The Observation of Isochronous Behaviour in a Low Energy Electrostatic Storage Ring

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The Observation of Isochronous Behaviour in a Low Energy
Electrostatic Storage Ring

By

Theresa Spanjers

A Thesis
Submitted to the Faculty of Graduate Studies
through the Department of Physics
in Partial Fulfillment of the Requirements for
the Degree of Master of Science at the
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The Observation of Isochronous Behaviour in a Low Energy Electrostatic Storage Ring

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DECLARATION OF CO-AUTHORSHIP

I hereby declare that this thesis incorporates material that is result of joint research, as follows:

The work was completed using an existing experimental system, the details of which are presented in Chapter 2. The system was designed by Dr. Tim Reddish and Dr. Peter Hammond, and its implementation was completed by Dr. David Tessier. The addition of an ion source and the computer controlled data acquisition scheme were performed by Michael Sullivan. The material presented in Chapter 3 on transfer matrices and regions of stability for the storage ring were developed and previously published by Dr. Peter Hammond. All the work listed above was completed prior to my start date in the summer of 2010. All analysis and discussion of storage modes within this experimental system is the unique contribution of the author and all data presented to support this work was obtained by the author.

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ABSTRACT

An isochronous behaviour has been observed for electrons within a unique, ultra low energy electrostatic storage ring when operating using asymmetric lens potentials. In this work, isochronous refers to an unchanging temporal width of the stored pulse with increasing storage time. Evidence of changing behaviour from dispersive to isochronous has also been observed when storing singly and doubly charged ions. Isochronous behaviour is important when using the storage ring as a mass spectrometer. These observations have been investigated for the purpose of improving the mass resolution of the storage ring. A complete discussion of how this behaviour was observed and a comparison to similar observations in other electrostatic storage devices are presented in this thesis.
To Tim Reddish,

for his invaluable guidance throughout my studies;

and to Anthony Karloff

for his constant encouragement and support.
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CHAPTER I
INTRODUCTION

The containment of charged particles has been of significant interest for atomic and molecular studies and the development of apparatuses for this purpose has evolved into its own field of study over the past 60 years. The ability to contain and control charged particles has generated a wide variety of technologies such as, x-ray machines, electron beam welding, neon and television tubes and the large hadron collider at CERN. This has driven the design of myriad traps and storage rings with diverse energy ranges from meV to GeV, and physical sizes from handheld to spanning several miles [1].

There is a continuing need for systems that can provide acceptable control of the properties of the contained particles. This may include limiting or improving particle energy, energy resolution, target density, current density and mass resolution. Consider the optical analogue in which different technological and experimental applications require visible, ultraviolet or infrared light depending on their desired function. In the field of particle physics, this has lead to the development of large scale rings that use predominately magnetic fields to store particles at relativistic energies. While in atomic and molecular physics, the need to store more massive particles such as clusters and biomolecules has lead to the development of electrostatic storage rings and ion beam traps. These electrostatic storage devices are advantageous in that they are usually smaller, function at lower energies (<200 keV) and as a result are easier and less expensive to operate [1].

All experimental work for this thesis was done using a passive electrostatic storage ring designed to store low energy charged particles of 1-150 eV [2]. In this chapter an introduction to electrostatic storage devices, charged particle storage modes and other comparable experimental set-ups is presented to provide the necessary background information that places the work of this thesis in context. In particular, the importance of characterizing charged particle storage and accessing isochronous storage modes is presented.
1.1 Electrostatic Storage Devices

Electrostatic storage devices (ESDs) rely solely on electric fields to trap (hold stationary) or confine charged particles, such as electrons and ions, within a vacuum. These devices have been used over the past 60 years for a variety of atomic and molecular studies and exist in a large range of geometries, sizes and designs. They are well known to have an upper limit on the energy of the particles being stored and therefore the bulk of electrostatic storage devices operate in a low energy regime of 1-1000 keV [1].

The simplest form of an ESD is a static trap that confines the charged particles in a localized space (i.e. localized Paul trap, shown in Figure 1.1). This trap consists of an arrangement of electrodes (usually hyperbolic), each carrying an electric potential designed to generate electric fields equal in magnitude and opposite in direction that focuses and confines ions into a small region of space [3]. Once confined, the charged particles can then be perturbed by any number of beams (electron, ion or laser) to characterize the properties of the trapped particles and their interactions [1].

Charged particle storage is also achieved by continually moving particles in a linear or orbital path. The same principles of static trapping, where electric fields exert forces on charged particles, have been extended to dynamic traps. For electrostatic systems, these can largely be grouped into two categories: ion beam traps and storage rings [1].

![Figure 1.1](image_url)

*Figure 1.1:* A schematic diagram of the hyperbolic electrodes used in the localized ion trap developed by Wolfgang Paul. The electric potential applied to each electrode (equal in magnitude and opposite in direction) exerts a force on the charged particles confining them in a localized space [3].
1.1.1 Electrostatic Ion Beam Traps

In an electrostatic ion beam trap or EIBT, charged particles oscillate along a fixed path much like photons between two mirrors. This path is usually surrounded by cylindrical electrodes (often electrostatic lenses) with hyperbolic electrodes at either end. Charged particles are repelled by the opposing electric field at one end of the path, accelerating along the axis of the trap until reaching the opposite electrode. Due to the electric field generated by the electrodes at the other end of the track, the particles then slow, stop and begin accelerating back in the direction they came from. Lenses along the linear path focus the charged particles along the optical axis of the trap. One such system has been developed at the Weizmann Institute of Science to dynamically store ions (see Figure 1.2) [4]. Significant work has been done to characterize the behaviour of the stored particles in this system. The details of this characterization are presented in Sections 1.1.3-1.3.3, and form the basis for how storage is characterized in this thesis.

![Figure 1.2: A schematic diagram of the linear electrostatic ion trap (40.7 cm in length) developed at the Weizmann Institute of Science. Electric potentials ($V_1$, $V_2$, $V_3$, $V_4$ and $V_z$) are applied symmetrically to the electrodes. The entrance electrodes are “turned off” as ions are injected into the trap with energy of $E_k$, and then rapidly turned back on. Ions then oscillate between the two sets of electrodes and are detected as they pass through a ‘pick-up’ ring in the centre of the trap [5].]
1.1.2 Electrostatic Storage Rings

Electrostatic storage rings confine charged particles by using electric fields to guide a particle beam around a defined track formed by electrostatic lenses and bending elements. The sustained orbit of particles creates storage since the particles are in constant motion within the ring. An example of this type of system is the ELectrostatic Ion Storage Ring, Aarhus or ELISA (see Figure 1.3) which was built to store heavy ions. The racetrack geometry of the ring is formed by connecting two bent sections of vacuum pipe with straight sections. Each section has electrostatic steering elements that transport charged particles along the axis of the ring and experimental set-ups have been inserted into the straight sections of the track [6]. The experimental apparatus used in this thesis is also an electrostatic storage ring, though of much smaller size (~0.65 m). Details of its operation will be discussed in Chapter 2 and further information can be found in [2].

![Figure 1.3: A schematic diagram of the electrostatic storage ring, ELISA, equipped with an internal electron target (ETRAP), pulsed laser, ion source and various detectors. Ions with energies up to 22 keV are injected onto a straight section of the ring and bent through two 160° sections to form a closed path 8.3 m long [1].](image)

1.1.3 Comments on Injection Methods and Loss Mechanisms

Successful long term storage of charged particles in dynamic traps and rings depends heavily on effective injection methods and reduction of loss mechanisms. This section highlights some basic information pertaining to these two areas. The details and specific
discussion of the injection methods and loss mechanisms for the experimental apparatus used for this thesis are given in Chapter 2 and Appendix A respectively.

In many ESDs, charged particles are generated by a source outside the ring and then injected into the ring in a pulsed manner. Typically a pulsed injection method is used where the electric fields used store the beam are pulsed off briefly to create a field free region for injection, before being pulsed back on again to allow storage. The external source may produce charged particles in a range of energies, positions and emission angles with respect to the local optical axis of the storage ring. For successful storage these must be ‘matched’ to the phase space requirements of the system. This means that position, angle and energies of particles produced by the source (which may be expressed in phase space), must be able to physically enter the electric field of the storage device with a trajectory that can be stored. This can be optimized by construction of an ideal source for the system or by choosing an injection method which is energy selective [7].

There are also a number of loss mechanisms, both design related and inherent, which inhibit long term storage of charged particles. Loss mechanisms that affect the charged particles’ trajectories such as residual magnetic fields and mechanical misalignment of components, can be minimized through design processes by choosing appropriate materials and electrostatic components and by rigorous mechanical construction. ‘Patch’ or ‘parasitic’ fields on lens elements can be reduced by ensuring clean surfaces through heating and voltage supply instabilities can be reduced by using very stable power supplies [7].

Inherent loss mechanisms present in all electrostatic storage rings, include losses due to radiation, space charge effects, collisions with residual background gas and aberrations in electrostatic lenses. Losses due to the radiation generated by orbiting charged particles are insignificant for low energy storage rings. Space charge effects refer to the repulsive Coulomb forces between charged particles within the ‘pulses’ travelling the ring. These effects increase the energy width of the beam as ‘like’ charges repel each other and ultimately limit the beam diameter. Aberrations in electrostatic lenses are of most
significant concern for bending elements, (as in optical systems when considering spherical mirrors) and therefore affect storage rings more than linear traps. Finally, while collisions with residual background gas can not be entirely avoided, they can be minimized by improving vacuum systems, reducing pressure within the ESD [7].

1.2 Characterizing Storage Modes

The motion of charged particles in ESDs is governed by the electric fields of the lenses and bending components they pass through. When considering the pulsed injection of charged particles, a number of particles will travel through the system in a ‘pulse’ or ‘bunch’. These pulses can be characterized using a variety of related parameters that include: 1) energy width (the range of particle energies within the pulse), 2) temporal width (the range of ‘orbit’ or ‘oscillation’ times that the particles have, or alternatively the time interval it takes for the entire pulse to pass a given point in the storage device), 3) spatial width (the physical size of the pulse, length of the track occupied by the pulse) and 4) current density (the number of particles per unit volume within the pulse). The characteristics of the pulse depend both on the initial injection conditions (dependent on the source, extraction field and injection system) and pulse’s evolution (in time and energy) as it passes through the various components of the storage device. The evolution of a pulse as a function of storage time can in turn be used to characterize storage modes within the storage device.

For the discussion of storage modes in this thesis, a pulse will be described in terms of its ‘temporal peak width’. This refers to the full width at half maximum (FWHM) of the peak produced when detecting the individual charged particles passing a given point in the storage device. By examining the evolution of peak width as a function of storage time, it is possible to gain insight into how the particles are orbiting or oscillating within the storage device [8]. Ideally, the peak width should either remain unchanging (so that pulse characteristics are controlled by careful injection) or improve as the pulse completes multiple orbits of the storage ring such that the ring acts as an energy and trajectory filter [9,10].
In practice, the peak width of a stored pulse changes with storage time in different ways that are characterized as distinct storage modes [8]. If the peak width is unchanging with storage time, then the storage mode is classified as isochronous [9]. Isochronous behaviour has been observed in several systems and these observations, as well as discussion of what causes the behaviour, will be discussed in the next section.

1.3 Observations of Isochronous Behaviour

To put the work of this thesis in context a selection of electrostatic storage devices in which isochronous storage has been observed are presented in this section. These systems are currently being operated and were developed simultaneously with the experimental apparatus used in this work. They include a linear electrostatic trap developed by Zajfman et al. [4], a series of multi-pass time-of-flight (TOF) mass spectrometers developed by Toyoda et al. [11] and a linear multi-pass TOF mass spectrometer developed by Wollnik et al. [12]. Each of these groups proposed a different explanation for the isochronous behaviour they observed and these explanations are re-examined in Chapter 4 Section 4.6 to determine if they can be applied to the observations made in our electrostatic storage ring. Also, the process of characterizing storage used in this thesis, as dispersive or isochronous, has been modelled after the method used by Zajfman which is presented Sections 1.3.2 and 1.3.3.

1.3.1 Electrostatic Linear Ion Trap

A linear electrostatic ion trap (or ion trap resonator) was developed by Zajfman et al. [4] (previously shown in Figure 1.2), where ions oscillate between a pair of electrostatic mirrors similar to photons in an optical resonator. Ions with energy of a few keV are stored by continually oscillating along the central axis of the trap. An ion ‘bunch’ is injected into the trap by grounding the entrance electrodes while the electrodes on the opposite side are maintained at a high potential. The bunch slows as it penetrates the opposing electric field until, changing direction, it is reflected back towards the entrance electrodes. By this time (100-200 ns), the entrance electrodes also carry a high potential.
The trap is maintained with symmetric operating potentials, such that the electrodes on either side of the trap’s centre are kept the same, resulting in symmetric oscillation of the bunch. The trapped ions are detected by a cylindrical pickup electrode in the centre of the trap whose amplified signal is recorded on a digital oscilloscope. Pedersen et al. [8] presented experimental evidence of two distinct storage behaviours observed in their ion trap referred to initially as ‘diffusion’ and ‘synchronization’. Later these behaviours were referred to as a *dispersive mode* and *isochronous mode* respectively [13].

### 1.3.2 Coherent and Non-Coherent Diffusion

In general, dynamics of a trapped bunch are examined by analysing the evolution of its size. Pedersen et al., proposed that longitudinal spreading of an ion bunch in their trap was due to four mechanisms. These include: spread of oscillation times ($\Delta T_i$) due to differing trajectories for each ion, initial energy spread of ions from the source ($\Delta E_o$) causing additional width in oscillation time ($\Delta T_v$), external perturbations due to scattering off residual gas and noise on electrode potentials, and internal perturbations caused by Coulomb repulsion between ions [8].

By considering the influence of differing ion trajectories and the initial energy spread of ions within the bunch and no external and internal perturbations, it can be shown that the bunch width after $n$ oscillations, $W_n$ becomes:

$$W_n = \sqrt{W_o^2 + n^2 \Delta T^2}$$  \hspace{1cm} (1.1)

where $W_o$ represents the initial width of the bunch and $\Delta T$ represents the spreading in orbit time. $\Delta T$ arises from the combined influence of independent parameters $\Delta T_i$ and $\Delta T_v$ according to the relation:

$$\Delta T = \sqrt{\Delta T_i^2 + \Delta T_v^2}$$  \hspace{1cm} (1.2)
This type of diffusion is called *coherent diffusion* and is characterized by a quadratic trend in the bunch width as a function of oscillation number, \( n \). This is the most commonly observed storage behaviour (called *dispersive mode*) in this linear electrostatic trap (see sample data in Figure 1.4), as well as in our electrostatic storage ring, (where \( n \) oscillations can be replaced with \( n \) orbits) [8].

