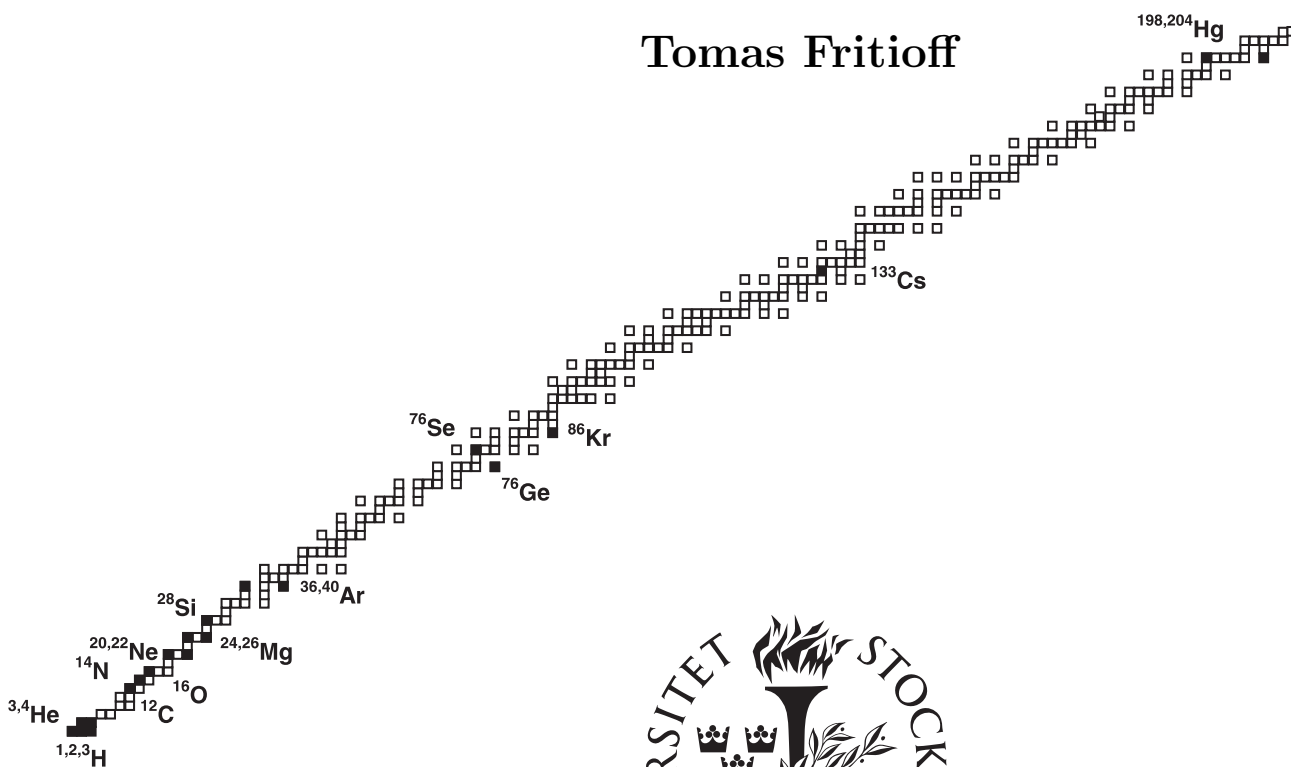


Precision Improvements of Penning Trap Mass Measurements Using Highly Charged Ions

Applications to solving current problems in
fundamental physics

Tomas Fritioff



Department of Physics
Stockholm University
2002

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Abstract

In my thesis I describe the improvements of the Penning trap mass spectrometer SMILE-TRAP. The objective of these improvements have been to increase the reliability and the accuracy with which an atomic mass can be measured using highly charged ions. The improvements have been achieved by stabilizing both the electric and magnetic fields of the trap and by improving the technical performance of the trap system. As a result it has been possible to measure accurately the mass of several atoms ranging from hydrogen to mercury using charge states from $1+$ to $52+$. It was only possible to use the highest charge states after applying a successful cooling of these ions with Helium during the charge breeding.

The technical improvements made a number of interesting accurate mass measurements possible. The measurements of the ^3H , ^3He , and ^4He masses showed that the previously values were wrong. The mass difference between ^3H and ^3He which is the Q -value of the tritium beta decay has been determined to $18.588(3)$ keV. The Q -value of the double β -decay of ^{76}Ge was measured at an accuracy of 50 eV. This value is indispensable for the evaluation the Heidelberg-Moscow experiment which aims at finding a possible neutrino-less decay which if present would be a violation of the standard model. The mass ratio of m_{Cs}/m_p is used to determine the fine structure constant independent of QED calculations. The two decades old anomaly in the mass values of Hg was solved by the mass determination of ^{198}Hg and ^{204}Hg . The mass of ^{24}Mg was measured at an uncertainty of 0.6 ppb and will be used in the determination of the g -factor of a bound electron in a hydrogen like ions.

To (is it to you?)

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List of Papers

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- I SMILETRAP-A Penning trap facility for precision mass measurements using highly charged ions**
I. Bergström, C. Carlberg, T. Fritioff, G. Douysset, J. Schönfelder and R. Schuch
NIM A**487**, 618-651 (2002)
- II A New Determination of the ^3He and ^4He Masses in a Penning Trap**
T. Fritioff, C. Carlberg, G. Douysset, R. Schuch and I. Bergström, Eur. Phys. J. D **15**, 141-143 (2001).
- III Mass determination of ^{22}Ne , ^{36}Ar , and ^{86}Kr using highly charged ions in a Penning trap**
T. Fritioff and G. Douysset, submitted to Nuclear Physics A
- IV High-Precision Mass Measurements of Hydrogen-Like $^{24}\text{Mg}^{11+}$ and $^{26}\text{Mg}^{11+}$ ions in a Penning Trap**
I. Bergström, M. Björkhage, K. Blaum, H. Bluhme, T. Fritioff, Sz. Nagy, and R. Schuch, submitted to Eur Phys J D.
- V On the Masses of ^{28}Si and the Proton Determined in a Penning Trap**
I. Bergström, T. Fritioff, R. Schuch, and J. Schönfelder, Physica Scripta **66** (2002) 201-207
- VI Determination of the ^{76}Ge Double Beta Decay Q-value**
G. Douysset, T. Fritioff, C. Carlberg, I. Bergström and M. Björkhage, Phys. Rev. Lett. **86** (2001) 4250-4262
- VII Determination of the ^{133}Cs and Proton Mass Ratio Using Highly Charged Ions**
C. Carlberg, T. Fritioff and I. Bergström, Phys Rev. Lett. **83** (1999) 4506
- VIII Shedding Light on the Mercury Mass Discrepancy by Weighing Hg^{52+} Ions in a Penning trap**

T. Fritioff, H. Bluhme, R. Schuch, I. Bergström and M. Björkhage, submitted to Nuclear Physics A.

Papers not included in this thesis

IX Recent Progress with the SMILETRAP Penning Mass Spectrometer

T. Fritioff, Sz. Nagy, R. Schuch, I. Bergström, M. Björkhage, G. Douysset, and H. Bluhme, to appear in Proc. of Conference on Physics Beyond the Standard Model (Beyond the Desert 02), Oulu, Finland, June. 2-7, 2002

X Back to the Line of Stability, Chasing Mass Accuaracies Below 10^{-9} Using a Penning Trap and Highly Charged Ions

T. Fritioff, H. Bluhme, R. Schuch, I. Bergström and M. Björkhage to appear in Proc. of 3 rd International Conference on Exotic Nuclei and Atomic Masses (ENAM 2001), Hämeenlinna, Finland, July 2-7, 2001

XI Recent Progress with the SMILETRAP Penning Mass Spectrometer

T. Fritioff, C. Carlberg, G. Douysset, R. Schuch, and I. Bergström, in Proc. of 2nd Euroconf. on Atomic Physics at Accelerators: Mass Spectrometry (APAC 2000), Cargèse, Corsica, France, Sep. 19-23, 2000, eds. D. Lunney, G. Audi, and H.-J. Kluge, Hyperfine Interactions Vol. **132** Nos 1-4 (2001)

XII Precision Mass Measurements Using Highly Charged Ions From an Electron Beam Ion Source

I. Bergström, C. Carlberg, G. Douysset, T. Fritioff and R. Schuch, in Proc. of 8th Int. Symp. on Electron Beam Ion Sources and Traps, and their Applications: 8th International Symposium, Upton, New York, Nov. 5-8 2000, ed. Krsto Prelec, AIP Conference Proceedings **572** (2001)

XIII Present status of the Stockholm electron beam ion source and its scientific program

I. Bergström, Mikael Björkhage, Håkan Danared, H. Cederquist, T. Fritioff, L. Liljeby and R. Schuch in Proc. of Electron Beam Ion Sources and Traps, and Their Applications: 8th International Symposium, Upton, New York, Nov. 5-8 2000, ed. Krsto Prelec, AIP Conference Proceedings **572** (2001)

XIV High Precision Mass Spectroscopy Using Highly Charged Ions in a Penning Trap

G. Douysset, T. Fritioff, C. Carlberg, and I. Bergström, in Proc. of 10th Int. Conf. on the Physics of Highly Charged Ions (HCI 2000), Berkeley, California, USA, July 30-Aug. 3, 2000, Physica Scripta Vol **T92** (2001)

XV Accuracy Tests of Atomic Mass Measurements in a Penning Trap Using Externally Produced Highly Charged Ions

C. Carlberg, H. Borgenstrand, T. Johansson¹, R. Schuch, I. Bergström, G. Rouleau, J. Stein, and U. Surkau, in Proc. of 8th Int. Conf. on the Physics of Highly

¹I changed my name from T. Johansson to T. Fritioff in 1998

Charged Ions (HCI96), Omiya, Saitama, Japan, Sep. 23-26, 1996, ed. Y. Awaya, and T. Kambara, Phys. Scr. **T73**, 347-353 (1997).

Chapter 1

Introduction

A Penning trap is a storage device for ions using a combination of static electric and magnetic fields to confine the ions. In 1989 Hans Dehmelt shared the Nobel prize in Physics "for the development of the ion trap technique" together with Wolfgang Paul and Norman Ramsey. He gave his trap design, with a homogeneous magnetic field and an electric quadrupole field, the name Penning trap to honour the Dutch physicist Frans Michel von Penning. In 1937 von Penning published a paper on a novel vacuumeter [1] that measures the current between a ring anode and two plate cathodes (one on each side of the ring). The vacuumeter used a homogeneous magnetic field perpendicular to the plates that forces the electrons to travel in spirals towards the ring and therefore each electron can collide and ionize several rest gas atoms before it is collected on the ring. In 1949 an improved design was published [2] using a cylinder instead of a ring (Figure 1.1). It was this vacuumeter that Dehmelt remembered in his search for a device in which he could study ions at rest for long periods of time. In one of Dehmelt's measurements with his trap $g - 2$ of the electron and the positron was measured at an accuracy of 1 part in 10^{12} [3].

A short introduction to mass measurements using Penning traps

When applied as a mass spectrometer the Penning trap makes use of the fact that frequency is the quantity that can be measured most precisely. The combination of an electric quadrupole field and a homogeneous magnetic field used in the trap separates the motion of an ion into three eigen-motions with simple analytic solutions. From these frequencies it is possible to determine the free cyclotron frequency ν_c which is related to the mass via the simple relation:

$$2\pi\nu_c = \frac{qB}{m}. \quad (1.1)$$

Here m is the mass of a particle with charge q that is moving perpendicular to a magnetic field B .

