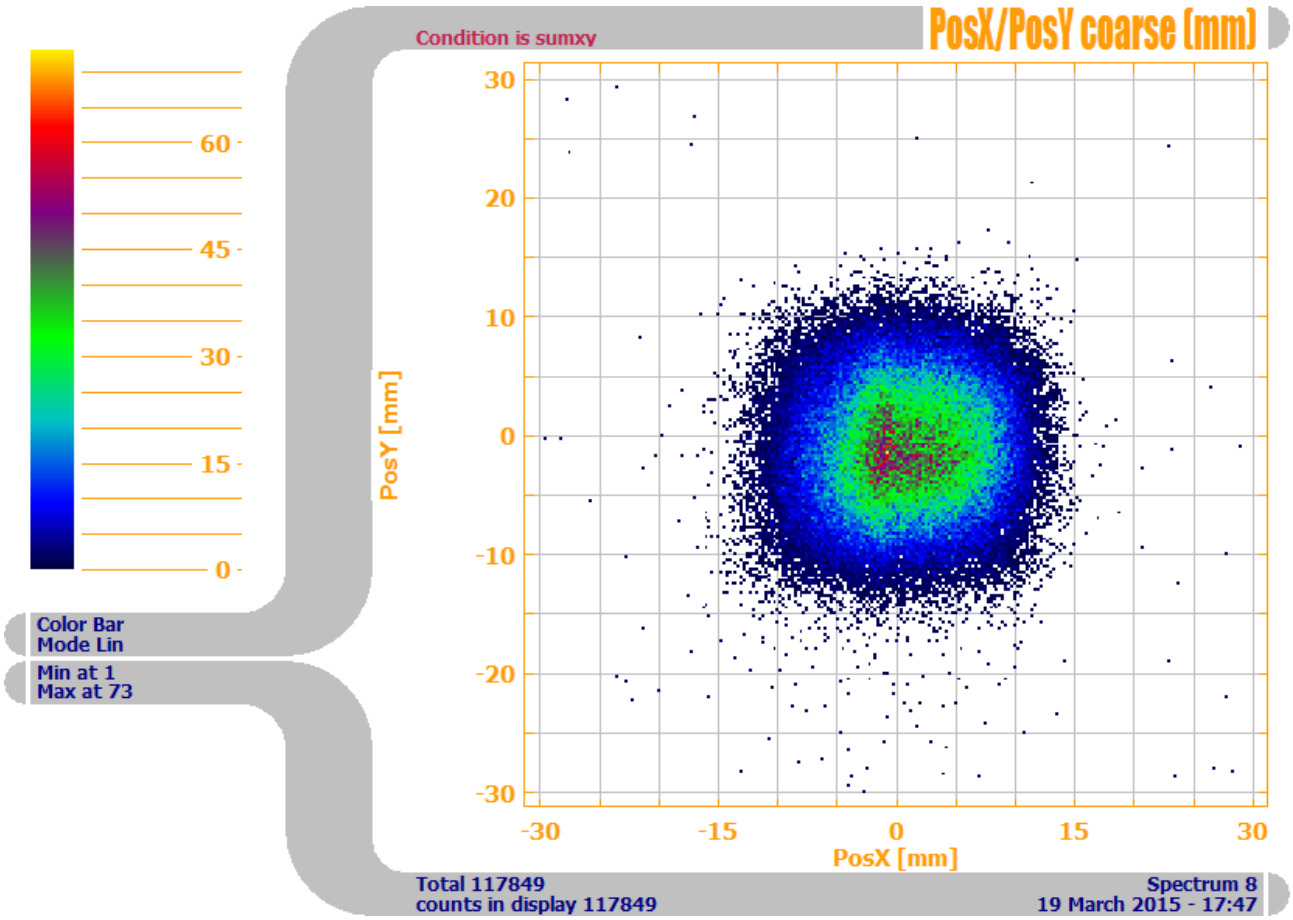


Figure: velocity map image for nitrogen ions with laser polarization parallel to TOF axis

4.2 Conclusion and Future Directions

Velocity map imaging by far is an advance technique to study the dynamics of a chemical reaction. Experiment done with nitrogen gas were compared with the previous experiments and the result matches quite well the previous studies. The velocity map imaging spectrometer is now calibrated and in future will be used to study the ultrafast dynamics induced by coherent x-rays in small and complex molecules.

A similar set-up was developed to perform experiment with Elettra Synchroton Source. There my research group studied the dynamics of helium clustur doped with different organic compounds like CH_2F_2, CHF_3, CH_4, CF_4, CH_3OH. The results will be analysed in near future and the further similar experiment are planned to perform time resolved measurement to go in further details of the dynamics.

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