![Sample data set showing the peak width \( W_n \) as a function of orbit number \( n \) of 4.2 keV Ar\(^+\) where the initial width of the bunch was 170 ns and \( V_1 = 5.5 \text{ kV} \). The dots represent measured bunch widths. The solid line shows a fit to the data for coherent diffusion (1.1), where \( W_o = 167 \text{ ns} \) and \( \Delta T = 5.9 \text{ ns} \). The dashed line shows a fit to the data for incoherent diffusion (1.3), where \( W_o = 167 \text{ ns} \) and \( \Delta T = 46.3 \text{ ns} \) [8].](image)

**Figure 1.4:** A sample data set showing the peak width \( W_n \) as a function of orbit number \( n \) of 4.2 keV Ar\(^+\) where the initial width of the bunch was 170 ns and \( V_1 = 5.5 \text{ kV} \). The dots represent measured bunch widths. The solid line shows a fit to the data for coherent diffusion (1.1), where \( W_o = 167 \text{ ns} \) and \( \Delta T = 5.9 \text{ ns} \). The dashed line shows a fit to the data for incoherent diffusion (1.3), where \( W_o = 167 \text{ ns} \) and \( \Delta T = 46.3 \text{ ns} \) [8].

Extending this model to account for external perturbations, Pedersen *et al.* developed an expression for *non-coherent diffusion* of the stored beam (shown as the dotted line in Figure 1.4). This storage mode is characterized by the bunch width changing with oscillation number according to:

\[
W_n = \sqrt{W_o^2 + n\Delta T^2}
\]  

(1.3)

The change in the second term \( n\Delta T^2 \) from \( n^2\Delta T^2 \) arises from accounting for the potential change in oscillation time of each ion in the bunch after each oscillation. Without the added external perturbations, the distribution of oscillation times has a constant mean such that the spread in oscillation time is described by (1.2). The external perturbations,
(due to collisions with residual gas and noise on electrode potentials), influence the oscillation of each ion in the bunch stochastically, (e.g. there is a random chance with each oscillation that each ion may be perturbed). By considering the cumulative effect these perturbations have on the bunch, it can be shown that if the perturbations begin on the first orbit, the net effect of these perturbations are to decrease the width of the distribution of orbit times by a factor of \( n \). Full derivation of the expressions for coherent and non-coherent diffusion can be found in [8]. Finally, it can also be shown that the expressions for bunch width given in (1.1) and (1.3) are the bounds for incorporating purely stochastic perturbations on the bunch, where (1.3) represents stochastic perturbations from the first oscillation and (1.1) represents no stochastic perturbations [8].

\[ 1.3.3 \text{ Self-Bunching Storage Mode} \]

Storage was observed to change from a dispersive mode to an isochronous mode in the linear electrostatic ion trap when using a different set of mirror electrode potentials (see Figures 1.5 and 1.6). The change in the mirror electrode potential may be thought as a change in the slope of a potential well, which in turn affects the dynamics of the interacting particles in the bunch. An investigation was undertaken into the effect the internal perturbations due to Coulomb repulsion could have on the ion bunch. Uniquely, in this system the current density of the ion bunch changes significantly as the bunch moves from the centre of the trap (low density) to the turning points (high density). A series of simulations which modelled the interactions of the ions in the region of the mirror electrodes were conducted and led to the development of criterion for the self-bunching mode, the full details of which may be found at [8,13].

Of most significance from these studies, is the conclusion that a repulsive force between the ions in the bunch is required for the self-bunching mode. For particles oscillating in a potential well such that ratio of time spread \((dT)\) and energy spread \((dE)\) of the bunch is \(dT/ dE > 0\), the fastest particles will have the longest oscillation times and will therefore localize in the back of the bunch while the slowest particles localize in the front. This
Figure 1.5: The observed signal (recorded over a 15µs window) for a short bunch of Ar⁺ at 4.2 keV after three different storage times (listed on each plot). Storage in the dispersive mode is shown in the 3 panels on the left and storage in the self-bunching mode is shown on the right [5].

Figure 1.6: Measured data for a 170 ns wide, 4.2 keV Ar⁺ bunch, showing the self-bunching mode. The solid line (1.1) shows the expected trend for coherent diffusion and the dotted line (1.3) the expected trend for incoherent diffusion. Note that the behaviour cannot be fit by either trend or any intermediate form [8].
means that in the turning points, where the Coulomb force is dominant, the fast particles will have their velocities slowed by the repelling force of the slower ions ahead. Similarly, the slow ions will have their velocities increased as they are pushed away from the fast ions behind. These competing effects continually compress the bunch such that its lateral size remains the same [13].

Through the simulations described in [8], it was determined that there is a minimum density of ions in the bunch for this to occur that depends on the initial energy distribution of the ion source. For example, for the ion source used to perform the measurements shown in Figure 1.4, the energy spread was ~ 1 eV, the bunch width ~ 200 ns and therefore a minimum of ~100 ions was necessary to observe self-bunching. This phenomena is considered similar to the onset of ‘negative mass instability’ that is observed in relativistic storage rings [13]. Recently, the self-bunching mode has been used to operate this system as a high resolution mass spectrometer, results of which are presented in [5]. It should be noted that there is one case where there is no minimum density requirement, when \( \frac{dT}{dE} = 0 \), and that this case exists at the boundary between storage favouring the dispersive mode \( (dT/ dE < 0) \), and storage favouring the self-bunching mode \( (dT/ dE > 0) \) [8]. The next two cases of observed isochronous behaviour also have no known minimum current density requirement and are examples of systems designed specifically to access isochronous storage to improve resolution for time-of-flight mass spectrometry [10,16].

1.3.4 Perfect Focusing Condition

Toyoda et al. have designed several multi-turn time-of-flight (TOF) mass spectrometers all of which rely on isochronous behaviour to conduct high resolution mass spectroscopy. While mass spectrometers in general are not necessarily storage devices, the need to create a compact but sufficiently long time-of-flight path for mass spectrometry has led to the development of multi-pass systems that operate as storage rings. In this section, the specific design requirements known as ‘perfect focusing’ used by Toyoda et al. to
consistently produce isochronous storage will be explored, as well as their application to electrostatic systems [11].

Perfect focusing refers to the idea of constructing a multi-turn system such that the ions return to the point of origin in the system. This means that the absolute value of the position and angle at the detector plane are the same as at injection. This eliminates the typical ion beam divergence (and corresponding mass resolution decrease) with increasing orbit. System geometries that support perfect focusing are determined by applying 9-fold focusing conditions on the transfer matrix of the proposed system. The nine constraints encompass: triple space focusing for both horizontal and vertical directions (angular focusing, energy focusing, and lateral magnification) and triple isochronous, or time, focusing (aperture angle, energy and source) [14].

In general, the constraints have been applied to geometries consisting of combinations of electric sector fields (electrostatic bending elements) such that systems are defined as planar, point or doubly symmetric (see Figure 1.7). It was determined that for planar symmetric systems (as would be most similar to the apparatus used in this thesis) there is no solution for the transfer matrix when all the perfect focusing constraints are applied. However, there are solutions for doubly symmetric systems and point symmetric systems (also referred to as ‘figure 8’ systems) [15]. Toyoda et al. have built several of these systems according to these design constraints (see Figure 1.8). Each design has improved upon the goal of creating a compact high resolution mass spectrometer to be used as part of the Rosetta Mars mission [11].

![Figure 1.7: Conceptualization of (a) point symmetric, (b) plane symmetric and (c) doubly symmetric geometries. The thin curved lines at turning points indicate an electric field sector of unspecified angle and the broad band between each set of turning points indicates potential ion trajectories.](image-url)
Figure 1.8: Images and development history of the multi-turn time-of-flight mass spectrometers designed and constructed by Toyoda and co-workers at Osaka University [11].
1.3.5 Energy-Isochronous Storage

At the same time Zajfman developed his linear electrostatic ion trap, Wollnik et al. developed a similar device for multi-pass TOF mass spectrometry [10]. One device developed was very similar in geometry to Zajfman’s (see Figure 1.9) but with the key difference that the reflecting mirrors were reflectrons, meaning they have additional energy focusing capabilities [10,17]. While the two systems operate similarly in that ions are reflected between two mirrors along a longitudinal axis, Wollnik’s design allows for additional control of the ions motion in the turning points, ensuring the device was energy-isochronous [10,12].

![Figure 1.9](image)

**Figure 1.9:** A schematic diagram of the linear multi-pass time-of-flight mass spectrometer developed by Wollnik et al. consisting of a pulsed ion source, a fast ion detector and two electrostatic mirrors separated by a series of lenses designed to focus the ions onto the optical axis [10].

To use this system for mass spectrometry, it is essential that the ions’ flight time between adjacent cycles characterizes its mass-to-charge ratio and not the ratio between kinetic
energy $K$, and charge $q$. If a reference ion with energy-to-charge ratio $K_0/q_0$ has an identical flight time to that of an arbitrary ion of equal mass but different $K/q$, this holds true and the system is energy-isochronous. Physically, this can be achieved by sending ions of higher energy further than the reference ion. By choosing the repeller field in the ion mirrors such that the more energetic ions penetrate more deeply, the ions’ flight path can be increased to compensate for their faster velocity. This condition has been fulfilled in Wollnik’s system such that not only is this condition true between the ions’ start and detection positions but also for a number of intermediate isochronous points [12]. Another way of expressing this condition is that $dT/dE = 0$ for the linear trap which according to the work done by Pedersen et al.[8] is the special case for which there is no minimum current density required to see self-bunching behaviour.

1.4 Summary

Control and containment of charged particles has been of significant interest for the development of experimental systems and technological devices. Storage rings and traps have been developed in many different sizes for a vast range of energies. One classification of these is electrostatic storage devices, which use applied potentials on electrostatic elements to generate electric fields that direct a charged particle’s motion in a defined path [1]. Electrostatic storage devices have been developed in a number of geometries including linear traps [4, 10], doubly symmetric systems [11] and rings [2,6]. The electrostatic storage ring used in this thesis is an example of this type of system, specifically designed for the storage of low energy charged particles (typically 1-30 eV).

Dynamic charged particle storage can be characterized by many different storage modes, which most broadly are described as dispersive or isochronous [8,10]. Isochronous storage refers to any storage characterized by the charged particle bunch passing the same point in the ESD with the same spatial distribution and the same cycle time [9]. Three examples of how this behaviour was achieved in different systems have been presented as: a self-bunching mode [8,13], perfect focusing [14] and energy-isochronous storage [10]. The application of high resolution mass spectrometry has been the leading
motivation to design electrostatic storage devices for isochronous storage and to access these modes within existing systems [11,12]. This information provides the basis for the 1) method of characterizing storage used in this thesis, 2) the potential explanations examined for the observed isochronous behaviour and 3) the motivation for using this electrostatic storage ring for mass spectrometry.
1.5 References

CHAPTER II: EXPERIMENTAL APPARATUS

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CHAPTER II
EXPERIMENTAL APPARATUS

The work presented in this thesis was performed using an electrostatic storage ring developed initially for storing low energy electrons (\(<\sim 150 \text{ eV}\)) with the intent to produce ‘monoenergetic’ electron beams with a narrow energy width, (details of the development of this system are presented in [1, 2]). The system was designed to be able to store different charged particles by modifying the charged particle source and the polarities of the ring’s electrostatic elements and in 2010 the storage ring was adapted to store ions [3]. Its racetrack geometry, desktop size, passive electrostatic elements and low operating energy make it a unique storage system with the potential to store a variety of charged particles including spin polarised electrons, positrons, atomic and molecular ions [4]. This chapter will give an overview of the storage ring’s geometry, electrostatic components and voltage configuration, electron and ion sources, and its injection and data acquisition methods.

2.1 Electrostatic Storage Ring

2.1.1 Storage Ring Geometry

The storage ring is an entirely electrostatic apparatus, desk-top sized and based on a race-track design, as is shown in Figure 2.1. It is composed of two 180° hemispherical deflector analyzers (HDA 1 and HDA 2) connected by two cylindrical lens stacks. The lens stacks consist of four 3-element electrostatic lenses; one before the entrance and exits of each HDA to accelerate/decelerate and control the charged particles’ trajectory. A pulse of charged particles is stored within the ring by completing multiple orbits through the lens stacks and HDA’s. Consider a pulsed beam of charged particles (sometimes referred to as a ‘pulse’ or ‘bunch’) injected into the cylindrical lens stack on the optical axis. The pulse is transferred by the lenses into the HDA which bends or ‘reflects’ the particles onto the second lens stack which in turn will transfer the particles
into the second HDA and back to the first lens stack such that a closed path of $\sim$0.65 m is formed for the particles to orbit. Depending on each particle’s trajectory in terms of its position from the optical axis of the lens stack and angle with respect to that axis, the particle may or may not continue orbiting the system thus being stored. Conditions for successful storage within the ring will be further discussed in Chapter 3. Potentials applied to the lenses and HDAs direct each particle’s position and trajectory allowing those particles whose path is close to the optical axis of the ring to be stored. It should be noted that all the elements in the storage ring are passive; meaning that there are no active feedback mechanisms within the system [1].

Figure 2.1: (left) Schematic diagram of the storage ring and (right) photo of storage ring with the top of HDA 2 removed. Note that the storage ring is made of gold plated oxygen free copper and is housed in an ultrahigh vacuum chamber lined internally with mu metal. Both these materials reduce the presence of stray magnetic fields inside the ring and the entire system is surrounded by Helmholtz coils.
2.1.2 Electrostatic Lenses

The source and interaction lens stacks are composed of electrostatic lenses that transport the charged particle pulse between the HDAs. These lenses may be made up of several lens elements each which act in an analogous way to optical lenses, (see Chapter 3) focusing and collimating the pulse. However, the elements differ from optical ones since they have no distinct boundary surfaces and therefore have a continually changing refractive index. Generally electrostatic lenses are classified based on the effect they have on a charged particle’s velocity (i.e. accelerating, decelerating or neutral) and the lens’ shape [5]. It should be noted that all the lenses in the storage ring have cylindrical symmetry, and ideally particles should travel near the central axis of the lens. This reduces deflection of the pulse and consequent losses as particles impact apertures in the ring. All are assumed to be free of magnetic fields. Physically, the lens is a series of metallic cylinders (lens elements) that have an aperture drilled in the centre of each (see Figure 2.2).

![Diagram of electrostatic lenses](image)

**Figure 2.2:** (upper) Conceptual diagram showing a charged particle’s energy while passing through a cylindrical 3-element lens along the central axis when voltage is applied. (lower) Schematic diagram of one of the lenses located at the entrance (or exit) of a HDA in the storage ring. Typically, $V_1$ is set to the same potential as the adjacent region, $V_2$ and $V_3$ are set to the same potential and $V_4$ is set to the same potential as the adjacent HDA.
A voltage is applied to each element creating a continuous field gradient of potential differences which directs the trajectory of charged particles. For a three element cylindrical lens, with elements of the same radius and voltages $V_1$, $V_2$, and $V_3$, the energy of the charged particles after it has passed through each lens element is $qV_1$, $qV_2$ and $qV_3$ respectively as was shown previously in Figure 2.2. The gradient of the electric potentials causes the charged particles to bend toward or away from the optical axis, similar to a converging or diverging optical lens (see Figure 2.3). It should be noted that while there is a finite energy change for all particles leaving a lens, inside the lens the particles’ energy is changing non-linearly as it passes through the field gradient and this determines the emerging particles trajectory [5].