The cyclotron frequency can either be determined by measuring the small image current created in the trap electrodes or by a time-of-flight technique that was proposed by Bloch[4] and later developed by Gräff *et al.*[5]. In the work presented in my thesis the latter method is used. After the ion cyclotron motion is excited with an azimuthal quadrupole radio frequency field the ions are ejected out from the trap. If the ion is in resonance with the applied RF-field the cyclotron orbit will increase and as a consequence its magnetic moment also increases. In the gradient ∇B of the magnetic field B a force F will act upon the ion.

$$\vec{F} = -\vec{\mu} \cdot \nabla \vec{B} \quad (1.2)$$

The corresponding acceleration will increase the axial energy and as a consequence of this effect the ion flight-time to the detector decreases. Therefore, ions in resonance with the applied excitation have a shorter flight-time than ions out of resonance. By scanning the frequency and measuring the average ion flight-time for each frequency it is possible to detect a resonance as a pronounced minimum in the time-of-flight. In this experiment an excitation time of 1 second is used which results in a frequency line width of ~ 1 Hz, since it is a Fourier-limited process. The corresponding resolving power for an ion with $q/A=0.5$ is 3.6×10^7 but the center can be determined to ~ 1 % of the FWHM and thus it should be possible to reach a statistical uncertainty of a few parts in 10^{10} . In order to keep the resolving power high when heavier masses are measured it is an advantage to increase the charge state of the ion since

$$\frac{\delta\nu}{\nu} \propto \frac{m}{q}. \quad (1.3)$$

It is not possible to determine the magnetic field to a precision accurate enough than by determining the cyclotron frequency for an ion with a well known mass. The ideal

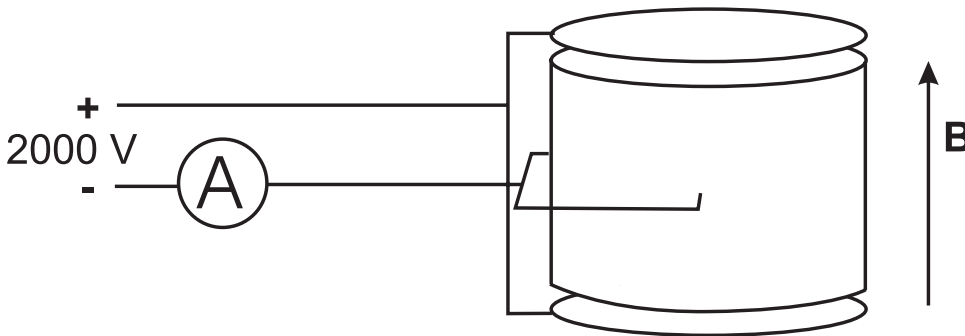


Figure 1.1: The second type of vacuumeter by von Penning inspired Dehmelt when he designed his trap. The vacuum is measured by the current between the anode cylinder and the cathode plates.

reference mass would be a ^{12}C ion since its mass by definition is 12 u. For experimental reasons H_2^+ ions were also used in this work, since this ion has $q/A=1/2$ *i.e.* similar to many highly charged ions used in this experiment. In order to eliminate a possible B -field dependence the cyclotron frequency of the reference ion and the highly charged ion of interest is alternatively measured in a time shorter than 1.5 minutes.

The mass of the ion is then deduced from the observed frequency ratios:

$$R = \frac{\nu_1}{\nu_2} = \frac{q_1 m_2}{q_2 m_1} \quad (1.4)$$

where the highly charged ion and the reference ion are denoted with subscript 1 and 2, respectively. It should be noted that the relevant quantity is a frequency ratio and thus several systematic errors cancel when the measurements are performed under similar conditions.

To deduce the atomic mass (M) of the measured highly charged ion one has to correct for the mass $q_1 m_e$ of the missing electrons and their total binding energies E_B :

$$M = \frac{1}{R} \frac{q_1}{q_2} m_2 + q_1 m_e - \frac{E_B}{c^2} \quad (1.5)$$

The SMILETRAP facility is a hyperboloidal Penning trap mass spectrometer with a 4.7 T magnet (Figure 1.2). The trap is connected to an electron beam ion source EBIS, named CRYISIS, that is able of producing highly charged ions of any stable element. The ions are transported to SMILETRAP where a 90° magnet selects the desired charge state. The ions are first retarded and then trapped in a cylindrical Penning trap, the pretrap. In this trap as a maximum a few thousand ions are trapped to be directly sent to the hyperboloidal precision trap. After a selection procedure as an average one ion is trapped. Thereafter, the ion is excited by a RF-field and its time-of-flight is measured as described above. The pretrap is not only used to remove the transportation energy of the ions. It is also used to produce the H_2^+ ions that are used as mass reference. This is done by electron bombardment of the rest gas.

The development of SMILETRAP

My undergraduate thesis presented in May 1997 was entitled "Accuracy Tests of The SMILETRAP Mass Spectrometer Using Singly Charged Light Ions". This paper reported comparisons of the and expected frequency ratios of the ions of H^+ , H_2^+ , and H_3^+ . The intention was to include a measurement using singly charged ^4He ions with H_2^+ as reference ion. However, due to the large deviation between my result and the accepted ^4He mass the result was discarded. The deviation was at the time blamed on poor statistics and,

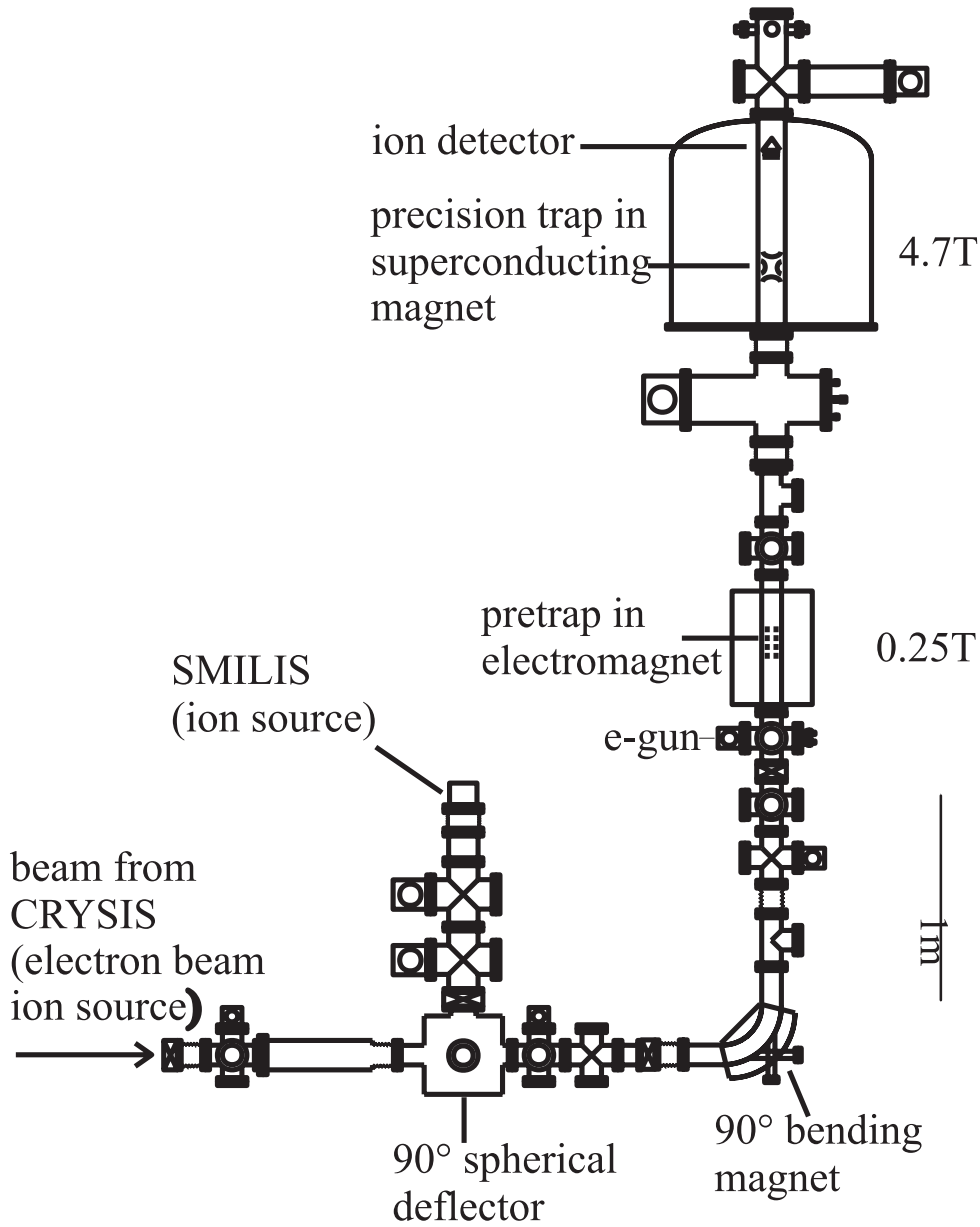


Figure 1.2: The SMILETRAP setup at the Manne Siegbahn, Stockholm University. The sketch shows the 90° charge state selection magnet, the electromagnet for the pretrap and the superconducting magnet housing the precision trap where the mass measurements are performed. The detector on top of the apparatus is used for the time-of-flight determination of the cyclotron frequency.

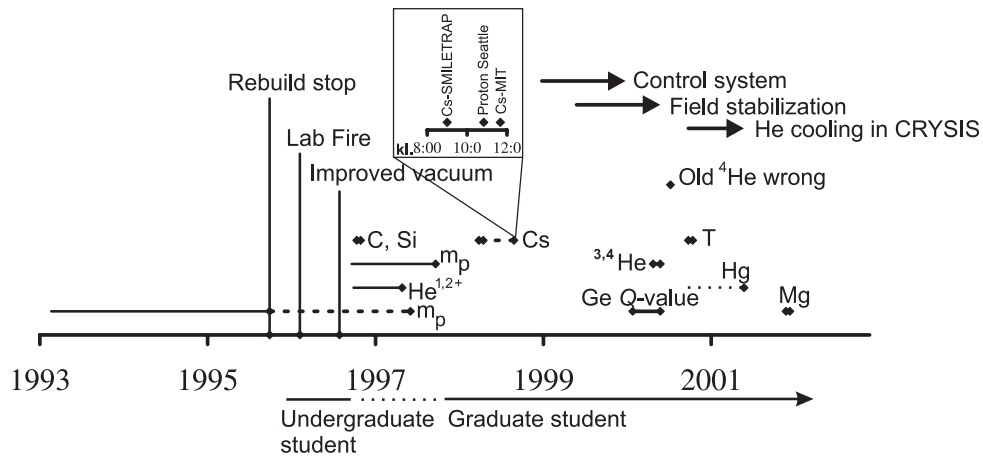


Figure 1.3: Important events from the time when the trap was moved to Stockholm. Interesting to note is the conference in 98 when three important mass measurements were presented in less than three hours.

as I felt it, on the poor handling of the equipment by an inexperienced student. Later the mass of 4He was re-measured. The mass value turned out to be in perfect agreement with my first result that was discarded. It was then correctly concluded that the accepted value of the 4He mass was wrong.

The improvements of SMILETRAP during my time as a graduate student are the following ones:

1. Improved technical performance and more efficient data taking
2. Improved mass accuracy
3. Measurements of atomic masses for solving current problems in fundamental physics

In figure 1.3 I have indicated some important events during the development of SMILETRAP and when I entered the project. My thesis is the 5'th that is based on construction, improving and using SMILETRAP for interesting experiments (Figure 1.4).

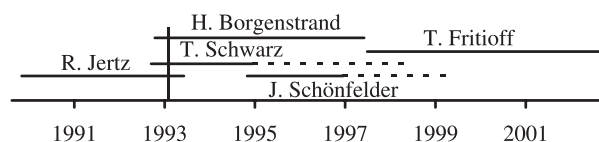


Figure 1.4: All five PhD. students that have based their work on SMILETRAP. In February 93 the trap was moved from Mainz to Stockholm indicated by the vertical line.

In the table below a summary of the most important mass determinations in which I have been involved in listed in order of increasing mass. An extensive description of the SMILETRAP mass spectrometer is described in Paper I, [6].

	Atom Mass in u	Published in or submitted to	Objective
1. ^1H	1.007 276 466 86(21) ¹	I. Bergström et al., <i>On the Masses of ^{28}Si and the Proton Determined in a Penning trap</i> . Accepted by Physica Scripta, May 2002	Accuracy test, only statistical uncertainty
2. ^3H	3.016 049 278 4(10)	I. Bergström et al., <i>On the Q-value of the Tritium Beta-decay and the Double Beta-decay of ^{76}Ge</i> . To appear in the Proceedings of Beyond the Desert 2002, Oulu, Finland, June 2002.	Related to efforts to find a finite value of $m(\nu)$
3. ^3He	3.016 029 323 5(28)	T. Fritioff et al., <i>A New Determination of the ^4He and ^3He masses</i> , Eur. Phys. J. D 15, 141-143, (2001)	Check of accepted mass
4. $Q(^3\text{H})$	18.588(3) keV	See also 2	Related to $m(\nu)$
5. ^4He	4.002 603 256 8(13)	Same as 3	Check of accepted mass
6. ^{20}Ne	19.992 440 185(14) ²	Same as 7	Accuracy test
7. ^{22}Ne	21.991 385 115(19)	T. Fritioff and G. Dyoussset, <i>Mass Determination of ^{22}Ne, ^{36}Ar and ^{86}Kr using Highly Charged Ions in a Penning Trap</i> , Submitted to Nuclear Physics A.	Improved masses. First test of injecting mass separated $1+$ ions into CRYISIS

¹Statistical uncertainty only. To be compared with the accepted value 1.007 276 466 89(13)

²Deviation from accepted value 0.46 ppb

	Atom Mass in u		Published in or submitted to	Objective
8.	^{24}Mg	23.985 041 687(17)	I. Bergström et al., <i>Accurate mass measurements of Hydrogen-like $^{24}\text{Mg}^{11+}$ and $^{26}\text{Mg}^{11+}$ ions in a Penning trap</i> , Submitted to Eur. Phys. J.	Improved masses. Mass accuracy requirements in g-factor measurement of bound electrons
9.	^{26}Mg	25.982 592 979(32)	Same as 8	
10.	^{28}Si	27.976 926 536(8) ³	Same as 1	Check of accuracy
11.	^{36}Ar	35.967 545 105(29)	Same as 7	Improved mass. Check of charge-consistency
12.	^{40}Ar	39.962 583 122(39) ⁴	Same as 7	Accuracy test
13.	^{76}Ge	75.921 402 758(96)	G. Douysset et al., <i>Determination of the ^{76}Ge Double Beta-decay Q-value</i> Phys. Rev. Lett. 86, 4259-4262, (2001)	Check of controversial Q-value
14.	^{76}Se	75.919 213 795(81)	Same as 13	
15.	Q(^{76}Ge)	2 039.006(50) keV	Same as 13	
16.	^{86}Kr	85.910 610 730(110)	Same as 7	First check of high q (29+) trap performance
17.	$^{133}\text{Cs}/^1\text{H}$	131.945 355 91(28)	C. Carlberg, T. Fritioff and I. Bergström, <i>Determination of the Ratio of the ^{133}Cs and Proton Masses Using Highly Charged Ions in a Penning Trap</i> , Phys. Rev. Lett. 83, 4506, (1999)	Contribution to a new way of determining α indep.of QED
18.	^{198}Hg	197.966 768 44(43)	T. Fritioff et al, <i>Shedding Light on the Mercury Mass Discrepancy by Weighing Hg52+ Ions in a Penning Trap</i> , Sub-mitted to Nuclear Physics A	Check of controversial Hg-mass values
19.	^{204}Hg	203.973 494 10(39)	same as 18	

³Deviation from accepted value 0.05ppb⁴Deviation from accepted value 0.03ppb

Chapter 2

My contribution to SMILETRAP improvements

The SMILETRAP (Stockholm Mainz Ion LEvitation TRAP) project was initiated in 1989 by I. Bergström (still today he is active on a daily basis). In order to get a quick start on the project the trap was designed, built and tested at the physics department at the Johannes-Gutenberg University in Mainz. After this period which lasted from 1990 to 1992 the trap was disassembled and moved to the Manne Siegbahn Laboratory in Stockholm. The objective was now to connect the trap to the ion source CRYISIS to make use of the highly charged ions produced ion this source. From 1990 to 2000 Conny Carlberg was responsible for the development of SMILETRAP and the graduate students that have worked with the trap. Four graduate thesis are based on experiments using SMILETRAP , by three German and one Swede; Roland Jertz 93 "*Direkte Bestimmung der Masse von ^{28}Si als Beitrag zur Neudefinition des Kilogramm*" [7], Tobias Schwarz 98 "*Hochpräzise Massenbestimmung hochgeladener Ionen mit SMILETRAP*" [8], Johannes Schönfelder 99 "*Hochpräzise Massenbestimmung von ^{28}Si und des Protons durch Penningfallenmassenspektrometrie an hochgeladenen Ionen mit SMILETRAP*" [9], and Håkan Borgenstrand 97 "*An attempt to measure the proton mass using a Penning trap and highly-charged ions*" [10].

During the time in Mainz Georg Bollen made a monumental contribution based on his experience which he had achieved during the first phase of the ISOLTRAP [11] project at CERN. Thereafter Bollen continued to have a large influence on SMILETRAP through discussions with our group in Stockholm and through the three German graduate students.

Thus the buildup process was a typical team-work from which I have benefitted. It is therefore justified to specify in detail my contributions to the development of SMILETRAP. This work would not have come as far as it did without the friendly and fruitful cooperation by the French physicist Guilhem Dousset, Post Doc. between 1999-2000.

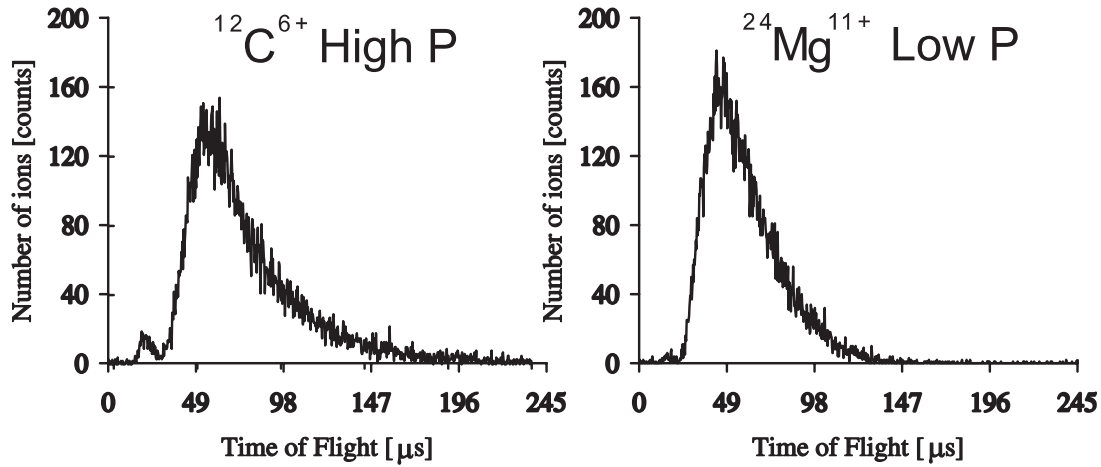


Figure 2.1: Time-of-flight (TOF) spectra before (left) and after (right) the vacuum improvements. In collisions between highly charged ions and the background gas (mainly H_2) protons and highly charged ion with charge state 1 and 2 lower than the original ion are created. These protons can be directly observed in the time-of-flight spectra as a small peak in front of the highly charged ions. The right plot is a $^{12}C^{6+}$ TOF spectra before the vacuum improvements and the left plot is a $^{24}Mg^{11+}$ spectra after the vacuum improvements. In these examples the proton peak corresponds to 2 % respectively 0.3 % of the total intensity.

2.1 Previous precision limitations

From the results presented in Håkan Borgenstrand's thesis [10] a lot was learned about the properties of the SMILETRAP Penning trap mass spectrometer including some weak features. Not only systematic uncertainties but also technical problems limited the precision of the mass measurements. A problem that was corrected for just before Borgenstrand's thesis was written was the relatively poor vacuum in the trap. It was estimated to be not better than a few times 10^{-10} mbar due to the limited conductance through the trap. The poor vacuum therefore limited the time during which highly charged ions could be stored inside the trap. The pressure was improved by adding pump capacity and by increasing or decreasing the conductance where it was necessary. This was done by adding NEG pumps (Non Evaporative Getter) close to the traps. The pretrap is surrounded by ~ 4 m of NEG equivalent to 4000 l/s pump speed. To decrease the gas load from the beam line to the precision trap a narrow tube was added above the pretrap. The precision trap itself was opened to increase the conductance. This was done by removing a lot of unnecessary construction material which increased the open area of the trap by more than a factor of 10 without changing the trap properties. As a result of these changes the vacuum is now $< 10^{-11}$ mbar. The improved vacuum can be directly observed from the time of flight plot (Figure 2.1).

The largest remaining problem was the disturbing drift and fluctuation in the magnetic field of the trap, which gave rise to a lack of reproducibility from run to run. On a few occasions there could be discrepancies as high as 5 ppb *i.e.* several times the statistical uncertainty. The drift in the magnetic field forced us to scan the cyclotron frequency over a larger window than necessary compared to the width of the resonance. On several occasions the resonance still drifted out of the window.

Another serious problem was the amount of time spent in transporting the beam from the ion source CRYISIS to the trap. If the beam "quality" and the amount of ions reaching the trap is not good enough CRYISIS and the beam line elements had to be reoptimized. Since the amount of allocated beam time is limited it is important to use it in an efficient way. The limited time for the measurements was, and still is, a problem that reduces the statistical uncertainty in the measurements. However, this limit has been pushed by several measures described in my thesis and in Paper I resulting in the use of ions with higher charge state and heavier mass than was before possible.

2.2 Technical improvements

Development of a new control system

From the start of SMILETRAP and until the spring of 1999 the control system was based on a 68030 controlled VME-bus with a graphical user interface on PC with Windows 3.11. Several problems with the old system led to the decision to replace it with a totally new control system. The old programs running on both the VME and the PC was written in C++ and since the knowledge of C programming was low among the SMILETRAP crew a LabVIEW based system was developed. From the users point of view the most important improvements with the new system can be summarized as:

- No computer crashes related to the control system
- Improved ability to correlate frequency drifts with ambient parameters like pressure, temperature and other quantities.
- Improved control over the ion transport/injection by adding new parameters for the two traps, several deflectors and other parameters.
- Information on important trap parameters like the trap potentials for both traps which now can be set and stored by the control system.

One of the advantages achieved with the new control system is a much lower number of crashes of the computer (zero crashes due to the control system). The frequent crashes of

the previous system resulted in loss of data and beam time. Since the time allocated for each experiment is limited by a users program this is indeed an important improvement.

Improved inter-trap optics

Among the hardware that has been replaced are the voltage power supplies which control the deflectors in the beam line in between the two traps. The advantage of this measure is that the deflectors now can be set differently for the reference ion and the ion species to be measured. This is in particular useful when the ions have different q/A values and during tests of the systematic effects when using different settings for the same ion species. With the new control system the optimization of the ion transport is more efficient which results in more captured ions and lower initial energies.

With the new control system it is possible to change the settings of all controlled electronic devices like voltages, ramp generators, pulse generators etc. within a time less than 2 seconds.

New trap electronics

The voltage power supplies for the different electrodes of the two traps have been replaced. The voltage of the different electrodes in the pretrap can now be set by the control system and they are more stable and less noisy than before. This is also true for the voltage power supplies which are now used to control the electrodes of the precision trap. The old supplies had a noise of at least 5 mV and an unknown repeatability and accuracy. The new supply (National Instruments NI 6704) has an average noise $< 50\mu$ V, an absolute accuracy of ± 1 mV and a relative accuracy $< 50\mu$ V. This corresponds to an improvement in the stability of the axial frequency from 120 Hz to ~ 1 Hz. The axial frequency is used to tune the E-field of the trap. Therefore, the more stable trap voltages is an important improvement of the trap.

Logging of ambient parameters

To correct for the influence of the measured cyclotron frequency from ambient parameters like air temperature and air pressure these parameters have to be accurately measured. Therefore, a barometer and several temperature probes were installed and monitored continously by a separate system. The barometer is a Tiltz HBA90 with a resolution of 0.1 mbar. The temperature probes that are now used are 4-wire PT100 probes together with a National Instrument DAQ4351 card. This measure makes it possible to measure

temperature changes to better than 0.01°C which is crucial for stabilizing the magnet field, the changes of which are due to the temperature dependent susceptibility of the trap and the surrounding construction materials.

2.3 Reduction of cyclotron frequency drifts

The improved precision that now can be achieved with SMILETRAP is to a large extent due to the stabilization of three quantities:

1. The trap temperature
2. The liquid Helium pressure
3. The frequency synthesizer

From fluctuations in both the room temperature and the air pressure one can observe a pronounced correlation in the measured cyclotron frequency. These fluctuations could sometimes be so large that it was impossible to accurately determine the cyclotron frequency. The cyclotron frequency of the ions could also drift out of the measured frequency window before the resonance was measured. When I started my PhD work both a pressure and a temperature stabilization existed, but they were not working properly. The improvements done on these two systems plus the stabilization of the frequency synthesizer is described below.