![Figure 2.3: Conceptual diagram of a three element electrostatic lens created in SIMION showing the simulated trajectories of electrons passing through a focusing lens from the same starting position, but with differing initial angles. Changes in the applied voltages $V_1$, $V_2$, and $V_3$ and the corresponding electric fields they generate cause changes in trajectories of charged particles as they pass through the lenses.](image)

Calculating the trajectories for a large number of particles passing through a non-linear gradient is complex for any system made of more than two lenses, and therefore this is done through numerical simulation. A general sense of the restrictions on the potentials of adjacent lenses may be gained by examining the Helmholtz-Lagrange relation:

$$
\sqrt{qV_1 r_1 \sin(\beta_1)} = \sqrt{qV_2 r_2 \sin(\beta_2)}
$$

(2.1)

where $q$ represents the charge on the particle passing from the lens carrying applied voltage $V_1$ to a lens carrying applied voltage $V_2$. The position of the charged particle relative to the lens’ central axis is given by $r_1$ and $r_2$ respectively, and the changing
trajectory is described by the corresponding pencil angles $\beta_1$ and $\beta_2$. This is analogous to Abbe’s sine condition in optics and defines the refractive index of the lens [7]. Helmholtz-Lagrange conserves the volume of phase space [6] such that the parameters of the particles in terms of energy, radial distance from the central axis and angle are constant. This constrains a charged particle beam for transport through a series of electrostatic lens elements by relating its initial and final energies (due to acceleration in the lens) to angular spread of the beam [7], and its radial extent.

For the three element lenses used in this storage ring, the overall acceleration ratio of the lens is given by $V_3/V_1$ and is essentially independent from the strength of the lens given by $V_2/V_1$. The focusing properties, tabulated by Harting and Read [8], for 3-element cylindrically symmetric electrostatic lenses were used to determine design voltages.

2.1.3 Hemispherical Deflector Analysers

The two lens stacks are connected at each end by a 180° hemispherical deflector analyser (HDA). The HDA serves two purposes within the storage ring: first, to join the lens stacks into a racetrack shape creating a closed loop that physically limits the path of charged particles and second, to act as an energy filtering element that limits the energy spread of the charged particle beam. Physically, the HDA consists of two different sized hemispherical surfaces, arranged such that the smaller hemisphere is placed concentrically within the larger hemisphere leaving a gap between them. A separate potential is applied to each hemisphere, generating a radial electric field in the gap between them. This is an energy dependent field, passing only particles within a specific energy range. An HDA acts as an energy filtering element by rejecting particles of energies outside the range of the pass energy and the HDA's energy. It does this by dispersing the energy of the beam within the HDA, analogous to a prism in optics [9].

The effective electric potential between the hemispheres is given by $V_o$ and the mean energy of charged particles travelling along the central axis of the HDA, commonly referred to as the pass energy, is given by $qV_o$. Ideally, charged particles with energy
equal to the pass energy will traverse through the centre of the hemisphere exiting it along the optical axis of the lens stack. Particles with energy greater than the pass energy will exit closer to the outer hemisphere and particles with energy less than pass energy will exit closer to the inner hemisphere. This creates an energy dispersed image at the exit of the HDA and by placing an aperture at the exit a finite range of energies for charged particles can be selected [9].

Since the electrostatic properties of a 180° HDA are determined based predominately on the voltages applied to the hemispheres and their geometrical size, the energy dispersing properties can be described by [10]:

\[
\frac{\Delta E_{FWHM}}{E_o} \approx \frac{r_s}{r_o} + 0.25\alpha^2
\]  

(2.2)

where \(E_o\) is the mean energy of particles in the HDA, \(\Delta E_{FWHM}\) is their energy resolution, \(r_s\) is the radius of the entrance and exit apertures, \(r_o\) is the mean radius of the HDA (or central trajectory) and \(\alpha\) is the beam half-angle in radians [9]. This determines the resolution of the HDAs for this storage ring to be \(qV_o \pm 0.03qV_o\), (or 3% of the pass energy).

Figure 2.4: Cross-sectional diagram of an HDA showing a potential trajectory (dotted line) for a charged particle. The central axis or mean radius (thin solid line) of the HDA shows the path for particles with energy \(qV_o\) entering with a trajectory along the optical axis of the proceeding lens. In this storage ring the lens element before the entrance aperture also has an applied potential of \(V_o\) and three hoops positioned concentrically within the hemisphere to correct for edge effects of electric fields at the lens-HDA interface [9].
2.1.4 Trajectories of Stored Particles

Each of the components (lenses and HDAs) composing the racetrack of the storage ring have a variable influence on the trajectories of stored particles within the ring depending on their applied voltages and the properties of the particles themselves. The trajectory of a particle orbiting the storage ring may be calculated by examining how it passes through each element in sequence. As previously mentioned, this is very difficult to calculate for a single orbit, and even more complicated to extend the calculation for many orbits. For this reason a model of the storage ring was created for symmetric lens and HDA parameters in Charged Particle Optics 3D (CPO3D), software capable of using numerical methods to perform direct ray tracing. This was used to simulate the trajectories of a single particle orbiting the storage ring for many orbits (see Figure 2.5) [4].

Figure 2.5: Numerically computed non-paraxial trajectory of an electron undertaking multiple orbits of the storage ring as generated by a CPO3D model [4].

This image provides a good visualization of the movement of a charged particle within the ring under the influence of the various electrostatic components. The next two sections will address how the charged particles are generated for injection into the system and the details of their injection and detection.
2.2 Charged Particle Sources

2.2.1 Injection Method

Both an electron and a positive ion source have been developed for this storage ring. In both cases the source was mounted at the base of HDA 1 as is shown in Figure 2.6. A field free region is created in HDA 1 by pulsing it ‘off’, such that the elements in that HDA have been set to the mean pass energy. While the HDA is ‘off’, charged particles are accelerated from the source through the hemisphere, which at this point is a field free region, onto the first lens stack. Conditions for storage are restored by pulsing the HDA back ‘on’ once the charged particle pulse has completed a partial orbit, enabling the deflecting potentials of the HDA.

Pulsing units designed to quickly turn the HDA 1 potentials ‘off’ and ‘on’ for injection and detection are based on AD811 video operational amplifier chips, which have a high slew rate of 2500 V/µs. The measured voltage pulses for the inner and outer hemisphere

\[ \text{Figure 2.6: A schematic drawing of the storage ring showing the location of the source and the channel electron multipliers (CEMs) used as single particle detectors.} \]
voltages when storing electrons are shown in Figure 2.7 with the response of the filament current power supply. This signal is produced by a similar pulsing unit and electrically floats on the source’s injection energy voltage such that the filament has a mean potential corresponding to the ‘0 eV’ energy and the potential difference between the filament top and HDA 1’s pass energy is equal to the voltage on HDA 1 [2].

![Figure 2.7: Plot of the (scaled) electron source (or filament) potential and the measured voltage pulses at the lower inner and outer hemispherical deflector analyzer for a 12 eV pass energy. The inner and outer hoop potentials are not shown, but have similar relative profiles. These pulses are driven by a TTL pulse (150 ns for the data above) [2].](image)

Lens and deflector potentials within the source are optimized for this energy so that a pulse of charge is transferred to the optical axis of the storage ring’s source lens stack. The pulse sequence (which is discussed in Section 2.3.1) is controlled by a BNC 565-8C digital pulse generator with a measured rise (and fall) time of the hemisphere potentials for 20-80% \(\Delta V\), which for electrons is \(~15-20\) ns. This includes the op-amp response as well as the vacuum voltage feedthroughs and the overall load impedance mismatch. Of more significance is that the hemisphere potentials have returned to stable deflecting potentials within 200-300 ns (an average orbit time) of the injection pulse, so that the injected particles can be stored. There are still additional losses in the first few orbits due to lingering transients that could be reduced by using faster electronics and improved impedance termination [2].
2.2.2 Electron Source

The electron source consists of a custom-built electron gun (see Figure 2.8) [11], mounted outside the storage ring on the back of HDA 1 such that the electron beam is injected onto the optical axis of the system. The electron source and HDA 1 potentials are pulsed, referred to as the filament pulse and hemisphere injection pulse respectively, for a width of ~85-120 ns, which is less than the time required for one orbit of the ring (~250-350 ns). Physically, this translates to a usable electron pulse of ~35-70 ns due to a rise and fall time of the HDA potentials of <~25 ns and the response of the electron gun potentials. Once injected the electrons are accelerated and transported through the first lens stack, HDA 2 and the second lens stack reaching HDA 1 again, which now has potentials applied for storage rather than injection.

![Diagram of electron gun](image)

**Figure 2.8:** A schematic drawing of the electron gun that acts as the electron source for the storage ring. Focusing and accelerating elements of the electron gun are given by $F_1$, $F_2$ and the gun base (GB). Aperture sizes of the elements are given by $\phi$.

2.2.3 Ion Source

The storage ring was adapted to store ions by replacing the electron gun with an ion source and switching the polarities of potentials in the lens stacks, hemispheres and detectors. To compensate for the increased mass of the ions, the timing electronics for injection and detection pulsing were changed from an order of 100 ns to 100 $\mu$s. The
The typical orbit time for ions is 50 µs – 200 µs depending on their energy, mass and charge state. Ion energies in the interaction region are ≤ 150 eV and storage lifetimes of more than 10 ms have been observed (~100 orbits of Ar⁺). Since most ion sources and transport systems operate in the keV energy range, this is a novel feature of this storage ring [3].

The ion source and injection lenses that replaced the custom electron gun are also mounted on the outside of HDA 1 such that the centre of the source region aligns with the axis of the lens stack. Ions are created by electron bombardment of a target gas that emerges from an effusive gas source as is shown in Figure 2.9. Electrons are emitted from a pulsed filament that has been seated within an extraction electrode of Pierce geometry [12]. The ~150 eV electron beam is accelerated and focused by a simple electrostatic lens onto the gas target. Any electrons that do not interact with the gas are collected by a Faraday cup located opposite the filament. The whole source region is differentially pumped with respect to the storage ring vacuum chamber with typical pressures being ~2x10⁻⁵ Torr compared to the chamber pressure of ~1x10⁻⁶ Torr [3].

Figure 2.9: A schematic diagram of the pulsed ion source.
The electron filament is pulsed for ~1-2 μs generating ions with thermal energies through electron impact ionization. It should be noted that while singly charged ions are produced in the greatest abundance, this ion source is also capable of creating 2+ and 3+ charged ions [3]. Ions are extracted over $4\pi$ sr through a 3 mm aperture using a field penetration technique [13]. Extracted ions pass through a short injection lens that focuses, steers and accelerates them into HDA 1 [3].

2.3 Data Acquisition

2.3.1 Detection Methods

In the central part of the ‘target’ lens stack, the charged particles may pass through an effusive gas beam and its scattered products may be measured using detectors adjacent to the ‘target’ region (or through HDA 1). Any charged particles that are not scattered will pass through and are stored as they orbit the storage ring again [1]. Alternatively, if there is no gas in the target region, storage of the beam can be measured by ‘dumping’ the pulse of charged particles through HDA 1 at a varying delay time so that the amount of particles stored per storage time is recorded. While it is possible to detect charged particles at several points in the storage ring, the data presented in this thesis concerns only measurements of storage performed through HDA 1 (i.e. no scattered measurements) and only the details of this data acquisition scheme are presented. Details of other detection methods can be found in [1].

A ‘Single Channel Electron Multiplier’ (CEM) is mounted below HDA 1, such that a hole in the outer hemisphere aligned with the optical axis of the ‘target’ lens stack permits charged particles to pass to the detector. HDA 1 can be pulsed ‘off”, meaning that all the potentials in the hemisphere are changed to the mean pass energy of the hemisphere. This causes the particles exiting the lens stack to see a field free region instead of being bent around the hemisphere to store. When the HDA is pulsed ‘off” after injection, the orbiting charged particles are able to enter the detector and the signal received is directly related to the number of charged particles in the stored pulse. This
means that HDA 1 is pulsed ‘off’ twice; initially an injection pulse allows charged particles to enter the system and then a second, independently controlled pulse allows particles to exit the system through the HDA for detection. The detection pulse only has width of ~ 10-100 µs for ions and ~100 ns for electrons. By sequentially changing the delay in a small enough step size it can be observed how many particles remain after each orbit.

These hemisphere pulses, as well as the filament pulse, are carefully sequenced and controlled using a series of delays as is shown in Figure 2.10. Typically, the filament in the source is pulsed first for a specified width (denoted by $A$ in Figure 2.10), and after a short delay HDA 1 is pulsed ‘off’ (for a width of $B$) the first time to allow for injection into the storage ring. The next pulse is sent again to HDA 1 (for a width of $C$) to allow for detection after a minimum delay time ($D$) equal to the time required for ~¾ of an orbit. This minimum delay time is required because the particles must travel ¾ of an orbit between injection and detection points (from one side of HDA 1 around the ring to the other side of HDA 1). This cycle of pulses with these delays may be repeated depending

![Figure 2.10](image_url)

**Figure 2.10**: Conceptual diagram showing the general TTL pulsing scheme for injection and detection. $A$ represents the width of the pulse sent to the filament once per each cycle, $B$ represents the width of the pulse sent that turns the HDA ‘off’ for injection and $C$ turns the HDA ‘off’ for detection. $D$ represents the delay between the 2 HDA pulses. $E$ shows the shift in delay $D$ between two cycles, and $F$ shows delay $D$ increased by $n$ shifts. It should be noted that each cycle may occur more than once before the next cycle starts, depending on the dwell time set for acquiring data at each storage time. See text for discussion.
on the dwell time set for each detection window, typically 0.1 s for electrons and 1 s for ions. When this is complete, the delay between the HDA 1 injection and detection pulses is shifted by a specified increment (E) and the next cycle of pulses begins. After many cycles, the delay between injection and detection pulses will have been sequentially increased such that the charged particles in the storage ring have been detected as a function of storage time.

While this detection is destructive to the pulse of charged particles, by conducting a series of scans one after another it was observed that the characteristics of the beam do not change significantly between scans. This suggests that this is an accurate method for measuring the beam as it is stored. It is also an improvement over measurement of storage by examining the beam scattered off gas as it reduces losses due to residual gas and eliminates the presence of neutral metastable atoms in the detectors.

2.3.2 Channel Electron Multipliers

The CEM is capable of single particle counting while preserving all the timing information of the stored pulse when it is used with a Time-to-Amplitude Converter (TAC) that begins counting with each injection and stops upon the first detected particle. The multiplier is attached to the base of HDA 1 and aligned with the optical axis of the lens stack, such that when HDA 1 is pulsed off stored electrons enter the CEM cone from the field free region. There is a ~2 kV potential applied across the detector which accelerates the electrons from the cone to the signal detection point. Inside the detector electrons collide with coated surfaces forming secondary electrons, which are successively boosted to create a measurable electrical signal as is shown in Figure 2.11 on the following page [14].

While its name suggests that CEM may only be used to detect electrons, the polarity and configuration of its internal components may be changed to detect ions. Positive ions in the storage ring are accelerated towards the now negatively charged CEM cone impacting
and releasing electrons, which in turn are amplified through the CEM as previously described.

![Conceptual diagram of the CEM showing how a single charged particle collides many times within the detector, creating each time a shower of particles that together create a measurable signal. (a) Shows the CEM configured for detecting stored electrons and (b) shows the CEM configured for detecting stored ions.](image)

**Figure 2.11:** Conceptual diagram of the CEM showing how a single charged particle collides many times within the detector, creating each time a shower of particles that together create a measurable signal. (a) Shows the CEM configured for detecting stored electrons and (b) shows the CEM configured for detecting stored ions.