Trap temperature

The previous temperature regulating system in SMILETRAP originated from the time the trap was operated in Mainz. It consisted of a few pieces of plumbing tubes a recycled hair dryer (both the fan and heater) and an ordinary axial fan. The tubes connected the lower and upper opening between the bore of the magnet and the vacuum tube. The fans made the air slowly circulate around the tube and a type-E probe measured the temperature. A simple temperature regulator normally used to control the baking of the vacuum system was used to regulate the heating power of the hair dryer heater. The trap temperature was only stable to within 0.5°C with this system and the system sometimes failed to keep the temperature due to large room temperature fluctuations, $\pm 2^\circ\text{C}$. Therefore we started to investigate how it could be improved. As a test the magnet bore was totally sealed and Styrofoam isolation was placed on the vacuum tubes that are accessible below and above the magnet. The result was both disappointing and promising. The measured temperature and frequency were indeed correlated as shown in Figure 2.2. The fan was

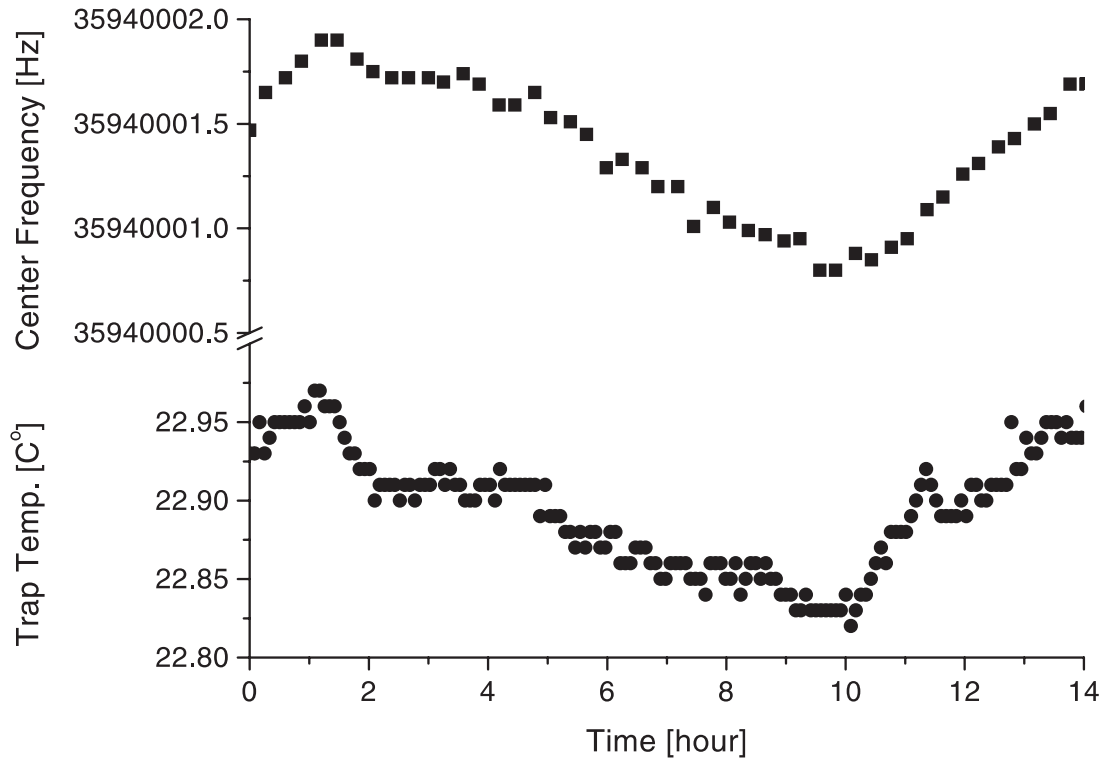


Figure 2.2: The upper curve shows the cyclotron resonance and the lower the trap temperature during the same time. As can be seen there is a strong correlation between the measured frequency and the temperature. Due to the time it takes to measure a resonance curve, in this example about 20 minutes, the fastest temperature variations are reduced.

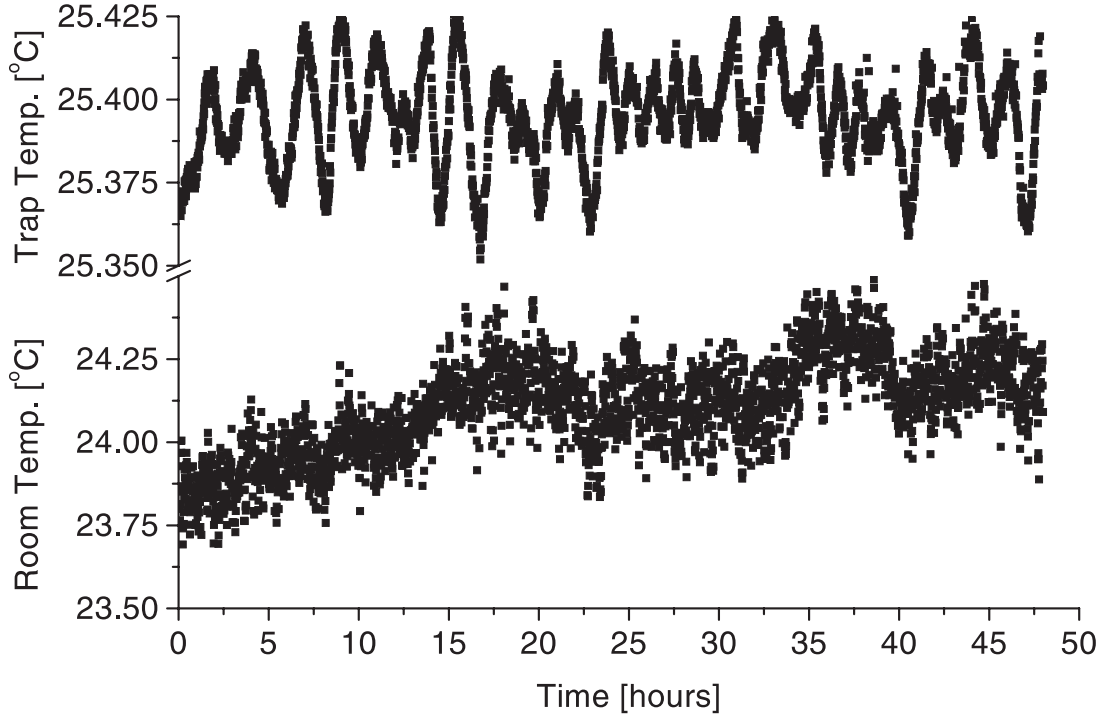


Figure 2.3: The trap temperature (upper curve) and room temperature (lower curve) measured each minute during 48 hours. Note that the upper scale for the trap temperature is 10 times larger than the lower room temperature scale. In this example when the room temperature is quite stable there is very little correlation between the two temperatures.

replaced with a stronger one that gave a faster response and additional isolation outside the tubes decreased the influence from the temperature of the laboratory air. The control unit was also replaced by one (FUG-3000) using a PT100 temperature probe that has a much better sensitivity and response time than the original device. In this way a much better regulation was reached if the room temperature is stable enough (Figure 2.3). The trap temperature is stable within $\pm 0.02^\circ\text{C}$ which corresponds to a resonance frequency stability of ± 0.2 Hz for an ion with $q/A \sim 0.5$. The B-field is only oscillating around a fixed mean value and the influence on the measured frequency ratio is less than 0.1 ppb. However, the regulation system introduces an oscillatory behavior of the trap temperature. By optimizing the parameters of the regulator this behavior has been minimized. A visual inspection or a FFT analysis of the temperature data shows that the remaining ± 0.2 Hz oscillation has a period of a little more than 2 hours which we have not managed to remove so far.

A justified question is why the regulation is done by air that is forced to circulate outside the trap instead of some more direct method. The simple answer is that other methods tested were not satisfactory enough. Without disassembling the whole trap it is not

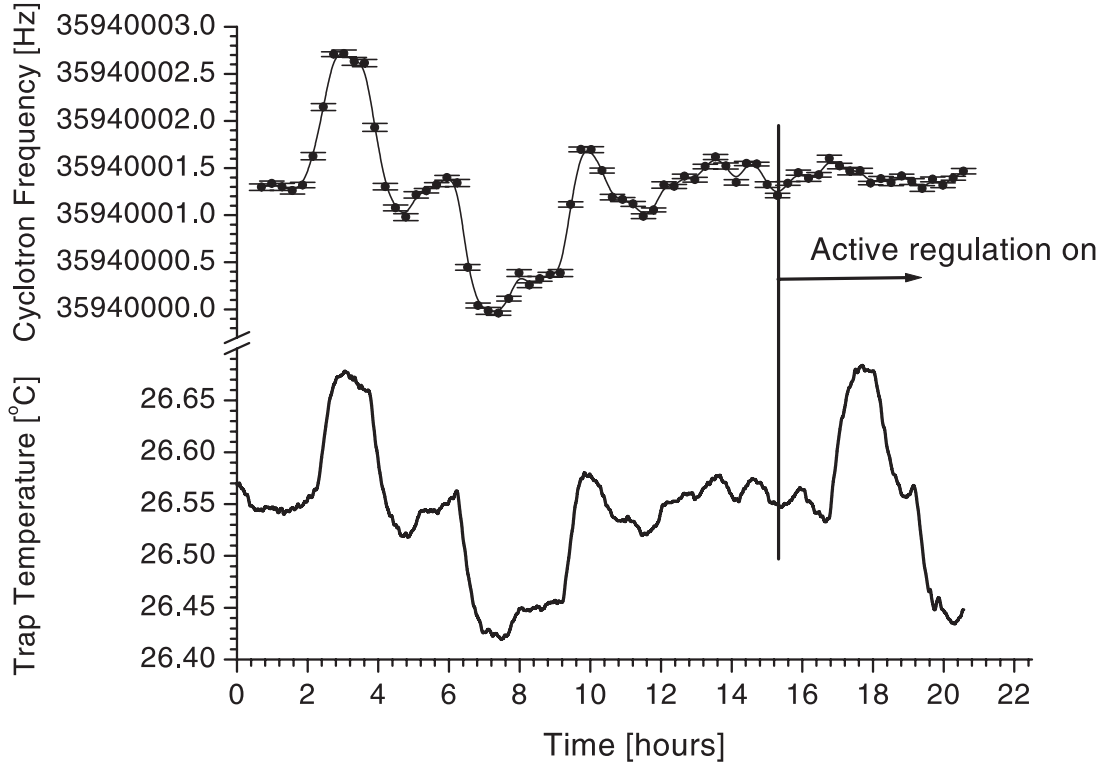


Figure 2.4: The upper curve shows the cyclotron resonance frequency using H_2 ions and the lower curve the trap temperature during the same time. Changing the set point of the regulation system induces jumps in the trap temperature of $\pm 0.1^\circ C$. After 15 hours the active regulation is turned on and even though the temperature is shifted by $\pm 0.1^\circ C$ which corresponds to a frequency shift of ± 2.2 Hz the cyclotron resonance frequency is stable.

possible to reach closer to the trap itself in order to implement a heater there. A test when the vacuum chamber that is sticking out from the magnet was directly heated was not working at all. This indicates that it is not only the trap itself that changes the B-field but also all the parts surrounding it, like the low susceptible stainless vacuum tube and the warm shim coils mounted inside the magnet bore just outside the vacuum tube in which the trap is located. The room temperature and therefore the B-field sometimes make drastic changes that makes the field unstable for a while which causes an oscillatory behavior in the regulation of the trap temperature.

Active feedback

To correct for these fluctuations that the temperature regulations fails to correct for an active feedback system was designed. A spare shim coil intended for correction of the B-

field in the z-direction is given a current that is proportional to a change in temperature relative to an arbitrary chosen reference temperature. The size of the correction current has been determined experimentally. The result is a system that drastically damps most of the remaining fluctuations of the trap temperature and the B-field (Figure 2.4).

Liquid He dewar pressure

By variations in the atmospheric pressure the pressure inside the liquid helium dewar is changed and therefore also the boiling point of the helium. Both the change in temperature and the change in the gas flow can cause field changes. This effect was the reason for the error in the He mass measured by VanDyck *et al.* [12]. When I started my thesis work G. Rouleau (Post Doc 92-93 and 95-97) designed and constructed a regulation system that was working electronically but still the pressure was not stable. In fact some minutes after the system reached the equilibrium point the pressure usually started to increase. The problem was due to leaking pressure transducers. As a matter of fact two identical transducers of the same type were tested and both were leaking.

These pressure transducers measured the pressure difference between a reference cavity and the LHe dewar. After the installation of a new type of pressure transducer with an internal reference cavity (TransMetrics P0202) and rebuilding the leaking He gas line from the dewar to the recovery system through the regulation valve the system now works perfectly. The only disturbance that can be observed is when there is a large load on the recovery line which can give a small pressure increase during about 5 minutes. It has been hard to measure the gain from this change in real numbers due to the sometimes long time constant of the system. It takes a long time before the whole bath of LHe changes its temperature. S. Brunner *et al.* [13] showed that in their magnet a pressure stability of ± 0.09 mbar resulted in a long term stability of 10^{-11} /h. The new SMILETRAP system is 1000 times more stable than what can be expected from natural fluctuations in the air pressure and as far as can be observed there is no pressure dependence in the magnetic field.

Frequency synthesizer

The frequency synthesizer that produces the RF-signal for the cyclotron excitation uses a crystal inside an oven achieve a stable 10 MHz signal that is used to create the output frequency. The synthesizer that we use had a guaranteed frequency stability of 10^{-9} per day when it was new. The fast switching between the measured ion and the reference ion that we use limits the problem of drifts but it may still cause broadening of the line width. Such a broadening makes it harder to trace other sources of drifts and broadenings. To make the frequency more stable a GPS (Global Positioning System) receiver was installed.

As well known this satellite system is usually used to accurately determine the position of various objects on earth, sea and air. The GPS receiver delivers a stable 10 MHz signal that is connected to our frequency synthesizer and locks the oscillator to this signal. In this way the synthesizer will deliver a frequency as stable as the reference signal from the GPS. The frequency stability reached by this method is better than 10^{-12} as long as the GPS receiver is in connection with at least 3 satellites (usually 7 or 8 satellites are within the range of the antenna at SMILETRAP). The frequency stability that the GPS receiver provides is sufficient as long as it takes more than a day to measure the frequency ratio to about the same order of magnitude as the stability or when using an excitation time shorter than 100 seconds (usually 1 s in SMILETRAP).

2.4 Improved interplay between CRYISIS and SMILETRAP

General properties of CRYISIS

A common problem during the experiments has been that although the beam intensity out from CRYISIS (Paper I, [14, 6]) has been satisfactory too few ions have neither reached nor been captured in the pretrap. The most likely explanation of this failure is due to the fact that the emittance of the ion beam has been larger than the acceptance of the pretrap. There is no equipment available close to CRYISIS to measure the emittance. Therefore it has not been possible to first optimize the beam close to CRYISIS and then transport the beam to SMILETRAP. The heavier the ions are and the higher the desired charge state and the harder it has been to trap a sufficient amount of ions. This is due the fact that the ions are not only ionized by the collisions with the electrons, as a side effect they are also heated. Since a high charge state requires a long storage time this effect will give hotter ions. The scientific program at SMILETRAP has in recent years asked for higher charge states. A lot has been learned over the past years on how the parameters in CRYISIS should be set to avoid ion heating and other unwanted features.

The ions in an EBI are axially confined by potentials on a set of electrodes (Figure 2.5) and radially by the space charge of the electron beam. The intensity of the electron beam also defines the maximum number of ion charges that can be stored and in the end ejected out from the source. A large electron current gives more ions but the radial energy spread increases as well and therefore fewer ions are trapped in the pretrap. The normal solution when there is a problem of capturing enough ions is to decrease the electron beam intensity and the CRYISIS trap depth. This gives less beam on the beam monitoring detectors close to CRYISIS but it usually gives a higher fraction of the ions transported through the 90° charge selecting magnet and consequently more ions are captured in the traps.

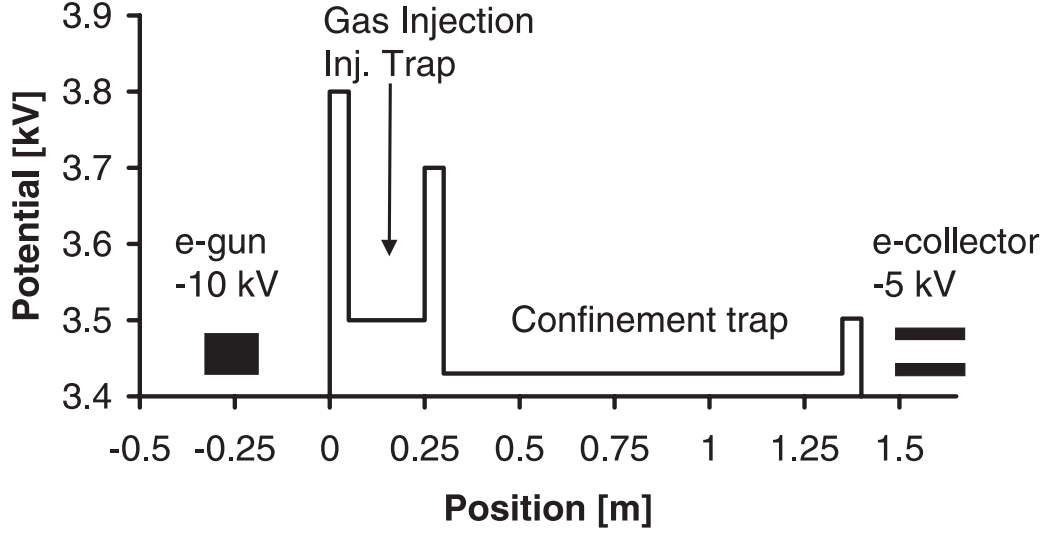


Figure 2.5: The potentials in CRYISIS that defines the smaller injection trap and the larger confinement trap used respectively for the cooling ions and charge breeding of the highly charged ions. The potential difference between the e-gun and the trap defines the energy of the electron beam, in this example 13.5 keV.

Ion cooling in CRYISIS

When the aim is, to measure the mass of highly charged ions, the lower electron beam intensity also decreases the maximum charge state that can be obtained. Therefore, this became a limitation and a new trick had to be introduced to increase the number of highly charged ions that can be trapped in SMILETRAP. On several occasions tests to cool the ions in CRYISIS by adding some other ions (by introducing a gas) have been performed. In the worst case the cooling ions took over and the intensity of the wanted species decreased. As an example Ar gas was injected while Cs ions were produced (Paper VII). The initial result was positive with signs of cooling but it was not possible to control the amount of Ar gas inside CRYISIS and after a short time most of the ions from CRYISIS were Ar ions. The real success came when He ions were used for cooling the highly charged ions. He gas was injected and ionized in a way that the He ions could pass through the confinement trap inside CRYISIS. Here the highly charged ions are stored during the charge breeding process. The previous problem that the cooling ions took over is avoided when:

1. The cooling ions are blocked during injection by the barrier between the gas inlet and the large confinement trap.
2. The cooling ions are injected with a higher axial energy than the highly charged ions.

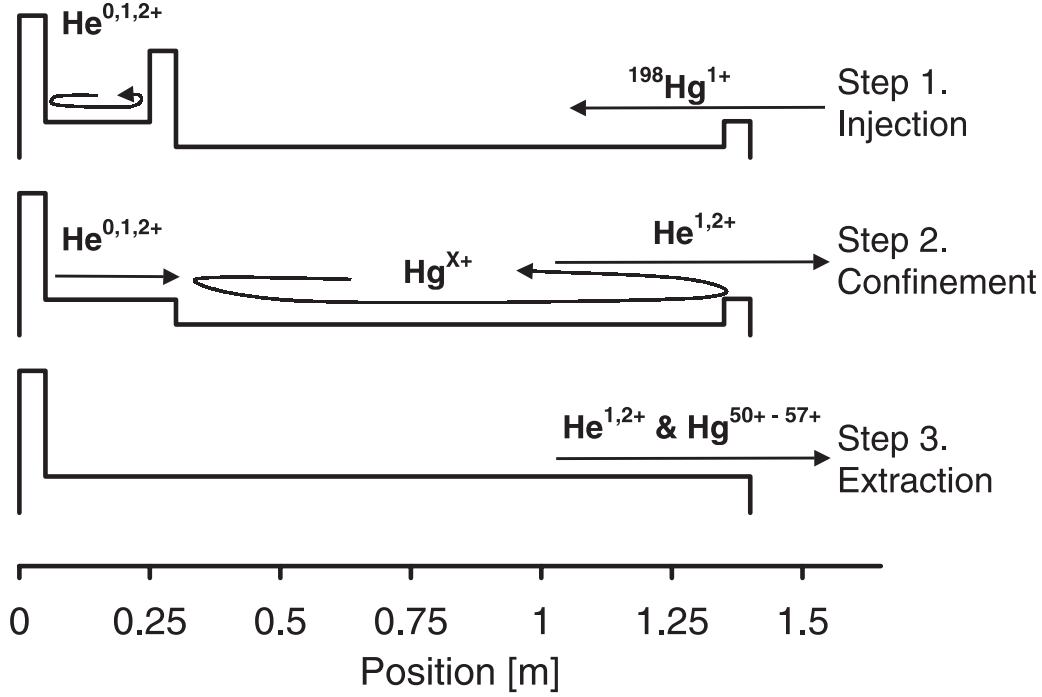


Figure 2.6: The different potentials in CRYISIS during the different stages of ion production with He cooling. The He gas producing the cooling ions are injected continuously to the left. (Step 1.) The cooling ions are blocked by a potential barrier inbetween the two trap sections while singly charged ions are injected from the ion source CHORDIS. (Step 2.) The He gas injected from the left is ionized by the electron beam and the He ions can freely move to the right through the cloud of trapped highly charged ions while the charge state is increased. (Step 3.) The mixture of He and highly charged ions is extracted and the cycle starts over again.

3. He ions are used for the cooling because:

- He gas is not trapped on the cold walls of CRYISIS and the optimum amount is therefore easier to control.
- He ions are not bound as strongly by the electron beam in the source as a more charged cooling ion would be.
- He has only two charge states that are easily distinguished from the highly charged ion spectra when reaching the pretrap.

The potentials on the different electrodes in CRYISIS are set as shown in Figure 2.5 and Figure 2.6. After injecting singly charged ions into CRYISIS in order to increase their charge state the cooling ions are injected into the confinement trap at the same potential as the barrier holding the highly charged ions inside the source. This will make the He ions very lightly bound, if bound at all. When they collide with the heavy ions which

are stored in the large trap they will be given some energy by collisions with the highly charged ions that will eject them out of CRYISIS. Since the barrier between the injection trap and the confinement trap is open during the whole storing procedure there will always be He ions in the trap to cool the heavy ions. But the barrier is closed during the injection of singly charged heavy ions and therefore the electron beam is empty and ready to trap these ions.

The result of the cooling is a more intense beam with less energy spread. In fact it is possible to use a much higher electron current and a deeper trap with cooling than what has been possible before. During a run without cooling the electron current was optimized to ~ 70 mA which gave a maximum number of trapped ions. With cooling it has been possible to use 130 mA and still trap more ions. The situation is similar for the trap depth which could be increased from 30 V to 70 V. With this method it has been possible to produce higher charge states than before, up to $^{238}\text{U}^{65+}$. In Figure 2.7 it is shown how the beam intensity increases when more cooling ions are applied by opening the He gas valve. The intensity drop for the highest charge states (low magnet current) for the setting with more gas is due to the fact that the beam energy changes slightly with cooling and therefore the beam line is not fully optimized. This is a problem during the whole beam optimization procedure and costs us a lot of time and patience.

It should finally be concluded that without the implementation of the cooling we would not have been able to perform the Hg mass measurements (Paper VIII). Though CRYISIS is not formally included in the SMILETRAP system I had to spend a lot of time to understand how it should be run to suit the SMILETRAP conditions, and to optimize properties which are not relevant for other users.