2.3.3 Computer Controlled Data Acquisition

The data acquisition process is computer controlled using a program developed in LabView by Michael Sullivan [15] that calls the appropriate software for recording and displaying the timing information of the stored particles and triggering the pulsing unit at appropriate time intervals. Data acquisition can be broken into three key events: 1) pulsing, referring to pulses sent to the storage ring to allow particles to enter and exit, 2) detection, referring the detection of single particles and transmission of pertinent signals and 3) display where the signals are processed and presented into a usable form. A flowchart showing the electronics used in each stage of data acquisition is shown in Figure 2.12 on the next page.

For stored electrons, a BNC Model 555 Pulse/Delay Generator sends TTL pulses to the storage ring to enable injection and start timing on a Time to Amplitude Converter (TAC). After a set delay another pulse is sent to the storage ring to enable detection and electrons are detected by the Channel Electron Multiplier (CEM). The CEM amplifies the signal of a single charged particle and then passes this signal to a Phillips Scientific
Figure 2.12: A flowchart showing the computer controlled data acquisition process from TTL pulses sent to the storage ring allowing particles to enter and exit to final display of the detected signal. See text for discussion.

Variable Gain Amplifier Model 777 (VGA), which further amplifies the signal for the TAC. The TAC receives the amplified signal and converts its time into a voltage signal between 0-10 V depending on the calibrated time scale. This voltage is passed to an Ortec TRUMP PCI-2k Multi-Channel Analyser (MCA) data acquisition card which places a count into a specified bin corresponding to the correct time. This data is displayed as a histogram using Maestro-32 software thereby forming a spectrum where each count in the histogram represents a particle that travelled the storage ring with a specified orbit time. For ions, a similar process is followed (also shown in Figure 2.12), however, there is a TAC built into the Ortec Multi-Channel Scaler (MCS) pci data acquisition card used for ion data and the data is displayed using MCS-pci Version 2.11 software.

A typical electron spectrum recorded using this data acquisition method is shown in Figure 2.13, with a filament injection pulse of 90 ns, an injection pulse of 91 ns after a 22 ns delay and a detection pulse of 100 ns that was shifted for a 700ns to 51300 ns delay.
Figure 2.13: Sample data set for stored electrons. (upper) A spectrum showing the number of electrons stored after progressively longer storage times. Each peak corresponds to an additional orbit of the ring. Peaks at later storage times are shown magnified at 20x and 200x. (lower) Logarithmic plot of the same spectrum showing the expected exponential decay due to loss mechanisms in the ring.
The chamber pressure was $3.7 \times 10^{-7}$ Torr and the particle energy† was 18 eV (since HDA 1 = 18 V, HDA 2 = 6 V and the source/interaction regions were 18 V). The peak heights in the spectrum show an approximately exponential decay after the first few orbits. This decay is associated with the natural losses in the pulse due to collisions with residual gas. The peaks in the spectrum are fit with a Gaussian curve to enable further analysis of the stored pulse, and the details of this are presented in Chapter 4.

In addition to collecting spectra for a single set of storage potentials, it was necessary to examine how storage is affected when lens voltages are changed (specifically the middle lens elements, $V_2$). LabView programming coupled with 6221 card was used to shift voltages, and repeat scans of the data acquisition program, saving files between each successive scan. This provided a series of spectra with varied lens voltages that were later processed to show areas of stable storage (discussed further in Chapter 3). These algorithms and programs were also developed by Michael Sullivan [15].

### 2.4 Storage Ring Voltage Configuration

Storage of charged particles within the ring is achieved when the lens and HDA potentials are set such that the pulse completes many successive orbits of the ring. Stable storage occurs when successive orbits of the pulse are sustained over a large range of lens potentials. Stable storage or long-term storage has been observed for both symmetric and asymmetric operating conditions, where symmetry refers to applying the same potentials to the HDAs and lens stacks respectively (i.e. HDA 1 = HDA 2, left lens stack = right lens stack). The ability to break symmetry in the storage ring is a unique feature generally not employed in other systems. This allows for greater flexibility in operation, for example: one part of the ring can be set to improve injection conditions while another part of the ring is set for varying experimental conditions. Figure 2.14 shows a schematic representation of a selection of symmetric and asymmetric operating conditions in the storage ring.

†Particle energy is defined by convention as the energy of the particles in the ‘target’ or ‘interaction’ region of the storage ring, (i.e. $qV_T$).
Figure 2.14: Three sets of operating potentials for the storage ring each with different degrees of symmetry. (a) Case 1 shows the ‘fully symmetric’ operating conditions. The source and target regions have the same potentials (i.e. $V_S = V_T$), as do the HDAs (i.e. $V_{HDA1} = V_{HDA2}$). The middle element of lens $U_1$ also equals that of $U$, hence lens $U_1$ is effectively the time inverse of $U$. (b) Case 2 shows asymmetry in the lens stacks since: $V_S = V_T$, and $V_{HDA1} = V_{HDA2}$; but $V \neq U$ (c) Case 3 shows asymmetry in the HDAs since: $V_S = V_T$, but $V_{HDA1} \neq V_{HDA2}$, and lenses $U$ and $W_1$ have different accelerating ratios.

The lens and hemisphere potentials that control the storage of charged particles are produced by a specific voltage configuration. The central element in each lens stack (referred to as ‘source region’ and ‘interaction region’), have been physically grounded inside the vacuum chamber. The charged particle energy in the interaction region is defined by a high precision voltage supply and all other voltage supplies reference this voltage. This creates a virtual ‘0’ energy such that the kinetic energy of a charged particle in the ring is given by the product of the charge on the particle and the potential of that region. The injection energy of the charged particles from the source must correspond to the mean particle energy in HDA 1 for the particles to be injected into a ‘field free’ region. As these potentials define the energy of the stored pulse, HDA 1 and the source injection energy must have low noise, drift and high stability. These potentials, as well as the potentials of HDA 2, are produced by high precision linear DC supplies (model TTi QL564P) that have meV accuracy and an output noise value of < 350 μV rms. Other lens voltages in the system that are less crucial for storage are produced by Acopian linear, resistor programmable 0-150 V DC supplies which have an output noise value of < 2 mV rms.
2.5 Summary

An overview of the construction and operation of the storage ring used for the work in this thesis has been presented. The racetrack style storage ring is composed of two HDAs separated by two cylindrical lens stacks each containing two 3-element electrostatic lenses separated by the ‘source’ and ‘interaction’ region respectively. All elements are electrostatic and passive, and successful storage is achieved by setting the potentials on all elements such that charged particles are transported through them with near paraxial trajectories. The voltages of these elements may be set so that the particles are transported through a set of symmetric or asymmetric operating potentials. The experimental work in this thesis was conducted using asymmetric potentials, such that the potentials and lens elements for HDA 1 and HDA 2 were different. It was under these conditions that isochronous behaviour was observed. The details of how voltage configurations for stable storage are determined are presented next in Chapter 3.

A simple electron and ion source have been constructed for the storage ring allowing for the storage of positively and negatively charged particles. The charged particles are injected into the storage ring by pulsing ‘off’ HDA 1, creating a field free region and allowing the particles to be accelerated onto the optical axis of the lens stack. HDA 1 is pulsed ‘on’ to allow for storage until a second ‘off’ pulse allows particles to exit the ring through the opposite side of HDA 1 into a CEM. The signal from the CEM is amplified and storage time of each particle is displayed in a histogram, forming a spectrum where each peak represents stored particles that have completed an additional orbit of the ring. These peaks were later fit with a Gaussian function to determine their temporal width, and the trend in the changing peak widths as a function of storage time was used to characterize storage modes. This analysis is presented in Chapter 4.
2.6 References


CHAPTER III: MATRIX FORMALISM & STABILITY OF THE STORAGE RING

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CHAPTER III
MATRIX FORMALISM & STABILITY OF THE STORAGE RING

This chapter briefly outlines how the storage ring has been modelled using transfer matrix formalism and how this model has been used to establish operating conditions of the apparatus that yield stable long term storage. The elements of the transfer matrix are expressed in terms of lens parameters and in terms of the position and angle of the particle with respect to the optical axis of the ring. By combining the transfer matrices of each electrostatic component of the ring, a transfer matrix for the entire system has been derived. Full details of this model are given in [1], and are outlined here to provide context for the discussion of storage modes and regions of stability.

The transfer matrix for an electrostatic storage ring can be used to determine the trajectory of a particle with a specific starting position, predicting its end position for any number of orbits. Restrictions were placed on the total transfer matrix for this storage ring to ensure particles perform multiple orbits and are successfully stored. These restrictions were extended to determine lens parameters for the electrostatic lenses in the ring that will correctly limit the range of angles and trajectories of the particles passing through them. It will be shown that these limitations lead to two sets of lens parameters for which there is stable storage, referred to as regions of stability. Lens parameters not included in these sets, describe configurations of the storage ring which do not support long term storage, and are referred to as regions of instability. Understanding the limitations on lens parameters and how those parameters can be changed to move the system from regions of stability to instability is necessary for the discussion of observed isochronous behaviour presented in Chapter 4.

3.1 Modelling the Storage Ring Using Transfer Matrices

The general conditions for stable storage (i.e. stable multiple orbits), of charged particles within the storage ring have been determined using a transfer matrix formalism. The
formalism provides a rigorous mathematical framework for predicting the storage capabilities of the storage ring for an arbitrary number of orbits, which may be extended further to account for higher order aberrations. The formalism differs from numerical electrostatic optics simulation programs (e.g. SIMION and CPO3D) in that particle transport through the system is derived from an analytical description of electrostatic optics instead of a numerical solution to Laplace’s equations describing particle motion through electric fields. As such, it gives an ideal, inclusive set of possible lens parameters for storage (i.e. storage is always found within these set of lens parameters). These lens configurations are further restricted by physical limitations of the system such as mechanical misalignment and fringe field effects. Currently, numerical simulation of 180° HDAs and modelling of storage rings that operate asymmetrically (discussed in Chapter 2, Section 2.4) need to be further developed if this approach is to be used effectively to model this storage ring for multiple orbits [1].

To determine the transfer matrix for a charged particle passing through the entire system, first transfer matrices were derived for each main electrostatic component in the storage ring. As was described in Chapter 2, the storage ring is ‘race-track’ shaped consisting of two 180° hemispherical deflector analysers and two cylindrical lens stacks. The lens stacks focus the charged particle pulse as it enters and exits the HDAs, which ‘bend’ the particles’ trajectories so that they may complete a closed loop. Each of the cylindrical lens stacks are divided into two parts such that there is a three element lens at the entrance and exit of each HDA (see Figure 3.1). These have been labelled as Lens 1-4 and the two HDAs as HDA 1 and HDA 2. For simplicity, the transfer matrix was determined for the storage ring operating under ‘symmetric’ conditions such that Lens 1=Lens 3, Lens 2 = Lens 4 and HDA 1 = HDA 2 [1].

The transfer matrix for each of these components was determined by considering the transfer of a single charged particle through the element. The particle’s initial and final positions are denoted in terms of \((r, \alpha)\) and \((r', \alpha')\) respectively where ‘\(r\)’ gives the distance of the particle from the optical axis of the element and ‘\(\alpha\)’ indicates its angle.
Figure 3.1: (upper) Schematic diagram of the storage ring with characteristic lens parameters and (lower) parameters for an electrostatic thick lens with focal lengths: $f_1$ and $f_2$; mid-focal lengths: $F_1$ and $F_2$; and position of target and image: $P$ and $Q$, respectively such that $K_1 = P - F_1$; $K_2 = Q - F_2$.

With respect to the optical axis. The transfer matrix then fulfills the following equation:

$$
\begin{pmatrix}
\alpha' \\
r'
\end{pmatrix} =
\begin{pmatrix}
a & b \\
c & d
\end{pmatrix}
\begin{pmatrix}
r \\
\alpha
\end{pmatrix}
$$

(3.1)

When considering beam propagation and focusing through the storage ring, charged particle optics is analogous to light optics. Each of the three-element electrostatic lenses can be treated as their optical analogue: a thick lens with corresponding focal lengths, mid-focal lengths and source/target positions as indicated in Figure 3.1. Using the general thick lens dimensions shown in Figure 3.1, the defining or cardinal lengths of Lens 1 is
given by: $K_1 = P - F_1$ and $K_2 = Q - F_2$ [2], where $P$ is the distance to the source and $Q$ is the distance to the entrance of HDA 1.

Using the normal conventions within charged particle optics, the transfer matrix for Lens 1 is given by [3-5]:

$$m_1 = -\frac{1}{f_2} \begin{pmatrix} K_2 & K_1 K_2 - f_1 f_2 \\ 1 & K_1 \end{pmatrix} \quad (3.2)$$

As discussed in Chapter 2, the HDA does not change the position of the charged particle with respect to the optical axis of the storage ring, but instead ‘reflects’ it around a 180° bend. The first order transfer matrix for the hemisphere (i.e. ignoring chromatic aberrations), is given by:

$$m_h = \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix} \quad (3.3)$$

Lens 2 is mechanically the same as Lens 1 for the symmetric case being examined, but the charged particles pass through it in the opposite direction such that its matrix is given by:

$$m_2 = -\frac{1}{f_1} \begin{pmatrix} K_1 & K_1 K_2 - f_1 f_2 \\ 1 & K_2 \end{pmatrix} \quad (3.4)$$

Combining the transfer matrices of Lens 1 (at the entrance of HDA 1), HDA 1 and Lens 2 (at the exit of HDA 1) the transfer matrix for a half orbit of the storage ring is:

$$M_{st} = m_2 m_h m_1 = \frac{1}{f_1 f_2} \begin{bmatrix} f_1 f_2 - 2K_1 K_2 & 2K_1 (f_1 f_2 - K_1 K_2) \\ -2K_2 & f_1 f_2 - 2K_1 K_2 \end{bmatrix} \quad (3.5)$$

This describes the transfer matrix for the source to target section of the storage ring under symmetric conditions, which in terms of particle energies means the source and target energies are the same. Similarly, the transfer matrices of Lens 3, HDA 2 and Lens 4 can be combined to form $M_{ts}$, to give the transfer matrix for the target to source section of the
storage ring. These two transfer matrices are combined to form the transfer matrix of the entire storage ring (source to source), $M_{ss}$.

$$M_{ss} = M_{st} M_{ts} \quad (3.6)$$

In standard circular accelerator theory [e.g. 6-9], $M_{ss}$ can also be expressed in terms of real quantities $\theta$, the angle of rotation of the phase space ellipse for the trajectory of a single orbit [5] and $L$, the ratio of the major and minor axis of the phase space ellipse.

$$M_{ss} = \begin{pmatrix} \cos \theta & L \sin \theta \\ \sin \theta & \cos \theta \end{pmatrix} \quad (3.7)$$

and it can be shown that the transfer matrix for $N$ orbits is given by [1]:

$$M_{ss}^N = \begin{pmatrix} \cos N\theta & L \sin N\theta \\ \sin N\theta & \cos N\theta \end{pmatrix} \quad (3.8)$$

Furthermore, for the symmetric case, the matrix for a half orbit is expressed as [1]:

$$M_{st} = \begin{pmatrix} \cos(\theta/2) & L \sin(\theta/2) \\ \sin(\theta/2) & \cos(\theta/2) \end{pmatrix} \quad (3.9)$$

which is extended to $H$, half orbits using [1]:

$$M_{st}^H = \begin{pmatrix} \cos H(\theta/2) & L \sin H(\theta/2) \\ \sin H(\theta/2) & \cos H(\theta/2) \end{pmatrix} \quad (3.10)$$
3.2 Stability Conditions for Symmetric Operating Potentials

It is well known that the general condition for stable storage [5, 6, 7, 10, 11] can be defined in terms of the transfer matrix for the entire storage ring, $M_{ss}$, as:

$$\frac{1}{2} |\text{Tr}(M_{ss})| \leq 1 \tag{3.11}$$

Physically, this implies that the overall linear ($M_{ss\ 11}$) and angular magnifications ($M_{ss\ 22}$) are $\leq 1$ which means they do not diverge with multiple orbits and therefore are considered 'stable' storage conditions. As the elements of $M_{ss}$ contain terms dependent on the electrostatic lens parameters, only lenses of certain cardinal lengths can be used to achieve stable storage within the ring. The transfer matrices derived in the previous section were used with the trace condition in (3.11) to determine focusing conditions that lead to regions of stable storage.

Consider a particle in the storage ring with a trajectory that after $H$ half orbits satisfies the trace condition. Using the matrix in (3.10), the trace yields the following limitation on the angle $\theta$ [1]:

$$\cos(H\theta/2) = 1 - 2\sin^2(H\theta/4) = \pm 1 \tag{3.12}$$

This limitation is satisfied when $H\theta/2 = m\pi$, and $m$ is any integer for $0 < m < H$.

Relating the matrix elements expressed in terms of lens parameters $f$ and $K$ in (3.5) to the matrix elements expressed in terms of $L$ and $\theta$ in (3.10) and applying the limitation above yields the general storage ring stability condition [1]:

$$\frac{K_1K_2}{f_1f_2} = \sin^2\left(\frac{m\pi}{2H}\right) \tag{3.13}$$
This limitation has been used to compute the cardinal lengths for lenses which enable stable storage. These storage modes, designated \((H/m)\) modes, describe a trajectory that if paraxial retraces itself every \(H/m\) orbits. The details of these calculations are provided in [1]. For this work it is sufficient to know that while the storage ring is operating under symmetric conditions, two regions of predicted stability exist within a range of cardinal lengths as a result of (3.13). These predicted regions of stability have been measured experimentally and an example of this data is presented in Section 3.4.

### 3.3 Stability Conditions for Asymmetric Storage Potentials

The matrix formalism describing the storage ring has been extended to include asymmetric operating potentials, where symmetry is broken first along a horizontal axis and then a vertical axis as shown in Figure 3.1 (and previously in Chapter 2). In type (a) asymmetry, symmetry is broken along reflection plane A, such that the pass energies of the hemispherical deflector analysers are not equal but the lenses at the entrance and exit of each hemisphere are, (i.e. \(\text{HDA}_1 \neq \text{HDA}_2\), \(\text{Lens } 1 = \text{Lens } 2\), \(\text{Lens } 3 = \text{Lens } 4\)). In type (b) asymmetry, symmetry is broken along reflection plane B such that the pass energies of the hemispheres are equal, the lens potentials in each lens stack are respectively equal, but the potentials of the source and target region are not, (i.e. \(\text{Source} \neq \text{Target}\), \(\text{Lens } 1 = \text{Lens } 4\), \(\text{Lens } 2 = \text{Lens } 3\)). The full details of the matrix formalism and stability conditions for the asymmetric case are presented in [12]. As in the symmetric case these limitations on lens parameters lead to two regions of predicted stability for a given set of cardinal lengths, which have been observed experimentally.

### 3.4 Experimental Measurement of Regions of Stability

Applying general conditions for stable storage to the matrix formalism of the storage ring yielded stability conditions that limit the lens parameters of the electrostatic lenses connecting each HDA. For any physical storage ring the lens \textit{geometry} is fixed and the lens \textit{parameters} \(f_1, f_2, K_1, K_2\) (defined in Figure 3.1) are controlled by applied voltages. The ratios of these lens voltages were initially determined using numerically calculated
design voltages by Harting and Read [13] and then adjusted for successful storage. As described in Chapter 2, for this storage ring the lenses are cylindrical three-element electrostatic lenses set such that the elements adjacent to the HDAs and source and target regions respectively share the same potential as that element (as is shown in Figure 3.2). For example, Lens 1 has one element set to the pass energy of HDA 1, one element set to the same potential as the source region and the middle ‘focus’ element between them is set to a potential $V_2$ which is varied to achieve stable storage. Under symmetric operating conditions all lenses have the same lens potentials and under asymmetric operating conditions one or more lenses may not have the same lens potentials according to asymmetry in the HDA pass energies or source and target regions.

Recall from Chapter 2 Section 2.3.3, that each peak in a spectrum represents the number of charged particles counted for a given orbit of the system and each successive peak
indicates longer storage or more orbits of the ring completed, (see Figure. 3.3 (a)). In both symmetric and asymmetric cases, regions of stable storage or ‘stability’ are determined experimentally by varying the $V_2$ lens elements while performing successive spectra measurements for a given set of HDA pass energies and source/target potentials. This effectively alters the cardinal lengths of the lenses.

These series of spectra are displayed in ‘contour plots’ and ‘area plots’, (Figure 3.3 (b) and (c) respectively) so that regions of stability are visually apparent. Figure 3.3 shows an example of these data representations for a small set of electron spectra. The $V_2$ potentials were varied in pairs where $V_{2,1}$ represents the middle lens elements on the
lenses adjacent to HDA 1 and $V_{2,2}$ represents the middle lens elements on the lenses adjacent to HDA 2. The potentials were incremented with small step sizes (0.5 V and 0.25 V) for ranges of $59 \leq V_{2,1} \leq 62$ and $60 \leq V_{2,2} \leq 61$ respectively. The operating potentials were symmetric since HDA 1, HDA 2, the source region and interaction region all had an applied voltage of 12 V. These measurements were made to probe the storage over small changes in voltage rather than to show the boundaries of the storage region, however, these plots clearly show how the same data is presented differently to give insight into changes in storage.

In the contour plot shown in Figure 3.3 (b), five spectra are shown as ‘stacked’ according to increasing $V_2$ potentials (y-axis) with the timing information of the spectra preserved in the x-axis. Each peak in a given spectrum is represented as a coloured point according to the number of counts it has: red for high and purple for low. Each spectrum is then represented by a horizontal line located on the y-axis according to $V_2$ potential, and the colour across the x-axis changes according to the number of stored particles.

The area plot in Figure 3.3 (c) shows how storage changes when the $V_2$ potentials for the lenses of each HDA are changed separately. An area plot is formed by adding up the number of counts for ‘long term’ storage in a given spectrum by including counts from all peaks in the spectrum except the first 5-10. This yields a single value expressing the number of stored particles for each pair of $V_2$ lens potentials. These values characterizing storage have been plotted for many spectra measured using a range of $V_2$ lens potentials, where the y-axis represents $V_{2,2}$ and the x-axis represents $V_{2,1}$. This means that a contour plot (like the one previously examined) is formed from any series of spectra taken along a line in the area plot. Area plots also display the spectra with higher counts as red and lower or no counts as purple and a clear progression from a region of low storage to high storage is observed.

In Figure 3.4 (shown on the next page), a contour plot and an area plot showing the entire regions of stable storage for a given set of operating voltages are presented. As was predicted from the matrix formalism presented previously in Section 3.2, there are two
broad regions of stability. The contour and area plots show electron storage as $V_{2,1}$ and $V_{2,2}$ increase in 1 V increments from 90 V to 150 V for a voltage configuration of HDA 1 = HDA 2 = 18 V and source and interaction region = 36 V. In the area plot, stable long term storage is indicated by lighter coloured regions separated and surrounded by regions of instability, indicated by dark purple. In the contour plot, the regions of stability are shown as two coloured bands and the white areas are where no stored particles were detected. *Edges of stability* refer to the area between bands of stability and instability.

**Figure 3.4:** (left) An area scan showing two regions of stable storage as $V_{2,1}$ and $V_{2,2}$ are increased from 90-150 V respectively. (right) A contour plot showing 2 coloured bands of stable storage, separated by a region of instability as $V_{2,1}$ and $V_{2,2}$ are increased from 90-150 V respectively. The black lines at 120 V and 123 V indicate an *edge of stability*.

It should be noted that storage is stable over a significant voltage change, for example in the area plot in Figure 3.4 (left) there is significant long term storage when $V_{2,1} = 140$ V and $V_{2,2} = 120$ V. The coloured area surrounding this point shows that strong storage is maintained for a voltage shift of 5 V to 10 V. This means that the storage is stable for a ~5% change in lens potential, which is deemed to be highly stable to any voltage fluctuations in the storage ring. For other lens potential combinations, there may be little stability (such as $V_{2,1} = 120$ V and $V_{2,2} = 95$ V) or the ring may be storing on the *edge of stability*. This means that if the voltage is shifted in one direction, storage in increased and if it is shifted in the other, storage sharply decreases. This is more easily observed in contour plots. In the contour plot shown in Figure 3.4 (right), for the black line
representing $V_{2,1}$ and $V_{2,2} = 120$ V, if both potentials are decreased the system moves further into a stable storage region, storing more counts, but if the potentials are increased by more than $\sim 2\%$ (e.g. when $V_{2,1}$ and $V_{2,2} = 123$V), then the system loses long term stable storage. Careful control of these lens potentials makes it possible to explore the edges of storage stability.

3.5 Summary

The storage ring has been modelled using a transfer matrix formalism which has been used to show that there are two regions of stable long term storage under both symmetric and asymmetric conditions. These areas of storage are expressed in terms of limitations on cardinal lengths for electrostatic lenses and ranges of operating potentials for lens elements. Regions of stability have been observed experimentally by recording successive spectra as the middle lens element potentials ($V_2$ in Lenses 1-4) were varied. This experimental data has been displayed in two ways such that areas of long term storage and edges of stability are visually apparent. Contour plots show long term storage while preserving storage time information and area plots show how storage areas change as the $V_2$ lens elements on Lenses 1 and 4 and Lenses 2 and 3 are varied independently. Experimental control of these lens elements provided a unique opportunity to examine storage at the edges of stability which has revealed isochronous behaviour in the stored pulse. The changes in the temporal properties of the stored pulses that characterize this behaviour will be presented in Chapter 4, as well as a discussion of potential explanations for this behaviour and the benefits of accessing an isochronous mode for mass spectrometry applications.
3.6 References

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CHAPTER IV
INVESTIGATION OF ELECTRON & ION STORAGE MODES

Long term stable storage of charged particles is a highly desirable feature of a storage ring. Even more desirable is stable storage that minimizes losses and spreading of the pulse either in time or energy. By altering the injection timing, lens voltages and pass energies within the storage ring, some control over these parameters has been achieved in this work. The most significant results from probing these parameters are presented in this chapter, including the discovery of isochronous storage behaviour for electrons, changing storage behaviour for ions and the ability to selectively store different charged species of ions. For the purpose of this work, isochronous behaviour refers to an unchanging temporal peak width at FWHM as a function of storage time (or orbit number) in a given spectrum. Identification of this mode of operation is advantageous if the system is to be used as a mass spectrometer (e.g. [1, 2]) or a high energy resolution electron or positron source [3].

Regions of long term storage have been observed for both symmetric and asymmetric operating voltages (e.g. different pass energies applied to the two HDAs). The regions of stability are characterized by a range of lens potentials under which storage is maintained for the same pass energies and injection conditions, as explained in Chapter 3. In this work, regions of storage were determined when the potentials of the middle lens element ($V_2$) of each of the four 3-element lenses were varied. The number of stored charged particles was measured for various pairs of $V_2$ potentials and a broad range of storage times were observed. The nature of the orbits within the storage ring was investigated at different lens potentials by examining the charged particle pulse’s peak width as a function of time. The pulses in the storage ring spread out over time due to: 1) the different paths the particles take around the ring, 2) the initial energy spread in the injected pulse, and, in principle, 3) space charge effects within the pulse. The temporal peak width variation for various pairs of lens potentials was shown to have different behaviours depending on voltage configuration, one of which provides evidence of isochronous behaviour of the charged particle pulse.
4.1 Typical Data for Stored Electrons

In normal operation, an approximately 35 ns wide (FWHM) pulse of electrons is injected into the ring through HDA 1, allowed to orbit for a specified length of time and viewed when it is extracted from the ring again through HDA 1. This injection and observation process is repeated while varying the time between injection and extraction pulses. This yields a spectrum of ‘peaks’ that represent the number of electrons extracted per orbit, as discussed in Chapter 2. An example of this type of data is shown below in Figure 4.1, for asymmetric operating conditions in which the pass energies for HDA 1 and HDA 2 were set to 18 eV and 6 eV respectively. For this data set, the injection delays and lens parameters were optimized for the maximum count rate in long term storage while still observing narrow, distinct peaks.

![Figure 4.1](image_url)

**Figure 4.1:** Typical spectrum showing the number of electrons stored as a function of storage time for HDA 1 – HDA 2 asymmetric operating conditions. It should be noted that the storage time on the x-axis is referenced from the detection time of the centre of the first measured peak.

Insight into the changing parameters of the electron pulse can be gained by fitting each peak in the spectrum. The height, area and temporal width at FWHM of the electron pulse for each peak was determined by fitting each peak in the spectrum with a Gaussian peak using a least squares fit. This fit is most accurate when each peak in the spectrum is symmetric and separate from neighbouring peaks. An image of an acceptable fit is shown in Figure 4.2. Trends in changing amplitude, area and width of the peaks are then determined with respect to storage time (or orbit number), as is shown in Figure 4.3 and Figure 4.4. For the spectra in Figure 4.1, the lifetimes for amplitude and area decay in the region of long term storage are 86 μs and 143 μs, respectively. These lifetimes are determined by fitting the exponential decay of the long term stored pulse. It should be
noted that for both peak area and peak height the decay rate changes after some orbits, which in this case is approximately 20-25 orbits.

Figure 4.2: An example of a peak from a typical electron spectrum (black squares), being fit by a Gaussian (red), using a least squares fitting procedure that minimizes the differences between the measured data points and fitted curves for peak height, width, position and flat background.

Figure 4.3: Logarithmic plot of peak area (red) and peak height (blue) as a function of storage time for the electron spectrum shown in Figure 4.1. The black lines show a linear fit which corresponds to the exponential decay of the long term stored pulse.
Figure 4.4: Temporal peak width at FWHM as a function of storage time for the spectrum presented in Figure 4.1. A good fit for coherent diffusion is observed after ~7 μs (i.e. long term storage), with \( W_o = 26.34 \pm 0.05 \) ns and \( \Delta T = 0.72 \pm 0.05 \) ns; see text for discussion.