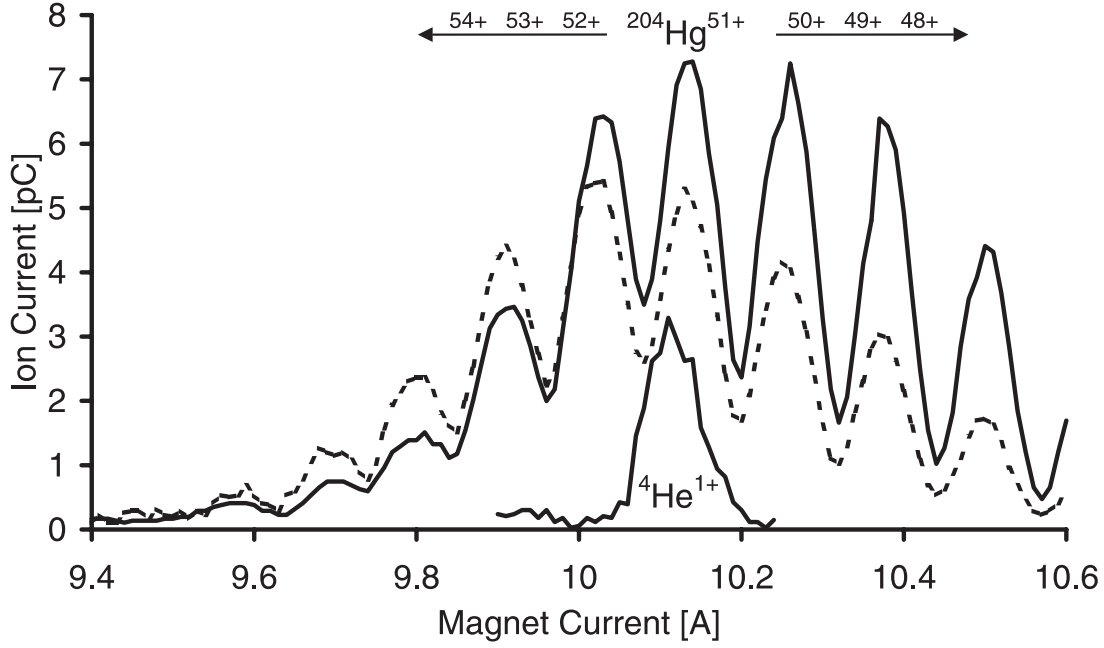


Figure 2.7: Charge state spectra of ^{204}Hg ions measured on a Faraday cup after the 90° magnet. The two charge spectra with many peaks are measured from two different settings of the He gas valve on CRYISIS. The solid curve corresponds to more He ions than the dashed curve, resulting in more efficient cooling and more ions. The total beam extracted from CRYISIS for the two settings was 1080 pC and 630 pC respectively. The lowest line with a single peak is due to $^4\text{He}^{1+}$ ions produced when no Hg ions are injected into CRYISIS. This mode is used to safely identify the different charge states.

Chapter 3

Post Scriptum comments to the results presented in Paper I-VIII

3.1 The masses of ^3H , ^3He , and ^4He

The measurements of these light isotopes had a long overture lasting for several years. The very first physics result after SMILETRAP was installed in Stockholm was an attempt to measure the proton mass using H_2^+ ions as a carrier for the proton mass and highly charged ions of ^{12}C , ^{14}N , ^{16}O , and ^{20}Ne as mass reference. The proton mass can be determined in this way by an accurate numerical relation between the mass of the proton and the H_2 molecule and the masses of the highly charged ions. The masses of these ions are namely known to an accuracy of about 0.1 ppb from the accurate mass measurements of N, O and Ne by the group of D. Pritchard at MIT [15, 16]. The p/ H_2 mass ratio is determined from accurate atomic and molecular data (Paper I [6]) at an uncertainty < 0.1 ppb. The proton mass derived from these measurements was presented in the thesis by H. Borgenstrand in 1997 [10].

Already at this stage several weak features in CRYISIS and SMILETRAP were noticed. The vacuum in the trap was not satisfactory which limited the charge state and time that the highly charged ions could be stored in the trap without charge exchange with the rest gas. More serious at this stage of SMILETRAP was the lack of reproducibility in the measurements. The proton mass determined from different runs using the same reference ion differed more than expected from the statistical uncertainty of the individual measurements. Therefore, the value of the proton mass presented in Borgenstrands thesis was assigned an uncertainty of 1.3 ppb compared to the statistical uncertainty of 0.3 ppb. The total uncertainty of the proton mass was also influenced by the the fact that a value of the proton mass using $^4\text{He}^+$ gave an unreasonably low proton mass.

At a conference in Ferrara 1997 Conny Carlberg reported a value of the proton mass [17] based on new measurements using highly charged ions of ^{12}C , ^{14}N , and ^{28}Si obtained after the vacuum improvement. To this data a couple of measurements using highly charged ^{20}Ne and ^{40}Ar ions were added. In all cases a consistent proton mass was obtained except when ^4He was used as a mass reference. The average proton mass excluding the ^4He result agreed well with the at that time accepted proton mass value [16] as well as with the present value [18] (Figure 3.1).

It has to be admitted that the strong belief that we had in the ^4He mass value reported by Van Dyck Jr. *et al.* disabled us for a long time. A systematic effect that could explain the deviation was desperately searched for but none was found. Therefore, it was finally concluded that the ^4He mass value from the Seattle group was wrong. After the B-field stabilization it was decided to return to ^4He and measure it's mass again. With the improved proton mass value [18] from the group of Van Dyck Jr. it was possible to turn the argument around to determine the ^4He mass. This was done in a q/A doublet measurement using H_2^+ ions as mass reference and the accurate relation between the proton mass and the H_2 mass. The new measurement confirmed the previous deviation from the accepted mass value at that time.

We were informed by Van Dyck Jr. that the discrepancy likely is due to a daily variation in the magnetic field in his spectrometer that was not known at the time of their measurements. The conclusion was that the mass obtained with SMILETRAP was more correct (Paper II [19]). Van Dyck Jr. *et al.* have remeasured the mass of ^4He using the new "Ultra-Precise" mass spectrometer [20, 18] and their last value is $4.002\,603\,254\,10(9)\,\text{u}$ [21]. This new value is in better agreement with the value in Paper II that deviates 5.5 sigma from the old Seattle value (Figure 3.2). The SMILETRAP value and the new Seattle value only deviate 2 sigma but the Seattle group achieved an uncertainty 14 times smaller. Since the group of Van Dyck Jr. also had measured the masses of ^3H and ^3He at the same time as the ^4He was measured it was decided to check these masses as well.

The difference between the atomic mass of ^3H and ^3He is the Q -value of the ^3H β -decay:

$$^3\text{H} \rightarrow ^3\text{He} + \beta + \nu \quad (3.1)$$

From measurements of $^3\text{H}^{1+}$ and $^3\text{He}^{2+}$ using H_2 ions as reference in both cases a Q -value of $18\,588(3)\,\text{eV}$ is obtained as compared to the Seattle value of $18\,590.1(1.7)\,\text{eV}$ [22, 23]. The relatively large uncertainty in our value is due to the q/A deviation between the singly charged tritium ions and the doubly charged helium ions. The tritium Q -value is expected to be improved to an uncertainty of $1\,\text{eV}$ by using $^3\text{He}^{1+}$ ions, which with $^3\text{H}^{1+}$ ions constitute a perfect q/A doublet measurement. The new so called KATRIN spectrometer planned in Karlsruhe will be capable of measuring the energy spectrum of the β electrons with a resolution 10 times higher than achieved in earlier spectrometers. It will be possible to set a lower limit on the neutrino mass of $0.3\,\text{eV}$ or to find a value in the region 2 to $0.3\,\text{eV}$. The neutrino mass would show up in the difference between

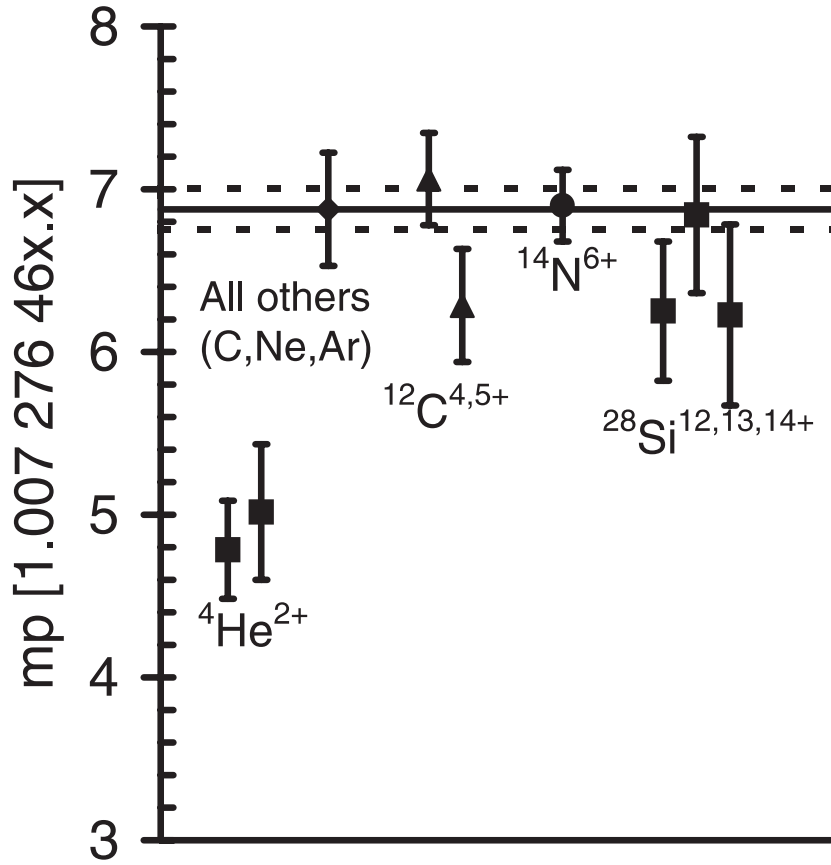


Figure 3.1: The mass of the proton determined by using different highly charged ions as mass reference. The solid line is the precise Seattle value [18], and the dashed lines indicates the uncertainty limits. The measured proton mass when the ${}^4\text{He}$ mass by the Seattle group is used as a mass reference clearly deviates, indicating either a systematic error in our procedure or a wrong accepted ${}^4\text{He}$ mass.

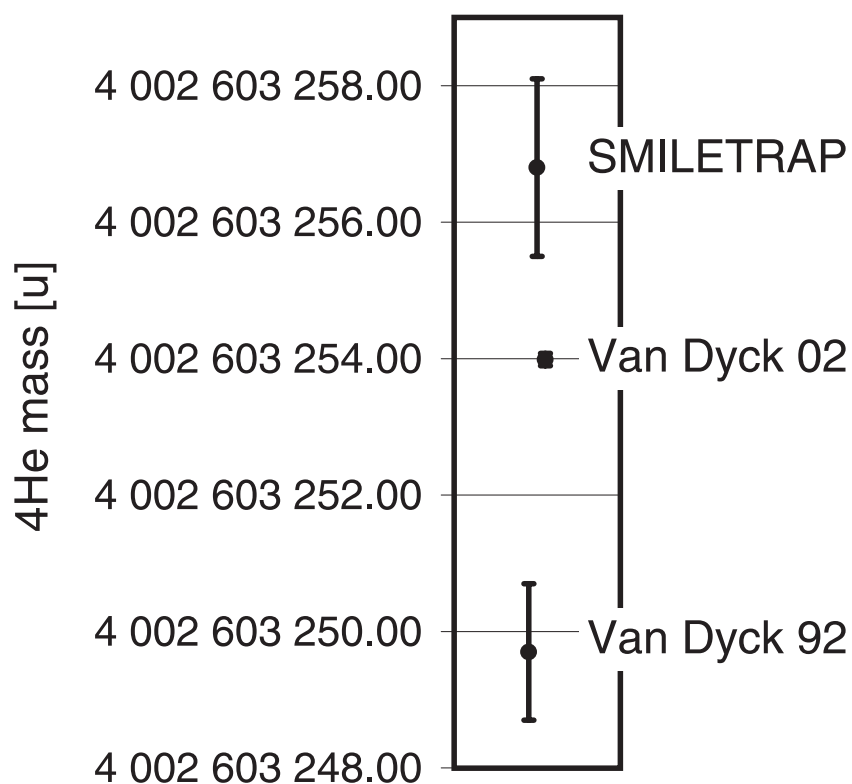


Figure 3.2: A comparison of ^4He mass measurements. The new Van Dyck Jr. value agrees much better with the SMILETRAP result. The new Seattle spectrometer has a much better stability and much better mass resolution than the old spectrometer.

the Q -value and the endpoint of the β -spectrum. Therefore there is a future need for a Q -value improved by at least a factor 5. It should be possible to reach such an improved Q -value in a new measurement using $^3\text{H}^{1+}$ $^3\text{He}^{1+}$ ions in SMILETRAP but the Seattle group with their new setup would provide an even more accurate Q -value.