The exponential decay in peak height and area represent the expected losses in the stored pulse due to various loss mechanisms within the storage ring such as collisions with apertures and residual gas. The temporal peak width however varies quadratically with respect to storage time according to the relation proposed by Pedersen et al [4], previously discussed in Chapter 1, Section 1.3:

\[
W_n = \sqrt{W_o^2 + n^2 \Delta T^2}
\]

(4.1)

where \( W_n \) is the temporal peak width of the \( n^{th} \) orbit, \( W_o \) is the initial peak width of the injected electron pulse and is a property of the source, and \( \Delta T \) is the time spread per orbit. This behaviour is characteristic of a dispersive storage mode, more specifically referred to as coherent diffusion [4]. The weighted least squares fit agrees within the scatter of the points after approximately 7 μs. The scatter in the peak widths is greater than the error in the calculated Gaussian fit and therefore will be used to discuss the accuracy of any curve fitting. The error in the peak widths shows the error in the Gaussian fit of each individual peak and is influenced by the symmetry and number of points in each peak. Therefore, it is expected to increase with storage time as the number of points in the peak decrease, affecting the reliability of the initial Gaussian fitting. The slightly larger error bars in the
first few data points are due to the narrowness of the initial peaks. All the counts in the peaks are spread over only a few channels, which means there are few points in the peak for Gaussian fitting.

The time spread per orbit, $\Delta T$, in (4.1) consists of two components: $\Delta T_{SR}$, the time spread within the storage ring for a charged particle of given energy due to possible trajectories and $\Delta T_s$, the inherent spread in the particles’ energies during injection at $t = 0$. $\Delta T_{SR}$ is a result of a differing $(r, \theta)$ of charged particles within the ring that in turn gives rise to a range of orbit times. Differences in stored particle trajectories depend on the geometrical configuration and operating potentials of the storage ring while $\Delta T_s$, is solely a property of the source. When storing electrons, the energy resolution of the storage ring is better than that of the electron source ($\Delta E_s > \Delta E_{SR}$). This is because the electrons, produced through thermionic emission from a heated tungsten filament, are accelerated directly into the storage ring and therefore have a thermal distribution of initial energies. The energy resolution of the storage ring, however, is determined by the HDAs and is typically ~ 200 meV for current operating voltages. The quadratic relation described by (4.1) assumes $\Delta T_{SR}$ and $\Delta T_s$ to be independent such that the total time spread for each orbiting pulse in the storage ring is given by [4]:

$$\Delta T^2 = \Delta T_{SR}^2 + \Delta T_s^2$$  \hspace{1cm} (4.2)

This also assumes that there is no coupling between these processes from internal mechanisms within the pulse such as Coulomb repulsion, or from external perturbers such as residual gas scattering or electrical noise. This implies that $\Delta T$ remains constant for all orbits as each charged particle moves independently of the others and preserves its initial orbital period.

This is the most consistently observed storage behavior (or mode) for stable storage of electrons within this storage ring. According to Pedersen et al [4], this is characteristic behaviour of charged particles in a pulse that are naturally spreading out from each other as faster particles move ahead of slower particles, where the density of the pulse is
insufficient for Coulomb repulsion to significantly influence its dynamics. This accurately describes the supposed dynamics of pulses within this storage ring which are of low current density. Furthermore, it is particularly accurate for symmetric operating conditions where the pulse is passed through the same operating potentials at each point in the ring and the hemispheres provide the only mechanism for slower particles to ‘catch up’ to faster ones (i.e. particles travel different path lengths in the HDAs according to their energy). In the next section it is shown that this characteristic quadratic behaviour is not limited to the stored electrons, but extends to stored singly and multiply charged ions.

4.2 Storage of Singly and Multiply Charged Ion Species

4.2.1 Storing Singly Charged Ion Species

The most abundant ion charge state produced by the ion source currently used with this storage ring is singly charged. The spectrum of stored ions produced by the storage ring is similar to that produced by stored electrons with only the orbit time and lifetime changing. Figure 4.5 shows the typical spectrum of a singly charged ion stored in the ring, in this case Ar\(^{+}\). Each successive peak represents an additional orbit of the 0.65 m track indicating that Ar\(^{+}\) completed 75 orbits and a total path length of ~50 m with an orbital period of 98.4 μs. The peak height decay is illustrated in both linear (upper) and logarithmic (lower) scales where a low signal-to-background ratio can be easily observed. The peak height of each successive peak decreases near exponentially as ions are lost from the initial injection pulse due to hemisphere pass energies, collisions with apertures, and collisions with residual gas.

This means improving injection will improve the overall number of ions present in a stored pulse and to achieve this improvement there must be good phase space matching in terms of energy (\(K\)), position relative to the optical axis (\(r\)), and injection angle (\(\theta\)) between the source and the ring. It should be noted that while the ion source is differentially pumped, there are additional residual gases in the chamber from the 3 mm aperture connecting the source and the chamber, that are not present when storing
Figure 4.5: A plot of the ion yield of Ar$^+$ as a function of storage time, displayed in both linear (upper) and logarithmic (lower) scales. Ions are detected in CEM (A) that have been sequentially pulsed out through the storage ring’s lower HDA using the data acquisition scheme described in Chapter 2, Section 2.3. The peak amplitudes and areas slowly decay with time due to residual gas scattering, and the peaks steadily broaden as discussed in the text.

The peak height, area and width at FWHM have been plotted as a function of storage time in Figure 4.6 for the data shown in Figure 4.5. The peak width variation clearly shows good agreement with the form of (4.1) with least squares fit values of $W_0 = 4.56 \pm 0.04 \mu s$ and $\Delta T = 0.11 \pm 0.09 \mu s$. It should be noted that like electrons, this dispersive mode is the typically observed storage behaviour for ions. The $\Delta T$ term is still described by the independent terms $\Delta T_{SR}$ and $\Delta T_S$ but unlike storing electrons, the energy resolution of the source is better than that of the storage ring ($\Delta E_{SR} > \Delta E_S$) when storing ions. This is because the field-penetration technique for injection is highly selective in
energy with a resolution of $\sim 10$ meV and the energy resolution of the storage ring remains $\sim 200$ meV for current operating voltages.

![Graph 1](image1.png)

**Figure 4.6:** (lower) Logarithmic plot of the peak height and area as a function of orbit number and (upper) plot of temporal peak width as a function of orbit number fitted for a dispersive storage mode using (4.1) for the Ar$^+$ data presented in Figure 4.5.

4.2.2 Storing Multiply Charged Ion Species

While the most abundant ion species produced by the storage ring’s ion source is singly charged, the pulsed electron beam in the ion source has sufficient energy to produce multiply charged ions. The different charged species (most often a combination of singly and doubly charged ions) have been stored simultaneously in the storage ring. Since the
storage ring is governed by electrostatic lenses, a doubly charged ion will experience twice the acceleration of a singly charged ion of the same atom, while still completing the same trajectory within the HDA and therefore it may be stored within the ring. This indicates that the storage ring could be coupled with any number of ion sources including those that produce highly charged ion species. These species can be separated from other molecules and products that may be present in a spectroscopy application using the well known relation \( M/qe \), where \( M \) is the atomic mass number, \( q \) is the charge on the ion and \( e \) is the charge of an electron.

For example, the ion source currently used is capable of producing \( \text{Ar}^+ \) and \( \text{Ar}^{2+} \) (or more generally \( \text{Ar}^{q+} \)) ions. The field-penetration technique sets an extraction energy of typically 0 meV to \( \sim 10 \) meV, giving the ions injected into the storage ring approximately the same initial energy. This energy range is suitable for ‘thermal’ ions and negligible when compared to the injection or storage energies of approximately 10 eV. This means that the initial ion of charge \( qe \), attains a kinetic energy of \( qK_1 \) when accelerated through a potential difference of \( \Delta V \), where the kinetic energy of a singly charged ion of mass \( M \) and velocity \( v \) is given by:

\[
K_1 = \frac{1}{2} Mv^2
\]  

(4.3)

The ions in this electrostatic storage ring experience a force,

\[
F = Ma = qeE
\]  

(4.4)

where \( E \) is the electric field strength and \( a \) the acceleration. In the case of the energy dispersive HDAs, a doubly ionized particle with exactly twice the energy of a singly charged ion will experience the same force as that singly charged ion. This can be shown for the median trajectory passing through the HDA, which follows a circular path of radius \( R \) and velocity \( u \) such that the acceleration of the ion is given by:

\[
a = \frac{u^2}{R}
\]  

(4.5)
Expressing the force on this particle in terms of the acceleration given above:

\[ F = \frac{Mu^2}{R} = \frac{2K_u}{R} = qeE \]  \hspace{1cm} (4.6)

where \( K_u \) is the kinetic energy of an ion travelling with speed \( u \). Rewriting this expression in terms of the physical constants of the storage ring shows:

\[ RE = \frac{2K_u}{qe} \]  \hspace{1cm} (4.7)

Therefore ions of energy \( K_u = qK_1 \), with corresponding charge \( qe \), will all have trajectories of the same radius \( R \), in the same electric field \( E \). Similar arguments can be made for multiply charged ions in the lens stacks of the storage ring, such that ions of arbitrary charge state, \( A^{+q} \), can re-circulate within the storage ring as long as their energies are given by \( qK_1 \). It should be noted that ions formed with significant initial kinetic energy \( K_0 \), will accelerate to \( K_0 + qe\Delta V \) during the injection process. These ions will not pass through the HDA unless \( K_0 \) is smaller than the resolution of the HDA. This, together with the nature of field-penetration technique, suppresses fast atomic ions produced by dissociative ionisation from having the appropriate energy for ion storage (i.e. \( K_0 \) is significant due to the conversion of bond energy to kinetic energy).

The thermal ions are injected into the storage ring using a field penetration technique that is controlled by selecting a specific injection time window after the ionizing electron beam has interacted with the gas. Specific subsets of charged species can be selected to enter the ring by changing the length of injection time window. If the window is shortened, only the faster moving multiply charged ions will penetrate far enough into the HDA to be stored before the HDA is pulsed back ‘on’. The storage of multiply charged argon ions is illustrated in Figures 4.7 and 4.8, where injection of \( \text{Ar}^+ \) has been suppressed to optimize the storage of \( \text{Ar}^{2+} \) and \( \text{Ar}^{3+} \) in the ring. Note that an \( ^{36}\text{Ar}^{2+} \) isotope has also been observed, which has a natural abundance of \( \sim 0.34\% \).
Figure 4.7: Spectrum of \(\text{Ar}^{2+}\), \(\text{Ar}^{3+}\) and isotope \(\text{Ar}^{2+}_{36}\) simultaneously being stored within the ring shown at 1x, 20x and 200x magnification. The series of peaks may be seen to pass through each other due to different orbit times. \(\text{Ar}^+\) ions were excluded from injection by shortening the injection window such that there was insufficient time for the slower singly charged ions to reach the lens stack before the HDA was pulsed back ‘on’ for storage.

Since each ion species has different orbit times, as they perform multiple orbits the ion pulses can merge and de-merge, passing through each other, with the possibility of interaction. Two possible interactions include: 1) the long range Coulomb interaction that results in space charge effects if the particle number density is high and 2) short range...
ion-ion collisions. Both processes are physically interesting and could result in further losses from the stored pulse however, an examination of the peak area decay in Figure 4.8 does not indicate any obvious changes or discontinuities in the loss mechanisms that may indicate a change in storage mode.

4.3 Probing the Storage Ring for Alternate Storage Modes

While the coherent diffusion model describes the peak width variation frequently observed in this storage ring, the ability to break the symmetry of operating potentials enabled further exploration into storage behaviour. Since at this time the energy and current density of the electron source is essentially fixed, different sets of operating potentials were tested to determine if other storage modes existed for this system. To that end, all the data presented in this chapter has been recorded using asymmetric operating conditions. In all cases HDA 1’s potential (the HDA for injection and detection) was greater than HDA 2’s potential such that the HDA 1 acts more like a mirror and HDA 2 more like a prism (as discussed in Chapter 2, Section 2.1.3). Therefore the resolution of the storage ring is defined by the operating potential on HDA 2. The middle lens elements ($V_2$) in the 3-element lenses, located before the entrance and exit of each HDA respectively, were set to the same potential (as discussed in Chapter 3, Section 3.4). Therefore changes in the operating conditions of the storage ring were defined by changes in a pair of $V_2$ voltages, where $V_{2,1}$ potential specifies the lens potentials for HDA 1 and $V_{2,2}$ for HDA 2.

Regions of storage, as described in Chapter 3 Section 3.4, for the storage ring were examined by performing an area scan where the $V_2$ potentials were varied sequentially for a set range of voltages and a spectrum was recorded for each $V_2$ pair. These spectra were fit with Gaussian peaks so that the peak width variation at FWHM as a function of storage time could be examined with changing $V_2$ lens potentials, identifying any links between storage modes and regions of stable storage.
4.4 Identification of Isochronous Behaviour in Electron Storage

4.4.1 Observation of Isochronous Storage Behaviour

An area scan for the asymmetric voltage configuration where HDA 1 = 18 V, HDA 2 = 6V, source/interaction = 18 V over the ranges 84.5 V ≤ V_{2,1} ≤ 85.5 V and 44.5 V ≤ V_{2,2} ≤ 45.5 V showed distinct changes in storage modes. In Figure 4.9, the peak width variations for two sets of lens potentials (V_{2,1} = 84.5 V, V_{2,2} = 44.5 V shown in red and V_{2,1} = 84.5 V, V_{2,2} = 45.5 V shown in blue) exhibited very different behaviours. The rate of change in the peak width decreased dramatically when the V_{2,2} potential was slightly (≤ 1V, or ~2% of V_{2,2}) de-tuned from the lens potential used for the spectrum in the area scan showing the highest count rate. The storage behaviour of the red data shows a dispersive storage mode characteristic of coherent diffusion and described by (4.1), where after ~10 orbits, \( W_0 = 23.85 ± 0.04 \) ns and \( \Delta T = 0.44 ± 0.04 \) ns. The storage behaviour of the blue data shows an isochronous mode, where after ~25 orbits the peak width stabilizes to a value of ~ 30-35 ns for the remaining 100 orbits. This second mode however, has significantly greater scatter in peak width values than the dispersive mode. This scatter is likely an artifact to the detection method that is more apparent for the changed.

Figure 4.9: Plot of peak width (FWHM) variations for long term storage (>4 μs) for dispersive (red) and isochronous (blue) modes. For the dispersive mode, after ~ 10 orbits the width varies according to (4.1) with \( W_0 = 23.60 ± 0.05 \) ns and \( \Delta T = 0.45 ±0.05 \) ns.
trajectories of the stored particles due to the change in the lens potentials between data sets. Basically non-paraxial electrons that would normally be passed through HDA 1 for storage are lost when travelling the field free region between the lens stack and the detector.

Isochronous behaviour is characterized by having a consistent peak width independent of orbit time. The total spread of peak width when plotted against orbit time for the isochronous data is significantly different than that of the dispersive data as shown in Figure 4.10. The peak widths for the isochronous data (blue) are limited to an ~10 ns spread between ~25 ns and 35 ns, whereas the dispersive data (red) shows peak widths over a 40 ns range between 20 ns and 60 ns. Both datasets show a similar range of orbit times as a function of orbit number, shown on the right of Figure 4.10.

**Figure 4.10:** Corresponding to the same data presented in Figure 4.9 *(left)* a plot of orbit time as a function of peak width for the isochronous data (blue) and dispersive data (red) and *(right)* a plot of orbit time as a function of orbit number. See text for discussion.

*4.4.2 Fitting Isochronous Data*

Pedersen *et al* [4], proposed two distinct forms of diffusion, *coherent* and *non-coherent* (as discussed in Chapter 1, section 1.3). Recall that non-coherent diffusion is given by:

\[
W_n = \sqrt{W_o^2 + n\Delta T^2}
\]  

(4.8)
which is a similar relation to the relation for coherent diffusion but with the dependence on orbit number $n$ reduced from $n^2$ to $n$. Non-coherent diffusion introduced the possibility of external perturbations (such as collisions with residual gas and noise on electrode potentials) while still excluding internal perturbations (such as Coulomb repulsion). Since the influence of external perturbations on any individual charged particle is based on the statistical chance that the particle has an interaction, it is possible that this effect is only noticeable after some number of orbits. The relation presented in (4.8) describes the case when external perturbations are present from the first orbit. Attempting to fit the data points in the isochronous data set was not successful using (4.1) or (4.8) (see Figure 4.11). This further confirms that this behaviour is not a dispersive storage mode (either coherent or non-coherent) and may indicate the presence of unknown internal perturbations. This is discussed further in Section 4.6.2.

![Figure 4.11](image-url)

**Figure 4.11:** The peak width variation has a unique trend which cannot be fit with either the quadratic curve characteristic of coherent diffusion (4.1) or the non-quadratic curve characteristic of non-coherent diffusion (4.8) relations for dispersive storage modes.

### 4.4.3 Isochronous Behaviour & Regions of Stability

It has been frequently noted that the predominant mode of storage observed for this storage ring is dispersive, which leads to the question of what operating voltage configurations are characteristic of isochronous storage. Initially, it was noted that peak widths became narrower when $V_2$ lens potentials were slightly detuned from those that yielded highest count rate for long term storage, (which typically indicates the voltage
configuration for longest storage). Plotting a series of spectra as a contour plot (discussed in Chapter 3, Section 3.4), one may observe that as the $V_2$ lens potentials are varied, the operating conditions of the system may pass from stable to unstable (and vice versa). Isochronous behaviour is observed on the edge of stability when operating conditions are changing between these stable and unstable regimes. Figure 4.12 shows a contour plot of two areas of stable long term storage, which are displayed as large bands of green, and in the transition between these two areas (see the black box superimposed in the plot) the peak width was plotted as a function of orbit time.

![Figure 4.12](image)

**Figure 4.12:** A contour plot (red indicates high counts, and blue indicates low counts) of the storage for 30 μs spectra collected while the $V_{2,1}$ lens potentials were set to 82 V and the $V_{2,2}$ lens potentials were varied in 0.4 V increments from 44.5 V to 49.3 V. (right) Plots of peak width as a function of orbit time for the corresponding $V_{2,2}$ potentials on the contour plot. These indicate that a change to isochronous behaviour occurred when the lens potential for HDA 2 was increased from 45.7 V to 46.1 V (＜1 %). See discussion on next page.
For successive spectra where $V_{2,2}$ was changed in 0.4 V increments, the dependence of peak width on orbit time is shown to be changing, becoming more isochronous as the system moves from one stable region to the next. In the fourth spectrum, which has a $V_{2,2}$ of 46.1 V, isochronous behaviour was observed before storage changed back to a dispersive mode at $V_{2,2} = 45.7$ V. The observation of isochronous storage only at the edge of regions of stable storage and when the system is detuned from maximum count rate, makes repeated observation of fully isochronous behaviour elusive. However, the observation of a dispersive mode becoming more isochronous (i.e. an increasingly flat quadratic) in these regions has been made several times for similar asymmetric configurations of operating potentials.

4.4.4 Edges of Stability and Changes in Decay Rates

Links between regions of stability and changes in temporal peak widths raised questions about other properties of spectra that change on the edge of stability. Specifically, questions about if there is any correlation between the changing behavior of the peak width variation and loss mechanisms within the storage ring. Examining the peak area and height decays for spectra on the edges of stability revealed that there are at least two distinct decay rates for each spectrum, regardless if the mode is dispersive or isochronous (see Figure 4.13 on the next page). The initial decay rate is very similar between dispersive (red) and isochronous (blue) storage however, the second decay rate that dominates for long term storage is greater for isochronous storage.

In Figure 4.13, the peak area as a function of storage time is shown for the two spectra on the edge of stability in Figure 4.12, one of which exhibits dispersive storage ($V_{2,2} = 45.7$ V (red)) and one that exhibits isochronous storage ($V_{2,2} = 46.1$ V (blue)). Since the trends in peak height as a function of storage time matched that of peak area, only the peak area is displayed for simplicity. Both datasets showed a distinct change in decay rate at ~ 20-25 orbits, after which the rate of decay decreased. Each data set was fit twice using a least squares fit; first the peak area was fit for orbits ~5-25 and then orbits ~35-85. The decay rates between the two fits changed less for the isochronous storage behaviour than the
dispersive storage behaviour, meaning that for long term storage the isochronous mode decayed faster. In general for this storage ring, a greater decay rate was always expected in the first few orbits due to the rejection of electrons from the pulse at apertures and in the HDAs due to energy and trajectory mismatch between injection and long term storage. The decay rate was then expected to level off when all these particles were eliminated from the pulse and any further losses were attributed to collisions with residual gases in the chamber.

Since the two data sets examined were measured consecutively, the pressure inside the storage ring was the same for both and the same decay would be expected for long term storage if this were true. These different decay rates imply that additional losses occur in the pulse during long term storage due to more than just collisions with residual gas. This suggests that there may be loss mechanisms in the isochronous storage mode which further refine the energy and trajectory spread of the pulse by continuing to eliminate outlying particles for many more orbits than the dispersive mode. A mechanism which continually reduces the energy and trajectory of stored particles with each orbit would be
of significant benefit to spectroscopy and high resolution source applications. To confirm that this is happening, the energy resolution of the stored pulse would need to be measured directly.

4.5 Evidence of Changed Behaviour in Ion Storage

Evidence of changing storage behaviour in which the peak width variation does not follow the quadratic trend has been observed for stored argon and nitrogen ions. For both datasets the storage ring was operating with HDA 1- HDA 2 asymmetry where HDA 1 = 6 V, HDA 2 = 12 V and the source and interaction regions = 6V. In Figure 4.14, the peak width variation of Ar$^{2+}$ is plotted as a function of time, clearly showing a change in storage behaviour. In the first 2 ms the peak width variation follows the common quadratic trend before switching to a much shallower curve for the remaining 5 ms. As previously discussed for electrons, we can speculate that in the first 2 ms, the injected pulse is independent of external perturbations and therefore shows the coherent diffusion described by (4.1) (shown in Figure 4.14 as a dotted line). After ~ 25 orbits, external perturbations, in particular interactions with residual gas may cause the storage to shift to non-coherent diffusion described by (4.8) (blue line). Alternatively, there is a shift in the

![Figure 4.14](image)

**Figure 4.14:** Plot of peak width as a function of storage time for Ar$^{2+}$ ions, showing a distinct change in storage mode after ~25 orbits. The same dataset has been fit for two cases: (a) shifting from coherent diffusion (dotted) to non-coherent diffusion (blue) and (b) shifting between two different coherent modes (from dotted to solid black line). See text for discussion.
coherent diffusion storage such that its behaviour is described by a much shallower quadratic (black line). However, considering the values of $W_o$ and $\Delta T$ produced by these fits, neither case describes the physical reality of the storage ring while this measurement was performed. Reasonably, we may neglect the changes in the values of $W_o$ as artefacts of the least squares fitting procedures being applied only after 25 orbits. However, $\Delta T$ should still accurately describe the time spread in the ion pulse due to $\Delta T_S$ from the source and $\Delta T_{SR}$ from the storage ring.

Since this data is from a single measurement of a spectrum, there is no reason to assume any changes in the source, meaning $\Delta T_S$ remains constant. $\Delta T_{SR}$ depends on the trajectories of ions within the storage ring. If storage changes from one coherent mode to another coherent mode there is no mechanism for $\Delta T_{SR}$ to change, so the peak width for long term storage should not be fit with (4.1). If storage changes from coherent diffusion to non-coherent diffusion, then according to Pedersen et al [4], external perturbations are beginning to affect the dynamics of the pulse after ~25 orbits. The only known external perturber in the storage ring is residual gas. However, for this fit $\Delta T$ increases significantly (from $0.27 \pm 0.05 \mu s$ to $1.53 \pm 0.07 \mu s$) and by comparison to other datasets that show purely coherent diffusion the pressure in the storage ring is relatively low (~1.3 x $10^{-6}$ Torr), which makes it unlikely that external perturbation as defined by (4.8) is the cause for the change in storage behaviour.

Figures 4.15 and 4.16 show stored $\text{Ar}^+$ and $\text{N}_2^+$ respectively, under the same asymmetric operating conditions and for both these cases there is no evidence of the quadratic peak width variation. Instead, a different non-linear behaviour is shown that is becoming more isochronous. It was difficult in many cases to determine with certainty the nature of storage modes for ions as there are fewer peaks in a spectrum to analyse and at times fewer counts within those peaks. Additional residual gas within the chamber significantly shortens the relative storage time under non-ideal storage conditions for ions when compared to electrons.
Figure 4.15: Peak width variation with respect to storage time for $\text{Ar}^+$ ions, showing non-quadratic, non-linear behaviour. Note, there are many fluctuations in peak width which contribute to the changing error bars.

Figure 4.16: Peak width variation with respect to storage time for $\text{N}_2^+$ ions showing non-linear, non-quadratic trend, seemingly representative of a non-coherent diffusion storage mode (see text for discussion).

Since all evidence of isochronous storage has been observed on the edge of stability this makes it difficult to collect sufficient data for these voltage configurations. If the vacuum system for the storage ring were updated to reduce the residual gases in the chamber from 

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~10^6 Torr and if true differential pumping was implemented for the source, storage times would be improved. Also, if an alternate ion source was implemented that is capable of shorter injection pulses (reducing $W_o$), it would be easier to obtain distinct separate peaks for fitting.

The Ar$^+$ peak width data shown in Figure 4.15 appears to be changing more linearly than the dispersive modes described by (4.1) or (4.8). A careful examination of the data points reveals that the variation in peak width is fluctuating, potentially showing a storage mode switching between dispersive and isochronous modes. The N$_2^+$ data shown in Figure 4.16 more clearly indicates a non-coherent diffusion mode described by (4.8) as the fit agrees within the scatter for all but the first few data points. However, as discussed previously, it is difficult to attribute this behaviour to external perturbations due to residual gas when this behaviour is not observed in other datasets measured with similar residual gas pressures. Since this storage mode was observed by making a change in the V$_2$ lens potentials, it suggests that the fit using (4.8) is a convenient mathematical model rather than derived from a physical model representing our storage ring. Both datasets presented in Figures 4.15 and 4.16 still provide evidence of alternate storage modes for ions and the potential for an isochronous storage mode to emerge with better long term storage.

4.6 Potential Explanations for Observed Phenomena

4.6.1 Matching Operating Potentials to Trajectories

In Chapter 1, three examples of isochronous storage were presented for three different electrostatic systems of different geometry than the storage ring used for this work. Each system had differing but related explanations of the isochronous behaviour they observed. The two systems designed for mass spectrometry [1,2] had physical parameters designed specifically to access isochronous storage. This was accomplished by using transfer matrices to predict trajectories through the proposed elements which would allow for particles of differing energies to arrive at the same point at the same time. This phenomena where particles of differing energies travel slightly different paths to achieve
isochronous storage, relates to the kinematic criterion described by Zajfman [4,5] for his observed self-bunching mode, where \( dT/dE = 0 \) for which there is no minimum charge density requirement. In all cases, specific operating potentials were used on electrostatic elements to let particles of faster energies penetrate further into bending elements than slower particles. As a result, a balance is achieved between particle speed and path length travelled and the charged particle pulse does not spread with increased storage time.

The most likely explanation for the observed isochronous behaviour in this storage ring is that this kinematic criterion has been met for specific voltage configurations, as described by Wollnik’s energy-isochronous mode [1]. This means that the combination of the energy distribution of the charged particles and the operating conditions of the system is such that their trajectories yield the same peak width independent of orbit number [1]. There is, however, a marked difference in how this is achieved in this storage ring from the previous systems mentioned as all the other apparatuses are operated solely using symmetric operating potentials. While further analysis of the pulse dynamics within this storage ring is required to fully identify the cause of the isochronous behaviour, some comparison to the previously identified self-bunching mode and perfect focusing mode as defined by Zajfman [4,5] and Toyoda [2] respectively is instructive.

4.6.2 Comparison to the Self-Bunching Mode

The self-bunching mode is specifically defined in terms of a charge density requirement such that the internal perturbation due to Coulomb repulsion is significant enough to affect the spreading of the pulse. The charge density requirement is met in Zajfman’s device by maintaining sufficiently large injection of ions and by the nature of the systems bending elements. Since the trap is linear, ions are slowed at the turning points, occupying a small space where the pulse will pass through itself. This allows for sufficient interaction time between ions in the pulse for competing Coulomb repulsion effects from fast and slow ions to compress the pulse to a constant lateral size (as discussed in Chapter 1, section 1.3) [4,5].
In this storage ring, neither of these conditions is met. Since charged particles are detected using single particle counting instead of pick-up electrodes the current density that can be safely detected is limited. To gain an idea of how many charged particles are in the ring at any given time, it should be noted that for a single peak in a spectrum, injection will occur as many times as the BNC pulsing unit cycles in the dwell time specified between shifting the HDA detection pulse. For example, for ions stored with a repetition rate of 7 ms, and a time between a shift in detection pulses of 1 s, there will be $\sim 142$ injection pulses detected separately to create the peak. For isochronous storage, the count rate is significantly less and therefore the timing between detection pulses is 8 s to 10 s, for the same repetition rate. This means that a peak of 50 000 counts is made up of approximately 1100-1400 pulses, and in each pulse there would be about 35-45 ions. Furthermore, the bending elements in the storage ring are HDAs spatially separate charged particles in two dimensions according to energy such that faster particles complete a longer trajectory than slower particles (see Chapter 2, Section 2.1.3) only bringing the particles back together at the exit of the HDA. This density is insufficient for the internal Coulomb repulsion within the pulse to create a self-bunching mode.

4.6.3 Comparison to Perfect Focusing

In the theoretical work presented in [6] by Sakurai et al, it was shown that the perfect focusing condition could not be achieved for a planar symmetric system of two sector field analysers (i.e. HDAs). It should be noted that in [6], the planar symmetric system treated is simplified for symmetric operation and does not account for any electrostatic elements connecting the two analysers. For this case, there is no solution that allows for the perfect reproduction of a charged particle pulse’s initial parameters at detection for every orbit. The transfer matrices and simulation of the current storage ring for symmetric operation (presented briefly in Chapter 3) suggests that the trajectories of stored particles will retrace themselves, but in two or more orbits depending on the storage potentials [7]. By definition, this behaviour does not represent perfect focusing. Furthermore, attempts to observe isochronous storage at the edges of stability for symmetric voltage configurations did not yield any convincing results. Whether it is
possible to meet all of the perfect focusing conditions when using an asymmetric operating configuration and by accounting for the contribution of the two cylindrical lens stacks is at this point unknown. Only recently has a generalized transfer matrix been developed for this storage ring (presented in [8]). It remains challenging to apply the nine constraints that define perfect focusing to a generalized transfer matrix for the storage ring or to simulate the corresponding particle trajectories for asymmetric operating conditions.