The tritium run was also a technical achievement in the sense that only a very small amount of tritium gas was used and very few parts of the equipment exposed to tritium and contaminated. To have enough gas a bottle containing 4 ml (10 Ci) tritium was supplied by RC Tritec. The tritium is absorbed in depleted uranium inside the bottle which therefore can be evacuated to remove the ^3He gas that is created through the decay of the tritium during the transport and storage. A small amount of gas was released into an evacuated volume of about 5 ml until a pressure of 400 mbar ($\sim 1/2$ atm) was reached by heating the uranium bed to $\sim 370^\circ\text{C}$. After the run the valve between the bottle with uranium and the small volume was again opened and the remaining tritium was absorbed back into the uranium bed. The amount of tritium gas used in CRYISIS was only ~ 0.14 ml *i.e.* 0.35 Ci.

During the summer of 2002 CRYISIS was opened for maintenance and it was concluded that the amount of radioactivity was not very high and easily decontaminated. It is interesting to notice that such a small amount of gas as 0.14 ml could be used in this ion source during one run week. The efficiency is not so impressive. From the measured beam intensity it can be estimated that this amount of gas correspond to an ionization efficiency of roughly 0.1%. Moreover, the overall efficiency from gas injection in CRYISIS to measured ion intensity in the trap is only 3×10^{-11} . This low efficiency is mostly due to the difference in the length of the ion beam pulse and the length of the pretrap and the fact that we through a selection procedure only trap one ion as an average each second .

3.2 The masses of ^{20}Ne , ^{22}Ne , ^{36}Ar , ^{40}Ar , and ^{86}Kr

It has to be admitted that the mass measurements of ^{22}Ne , ^{36}Ar , and ^{86}Kr were mainly done as tests of the performance of SMILETRAP. However, later mass determinations of ^{24}Mg and ^{26}Mg emphasized the need for accurate masses in the determination of the electron g-factor of bound electrons in hydrogen like ions. To qualify in such measurements the ion must have an even-even nucleus and these rare gas elements quoted above are such atoms. The masses of the ions in these measurements have to be known to 1 ppb or less. Therefore both ^{22}Ne and ^{36}Ar which are even-even nucleus could be used in these measurements since their mass now is known to about 1 ppb from these measurements. The measurement of the ^{36}Ar mass was the first accurate test using several charge states of the same ion species. During the same run week 13, 14, 15, and 16+ ions were measured with a derived ^{36}Ar mass that agrees within <1 ppb.

The Neon measurements were the first ones performed with the CHORDIS ion source connected to the isotope separator in order to test on-line mass separation. Previous tests with ion injection were done with other ion sources but their stability was poor. To successfully inject ions into CRYISIS one needs a stable beam and stable ion optics. The previous tests failed partly due to the fact that the ion sources had a tendency to change position resulting in a change of the ion path to CRYISIS which caused a very unstable beam of highly charged ions.

^{20}Ne and ^{40}Ar were selected as suitable test cases due to the fact that the MIT group had measured the mass of these isotopes with a high accuracy [15]. The masses of both Ne isotopes were measured with 10+, 9+ and 8+ ions but the 8+ data were unfortunately lost due to a problem with the old control system. These data would have added information on the q/A dependence and decrease the final statistical uncertainty in the measured masses. This failure showed again the vulnerability of the old system, and our previous lack of knowledge how to cure it.

$^{86}\text{Kr}^{29+}$ was used in a first test of the possibilities to handle very high charge states in SMILETRAP. The measurement resulted in an improved mass value [24] that was further improved by a factor of 10 after stabilising the B-field. At this level of accuracy the value of electron binding energy becomes so important that we had to use 26+ ions for which this energy has been calculated with a low enough uncertainty [25, 26].

3.3 Reevaluation of the ^{28}Si data

After our new ^4He mass measurement and the confirmation by Van Dyck Jr. *et al.* the mass uncertainty of some of the previous measurements were reconsidered. The Si mass presented in the thesis of J. Schönfelder [9] is 27.976 926 531(8)(28). The large systematic uncertainty was partly due to worries about the ^4He mass deviation. A reevaluation of the data decreased the total uncertainty by a factor of 3. The mass of the proton was calculated by comparing the cyclotron frequencies of H_2^+ and highly charged ions of ^{12}C , ^{14}N , ^{20}Ne , ^{28}Si , and ^{40}Ar . Through the good agreement of the weighted average of all these measurements with the accurate proton mass of the Seattle group [18] it can be concluded that the total systematical uncertainty is < 0.35 ppb (Table 3.1).

This way of experimentally determining the systematical error was not possible before due to the large uncertainty of the accepted proton mass. Data due to $q/A=1/2$ ions were excluded in the analysis because of the risk of impurities from CRYISIS. Furthermore $^{12}\text{C}^{5+}$ ions were compared to $^{28}\text{Si}^{12+}$ and $^{28}\text{Si}^{13+}$ since their q/A values are close. In this way a ^{28}Si mass of 27.976 926 536(8) u is obtained. This is in perfect agreement with the value measured by the MIT group [15], 27.976 926 532 4(20) u. The two mass values are both measured in Penning traps but using different charge states of the ions and different

Table 3.1: Comparison between our proton mass values (statistical uncertainty) and the accurately determined proton mass by the Seattle group.

	Proton mass [u]	
Seattle value	1.007 276 466 89(13)	Total uncertainty
All SMILETRAP data	1.007 276 466 74(16)	statistical uncertainty
Exculding $q/A=0.5$ data	1.007 276 466 84(21)	statistical uncertainty

methods to determine the cyclotron frequency resonance. For a new definition of the kilogram using a perfect silicon sphere [27] an uncertainty in the ^{28}Si mass of 1 ppb is required.

3.4 The ^{76}Ge - ^{76}Se double β -decay Q -value

The Q -value of the ^{76}Ge is indispensable in the search for neutrino-less double beta decay. In the decay mode allowed by the standard model for weak interactions:

$$^{76}\text{Ge} \rightarrow ^{76}\text{Se} + 2\beta + 2\nu \quad (3.2)$$

the electrons and the neutrinos share the energy and a continuous β -spectrum is observed. Although allowed, the decay is very rare with a half life of $1.55 \pm 0.2 \times 10^{21}$ years [28]. However, the decay mode:

$$^{76}\text{Ge} \rightarrow ^{76}\text{Se} + 2\beta, \quad (3.3)$$

is not allowed by the standard model since it would occur with a double lepton number violation. The position in the spectrum where one should look for a peak due to a neutrino-less decay is exactly given by the Q -value *i.e.* the mass difference between ^{76}Ge and ^{76}Se .

At a nuclear physics conference in Florida 1993 I. Bergström presented the SMILETRAP project and the possibility to measure atomic masses with an uncertainty of 10^{-9} . He was then asked by F. Avignone, who at the conference presented the latest results from the ^{76}Ge experiment in the Homestake mine [29], to measure the Q -value of ^{76}Ge double β -decay. A Q -value measurement had been carried out twice by the MANITOBA [30] group with different, and seemingly conflicting values [31, 32].

It was not until the spring of 2000, however, that the Q -value could be determined using highly charged ions in SMILETRAP. The reason for the delay was twofold. First there were experimental problems in the production of the ions and second there was a large uncertainty in the electron binding energies that has to be known accurately in order to get accurate atomic masses (Paper I [6]). The latter problem was solved by the very

precise calculations performed by E. Lindroth and P. Indelicato [25, 26]. However, several tests had to be done to produce Ge and Se ions, among them gas injection into CRYISIS with enriched GeH_4 gas. The gas is both explosive and has a freezing point higher than the temperature of the gas transfer line inside CRYISIS. Therefore, the gas condensed and formed ice plugs in the feed line. Later, after losing all gas in the bottle by a handling mistake a new approach was taken. But it took several years to reach a final solution. Ge ions were finally produced by xenon sputtering from slices of an old Ge detector inserted in the CHORDIS ion source. The Se ions were produced in CHORDIS from Se vapor by heating metallic Se in the ion source oven. In this way the measurements improved the mass uncertainties in the two atomic masses by a factor of 17 and, which is of more interest, the Q -value by a factor of 7 (Figure 3.3). The new result confirmed the later of the two MANITOBA measurements. The reason for the deviation between their two values came evidently from the influence of contaminating ions which they could not control during the first measurement.

The Q -value presented in Paper VI has recently been used in the analysis of the data collected in the Heidelberg-Moscow experiment located in the Gran-Sasso laboratory in Italy. This experiment has been running for the last 10 years using five enriched (86%) ^{76}Ge -detectors. In a recent article by H.V. Klapdor-Kleingrothaus *et al.* [34] it is suggested that the forbidden double beta decay has been observed (Figure 3.4). The obtained half life of the neutrino-less 2β -decay as derived from the measurement is $T_{1/2}^{0\nu} = (0.8 - 18.3) \times 10^{25}$ years. The authors also present an effective value for the neutrino rest mass of 0.11-0.56 eV (95% c.l.). For the analysis of the data from four of the detectors they emphasize that the accuracy of the Q -value presented in Paper VI is of decisive importance [35]. Important is also the fact that they can decide by pulse shape discrimination whether an event in the detector is localized *i.e.* originates from charged particles. These events are confined within a few mm^3 in the detector, as compared to multiple scattered γ -events from γ -rays which occupy a larger volume in the trap. In this way it is possible to discriminate 80% of the γ -events and significantly reduce the background in the β spectrum. The new result on ^{76}Ge double β -decay, though very interesting, has a very low confidence level (about 2-3 σ) and the final conclusion whether the neutrino is a Majorana or a Dirac particle has to await new experiments. The Heidelberg-Moscow group plans to build a new experiment that will use in its final stage 240 detectors of enriched germanium immersed in tons of liquid nitrogen. This detector, modestly called GENIUS, will have a background level 1000 times lower than the present setup and will hopefully be able to answer the question of neutrino less β -decay within a year of operation.