4.6.4 Generation of a Mono-Energetic Pulse

One final explanation for the isochronous behaviour is that the charged particle pulse is becoming more mono-energetic as more orbits are completed. Particles of differing energies are pushed farther off the optical axis by the HDAs until they are rejected by the apertures within the ring, such that only particles of a small energy range remain. Since particles of the same energy are subject to the same potentials and travel the storage ring at the same speed, a pulse of mono-energetic particles would be detected with the same width independent of storage time. Considering the initial energy spread of thermal electrons injected into the storage ring, this seems unlikely, but could be confirmed by measuring the energy resolution of the stored pulse directly. There were two attempts made to do this, first using a simple retarding field analyzer and then using a small hemisphere as an energy analyzer. Neither analyzer had a sufficiently small energy resolution to allow for reliable measurement. Further work is required in this area.

4.7 Implications of Isochronous Storage for Mass Spectrometry

One of the primary reasons to access an isochronous storage mode is to be able to perform high resolution mass spectrometry. In order to obtain a high resolution in mass spectrometry, (i.e. for the peaks representing different atomic masses to be observable as they separate in time), the width of the peak must be minimized. However, as was shown for the case of Ar⁺, under most operating conditions the peak width of the stored beam in our storage ring is increasing quadratically according to (4.1).
Consider the following quick approximation to see how mass resolution may be improved by changing the peak width variation as a function of storage time. If the time $t$ for a charged particle of mass $M$ (in amu), to orbit the storage ring $n$ times is given by:

$$ t_n = \frac{nL}{v} = \frac{nL}{\sqrt{2K}} \sqrt{M} \quad (4.9) $$

where $L$ is the path length of the ring, $v$ is the average particle velocity and $K$ is the average kinetic energy of the particle. Using the binomial expansion for the time difference between ions of adjacent mass numbers, one obtains:

$$ \Delta t_n = \frac{nL}{\sqrt{2K}} (\sqrt{M+1} - \sqrt{M}) \approx \frac{nL}{\sqrt{2K}} \frac{1}{2\sqrt{M}} \quad (4.10) $$

This quantity needs to be compared to the peak width $W_n$ given in (4.1):

$$ \left( \frac{\Delta t}{W} \right)_n \approx \frac{nL}{\sqrt{2K}} \frac{1}{2\sqrt{M}} \frac{1}{\sqrt{W_o^2 + n^2 \Delta T^2}} \quad (4.11) $$

which can be simplified further by recognising that $t_1$ is the orbit period and by considering $M$ as a unitless mass number, i.e.:

$$ \left( \frac{\Delta t}{W} \right)_n \approx \frac{nt_1}{2M} \frac{1}{\sqrt{W_o^2 + n^2 \Delta T^2}} \quad (4.12) $$

This relation indicates that the mass resolving power of the storage ring can be improved for any given $M$ and $n$, by reducing the initial temporal peak width $W_o$, or the time spread per orbit $\Delta T$. The initial temporal peak width is limited by the ion source and our ability to inject sufficient charged particles in a narrow time window coupled with the ability to rapidly ‘turn on’ and ‘off’ the lower HDA without introducing significant noise on neighbouring elements. $\Delta T$ relies on the injected particles being as mono-energetic as possible, which in our system, can again only be significantly improved by a change in the source and injection method.
Alternatively, it can be observed that this relation could be improved for the same $W_o$ and $\Delta T$ values if the temporal peak width variation $W_n$ is governed by an alternate relation that reduces its dependence on $n$. For example consider the case of non-coherent diffusion described by (4.8). Replacing the peak width relation in (4.12) and allowing $n \rightarrow \infty$, it becomes clear that with increasing orbit resolution improves:

$$
\left( \frac{\Delta t}{W} \right)_n \approx \frac{nt_1}{2M} \sqrt{\frac{1}{W_o^2 + n\Delta T^2}} \rightarrow \left( \frac{t_1}{2M\Delta T} \right) (\sqrt{n})_{n \rightarrow \infty} \Rightarrow \infty \quad (4.13)
$$

Extending this discussion to apply to a fully isochronous mode where the peak width remains constant, independent of orbit number; the peak width relation would be replaced with a constant $C$ that describes the initial temporal width of the pulse from the source and first orbit of the ring. This gives the following relation where mass resolution will improve with each orbit of the ring.

$$
\left( \frac{\Delta t}{W} \right)_n \approx \frac{nt_1}{2MC} \quad (4.14)
$$

As previously mentioned, this improvement in mass resolution by using isochronous storage has been used in linear traps developed (independently) by Zajfman [5] and Wollnik [1] and in Toyoda’s multi-turn TOF system [2] (as described in Chapter 1, Section 1.3). Operating in an isochronous mode with behaviour similar to Wollnik et al.’s [1] energy-isochronous behaviour, Zajfman et al.’s [5] self-bunching mode or Toyoda’s et al.’s [2] perfect focusing configuration would make a dramatic increase to the mass resolution of the storage ring. While isochronous behaviour has been observed in electrons and evidence of an alternate, non-dispersive storage mode has been observed for ions, at this time these observed modes have not been applied to mass spectrometry applications in this storage ring. A more in depth discussion of the storage ring’s performance to date as a mass spectrometer has been presented by Michael Sullivan in [8].
4.8 Summary

The storage behaviour of electrons and ions was examined for asymmetric storage configurations by comparing trends in peak width at FWHM as a function of storage time. Isochronous storage behaviour has been observed for electrons and evidence of changing storage behaviour between dispersive and isochronous modes has been observed for singly and multiply charged ions. Observations of these behaviours were challenging to achieve as they are made by probing the edges of stable storing voltage configurations of the storage ring under asymmetric operating potentials. Potential explanations for these observations have been explored, considering the possibility of non-coherent diffusion and self-bunching modes described by Zajfman, perfect focusing described by Toyoda and energy-isochronous behaviour described by Wollnik. It is the conclusion of the author that the observed isochronous behaviour is due to a kinematic matching of the energy spread of the particles in a stored pulse, the operating voltage configuration of the storage ring and the trajectories of the stored particles. The ability to access this isochronous storage mode provides the potential to conduct high resolution mass spectrometry.
4.9 References


CHAPTER V: FUTURE WORK & CONCLUSION

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By altering the injection timing, lens voltages and HDA pass energies within the storage ring, storage behaviours were examined with a particular focus on probing the edges of stable storage regions for asymmetric HDA pass energies. The changing behaviours that are observable at the edges of stable storage conditions show the existence of isochronous storage for electrons and evidence of a change in storage behaviour for ions. This isochronous behaviour yields a pulse width that does not change with storage time, a property that has been used successfully by Wollnik [1], Toyoda [2] and Zajfman [3] for high resolution mass spectrometry applications. Further theoretical and experimental work is required to fully characterize the isochronous mode observed.

5.1 Future Experimental and Theoretical Work

There are several obvious directions for future experimental work. The energy resolution of the system must be studied to confirm that the observed isochronous behaviour exists due to the ‘coupling’ of varying energies and trajectories of the stored particles and not because the pulse is becoming monoenergetic. The current density of the pulse should also be measured directly to inform about any significant internal mechanisms (e.g. space charge) affecting storage behaviour. To further examine mass spectrometry applications and improve data collection for analyzing the storage of ions, the ion source needs to be replaced with a source that can inject a pulse with a much smaller initial width (e.g. laser ionization source) and the source should be fully differentially pumped to further reduce the residual gases in the chamber, enabling longer storage lifetimes.

As part of future theoretical work, the storage ring should be modelled for asymmetric storage configurations and the resulting trajectories of particles examined. There is significant challenge in extending the current simulation of the storage ring created in CPO3D since the program relies heavily on symmetry to simplify the computation of trajectories. Tracking particle trajectories according to energy, especially at the edge of
stable regions, will identify where in the storage ring particles are being lost from the pulse. This may correlate to the observed dispersive and isochronous storage behaviour.

Furthermore, the constraints for perfect focusing should be applied to both the symmetric and general matrix formalisms developed for storage ring, (presented in [4,5]). The use of asymmetric operating potentials and the inclusion of the lens stacks in the matrix formalism may reveal the existence of a perfect focusing mode for a two sector field analyser system, contrary to the assertion in [6].

5.2 Conclusions

Extending the function of this storage ring to operating isochronously is a significant advancement when considering that the ring is uniquely able to operate asymmetrically and at such low energies. Being able to examine the behaviour of the stored pulse by probing edges of stable storage highlights the inherent flexibility of the ring and its potential to be adapted for to new applications. By exploring these possibilities in this work, the following has been achieved:

- Observed isochronous behaviour in electron storage
- Observed changing storage behaviour in ion storage
- Determined that these behaviours are observable at the edge of stable storage regions under asymmetric operating conditions
- Assessed potential explanations for observed behaviours through comparison to other systems operating with isochronous storage modes
- Determined the next steps required to confirm the proposed explanation

In conclusion, the analysis of peak width variation at the edge of stable storage for asymmetric operation of the storage ring has revealed the existence of isochronous behaviour in the stored pulse. This behaviour is observable by slightly detuning the middle lens elements ($V_2$) in the lens stacks of the storage ring by ~1-2% from the potentials required for longest stable storage. The observed behaviour for both electrons
and ions is most likely achieved by matching the voltage configuration of the storage ring with the trajectories and energies of the stored particles, such that the natural energy spread in the pulse is compensated by the differing path lengths travelled. Further measurements of the energy and current density of the pulse or simulation of particle trajectories in the storage ring under asymmetric potentials are required for confirmation. The observation of this behaviour relied on the inherent flexibility of the storage ring’s design and has the potential to enhance its applications to new endeavors such as mass spectrometry.
5.3 References

APPENDIX A

LOSS MECHANISMS FOR ELECTROSTATIC STORAGE RINGS

There are a number of loss mechanisms both design related and inherent which inhibit long term storage of charged particles. The loss mechanisms that have been minimized through design processes in our system include: residual magnetic fields near the electron orbital path, mechanical misalignment of electrostatic components along the optical axis, ‘patch’ or ‘parasitic’ fields on lens elements, and voltage supply instabilities. In brief, losses due to these elements have been minimized through the choice of appropriate materials and components, and rigorous construction. The full details of these design considerations may be found in Niu Y. et al., *The Design, Construction and Operation of an Electrostatic Storage Ring for Low Energy Charged Particles* (in preparation). Inherent loss mechanisms present in all electrostatic storage rings include: losses due to radiation, space charge effects, collisions with residual background gas and aberrations in electrostatic lenses (most notably, hemispheres).

Since this storage ring is operated at low energy the losses due to radiation are negligible. If we consider the classical formula for the radiated power $P$ from an accelerated electron as:

$$P = \frac{2}{3} \frac{e^2}{4\pi\varepsilon_0 c^3} a^2$$  \hspace{1cm} (A.1)

where $e$ is the charge of an electron, $\varepsilon_0$ is the permittivity of free space, $c$ is the speed of light and $a$ is the acceleration of the electron. For a non-relativistic circular orbit, acceleration is simply $v^2/r$, since for relativistic motion $a = \gamma^2 v^2/r$ and the time rate of change of $\gamma$ is $\sim 1$ for non-relativistic mechanics. Radiated power is then given by:

$$P = \frac{2}{3} \frac{e^2}{4\pi\varepsilon_0 c^3} \frac{v^4}{r^2}$$  \hspace{1cm} (A.2)
Converting the expression to one in terms of frequency using: \( v = r\omega \) and \( \omega = 2\pi f \) yields:

\[
P = \frac{2}{3} \frac{e^2}{4\pi \varepsilon_0} \left(\frac{2\pi f}{c}\right)^4 = \frac{2}{3} \frac{e^2}{4\pi \varepsilon_0} \left(\frac{2\pi}{c}\right)^4 r^4 f^4
\]

(A.3)

In our case the circumference \((2\pi r)\) or path length of the storage ring is \(\approx 0.65\) m, and the orbit period is \(\approx 250\)ns such that \( f = 4\) MHz. Applying these values to (A.3):

\[
P = \frac{2}{3} \frac{(1.6\times10^{-19})^2 \pi}{8.854\times10^{-12}} \left(\frac{0.65}{3\times10^8}\right)^3 \left(\frac{4\times10^6}{c}\right)^4
\]

\[
= \frac{2}{3} 9.08\times10^{-27} \times 1.565\times10^{-26} \times 2.56\times10^{26} = 2.425\times10^{-26} W
\]

\[
P = 1.5\times10^{-7} eV/s \approx 3.8\times10^{-14} eV/orbit \text{ (per electron)}
\]

This confirms that these losses are negligible for this application.

Space charge effects refer to the repulsive Coulomb forces between charged particles within the pulses traveling the ring. These effects increase the energy width \(\Delta E\) of the beam as ‘like’ charges repel each other. As charged particles travel along the optical axis, cylindrical lenses focus the bunch radially, limiting its spreading due to space charge effects. In the direction of the optical axis however, the pulse will spread more at the end points and less in the centre of the pulse as there is no clear direction to the repulsion force. This spreading increases the energy width of the pulse and as a result some of these particles may not meet pass energy requirements of hemisphere elements and be lost to the stored pulse. Space charge effects also limit the current that can be transmitted through an aperture. Both these effects cause the most significant losses when the stored pulse has high current density. In our system the stored pulses have low current density and this is thought to be a very small loss mechanism.
Of more significance to our storage ring, and any storage ring where there are interactions with inert gas are losses due to collisions with residual background gas. The mean free path $\lambda$, of any charged particle is related to the gas number density $n$ and total charged particle cross section $\sigma$ for the mean energy in the ring according to:

$$\lambda = 1/n\sigma$$  \hspace{1cm} (A.4)

Consider the ideal gas law:

$$n = \frac{P}{kT} = \frac{1.01\times10^5}{1.38\times10^{-23} \times 300} \frac{P}{760} = 3.21\times10^{22}P$$ \hspace{1cm} (A.5)

where $P$ (Torr) represents the average pressure along the optical axis near the stored pulse. An expression for the mean free path can be calculated in terms of pressure and particle cross-section with the storage ring. As the mean free path decreases with increased residual gas pressure, the losses increase.

Finally, there will be losses in storage rings due to aberrations in electrostatic components that affect the trajectories of charged particles. If the trajectories of charged particles stray too much from the optical axis of the ring, particles will be lost as they impact apertures, lens edges and are rejected by the hemispheres. Chromic aberrations are common in hemispherical analysers since their primary function is to be energy dispersive and charged particles of undesired energy will be lost from the pulse actually improving the energy resolution of the beam. As mentioned in Chapter 1 Section 1.3, electrostatic lens elements follow charged particle optics which are subject to all the standard optical aberrations. Most serious in terms of losses is spherical aberration. Many storage rings, including ours, use paraxial rays and cylindrical lenses instead of aperture lenses to minimize spherical aberration. However, this does not eliminate losses due to trajectories with large radial positions or trajectory angles that may be perturbed over many orbits and be lost through impact with apertures in the ring.
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