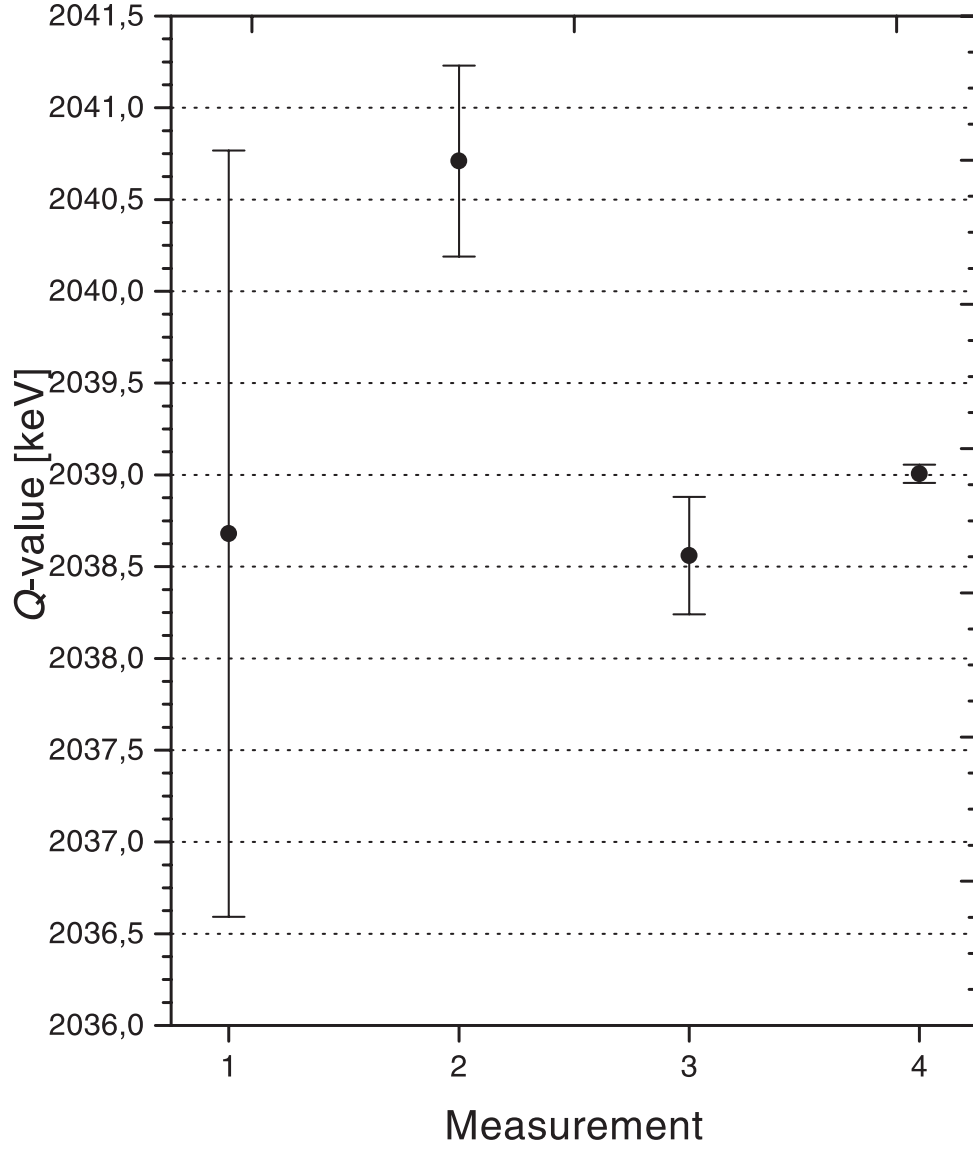


Figure 3.3: ^{76}Ge double beta decay Q -value. 1: Deduced from Audi *et al.* (1995) [16], 2 : Ellis *et al.* (1985) [31], 3 : Hykawy *et al.* (1991) [32], 4 : SMILETRAP 2001 Paper VI [33]. As can be seen the later of the two MANITOBA measurements (nr. 3) is in good agreement with the SMILETRAP result, although our value has a 7 times improved uncertainty.

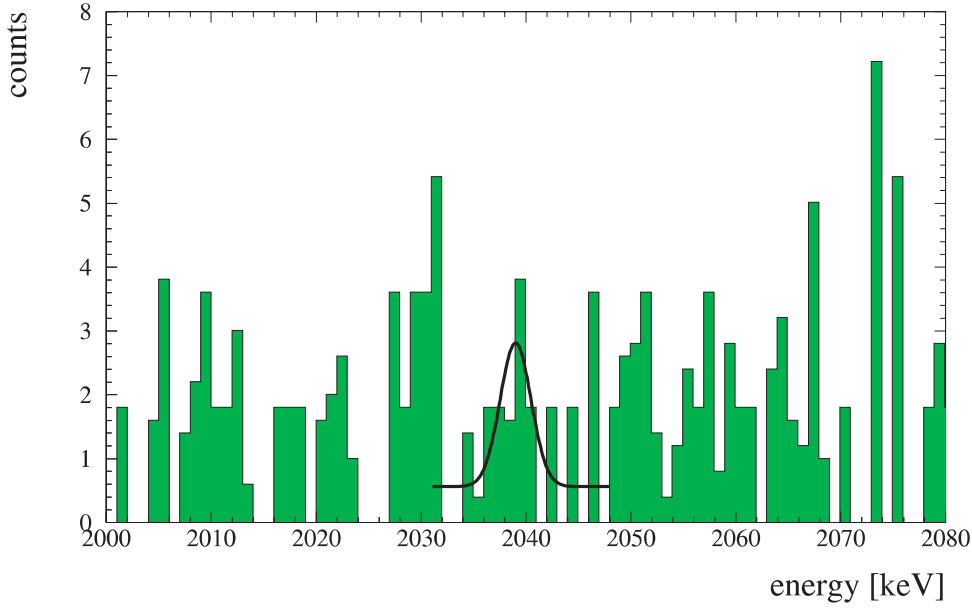


Figure 3.4: Evidence of neutrino less double beta decay in ^{76}Ge . $T_{1/2}^{0\nu} = (0.8 - 18.3) \times 10^{25}$ years with a the neutrino rest mass of 0.11-0.56 eV (95% c.l.). In the evaluation of this data our Q -value of 2 039.006(50) keV was essential. Courtesy of H.V. Klapdor-Kleingrothaus.

3.5 The ^{133}Cs and proton mass ratio

The present value of the fine structure constant is to a large part defined through the g-2 value of the free electron measured by Van Dyck Jr. *et al.* [3] and the QED calculations by Kinoshita [36]. There are obvious limitations in this procedure of deriving α and therefore new methods not as dependent on QED are of interest. The equation:

$$\alpha^2 = \left(\frac{2R_\infty}{c} \right) \left(\frac{h}{m_e} \right) \quad (3.4)$$

can be re-written as:

$$\alpha^2 = \left(\frac{2R_\infty}{c} \right) \left(\frac{h}{m_{Cs}} \right) \left(\frac{m_{Cs}}{m_p} \right) \left(\frac{m_p}{m_e} \right). \quad (3.5)$$

The Rydberg constant, R_∞ , and the mass ratio m_p/m_e are known to an uncertainty of 0.8×10^{-11} and 2.1×10^{-9} respectively [37, 38]. Since the uncertainty in the mass ratio between the cesium atom and the proton used to be 23 ppb we were asked by Barry Taylor at NIST to improve this value. The mass ratio was measured in April 1998 at an uncertainty of 2 ppb which at that time seemed good enough (Paper VII [39]). The result was shortly after confirmed and improved at MIT by Bradley *et al.* [40] (Figure 1.3) by measuring the mass of ^{133}Cs which was published in the same issue of PRL as our result.

Table 3.2: Comparison of α^{-1} values obtained by different groups (see [38]).

Primary source	α^{-1}	Relative uncertainty	Method
Γ_{90}	137.035 987 1(43)	32	Gyromagnetic moment of shielded proton
$\Delta\nu_{Mu}$	137.035 995 2(79)	57	muonium hyperfine splitting
$g - 2$	137.035 987 1(43)	3.8	g-2 of the free electron and <i>QED</i> calculations
R_k	137.036 003 0(27)	20	von Klitzing constant, quantized Hall resistance
h/m_n	137.036 008 4(33)	24	Planck constant / neutron mass
CODATA	137.035 999 72(50)	3.6	Average of values above
h/m_{Cs}	137.036 000 3(10)	7.4	Planck constant / Cesium mass

The m_{Cs}/m_p value was then again improved by using the new value of the proton mass by Van Dyck Jr. *et al.*[18]. What is important is that the two results agree within the required accuracy of a few ppb and are obtained in two quite different ways increasing the reliability.

Steven Chu, Nobel laureate in 1997, has measured h/m_{Cs} at an uncertainty (7.3 ppb) that results in a α^{-1} value with an uncertainty of 7.4 ppb. The fine structure constant obtained by this technique is compared to the best values from five other methods and the recommended CODATA value are compared in Figure 3.5 and table 3.2. As can be seen the value of Chu *et al.* agrees quite well with the CODATA value. The size of the uncertainties in the different ratios of equation 3.5 can be seen below:

$$\alpha^2 = \underbrace{\left(\frac{2R_\infty}{c}\right)}_{0.8 \times 10^{-11} \text{ ppb}} \underbrace{\left(\frac{h}{m_{Cs}}\right)}_{7.3 \text{ ppb}} \underbrace{\left(\frac{m_{Cs}}{m_p}\right)}_{\substack{0.2 \text{ ppb}^1 \\ 2 \text{ ppb}^2}} \underbrace{\left(\frac{m_p}{m_e}\right)}_{\substack{2.1 \text{ ppb}^3 \\ 0.7 \text{ ppb}^4}} \quad \begin{array}{l} 1. \text{ MIT} \\ 2. \text{ SMILETRAP} \\ 3. \text{ Seattle} \\ 4. \text{ Beier } et al. \end{array} \quad (3.6)$$

Chu now hopes that the uncertainty in their measurement of h/m_{Cs} can be improved from 7.4 ppb to 2.9 ppb by further improving their data evaluation [41]. This would give a α^{-1} value with an uncertainty of 3.1 ppb which is less than the value 3.8 ppb obtained from the g-2 measurement but without the input of *QED* calculations in that value. With a new setup Chu believes that it will possible to reach a value even below 1 ppb in the h/m_{Cs} ratio [42]. By using equation 3.5 and such a low value of h/m_{Cs} would give α^{-1} with an uncertainty 4-5 times lower than the value from the g-2 measurement which until recently was one of the best known atomic constants (1S-2S transition frequency is known to 1.8×10^{-14} [43]). A smaller uncertainty in fine structure constant can be obtained by using the very precise value of the electron mass by T. Beier *et al.* [44].

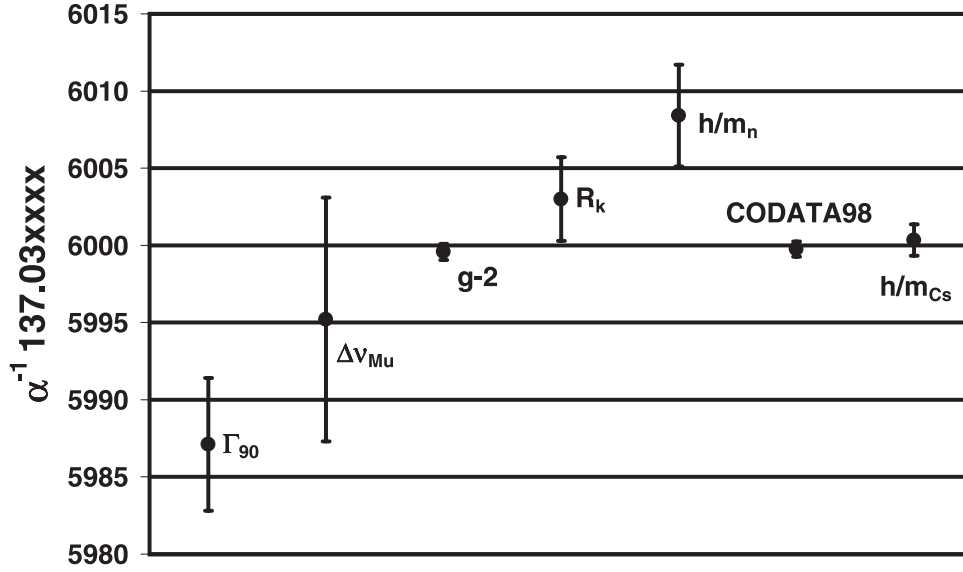


Figure 3.5: The most accurate values of the fine structure constant from five different methods in order of increasing value of α^{-1} [38]. Also shown is the recommended CODATA98 value that is almost only based on the $g-2$ value and QED calculations. To the right is the latest value from the Cs recoil measurements which is expected to be improved by a factor of 7 and thus lead to a more accurate value of α^{-1} than the value based on $g-2$.

With their electron mass value the mass ratio between m_p/m_e can be determined with an uncertainty of 0.8 ppb. This electron mass is derived from the accurate measurement of the bound electron g -factor in $^{12}\text{C}^{5+}$ and QED calculations. Compared to the fine structure constant derived from the $g-2$ measurement the value derived from this value would practically be independent of QED .

As is mentioned above, the MIT value of the ratio m_{Cs}/m_p is based on their measurements of m_{Cs} and the proton mass determined by the Seattle group. In Paper VII we presented a value that does not depend on the absolute mass of the proton. Since H_2 is always used as the reference in a SMILETRAP measurement (directly or as an intermediate mass reference) and the mass of H_2 is directly related to the mass of the proton (Paper I,[6]), one obtains the relation:

$$\frac{m_{Cs}}{m_p} = \left(\frac{m_{Cs}}{m_{H_2}} \right) \left(\frac{m_{H_2}}{m_p} \right) = \frac{m_{Cs}}{m_{H_2}} \times 2.000\,544\,600\,49(6) \quad (3.7)$$

Even better would be to directly compare the cyclotron frequencies of H_3^+ and $^{133}\text{Cs}^{45+}$ since the mass ratio between the proton and the H_3^+ molecule can be very accurately calculated (Paper I,[6]).

$$\frac{m_{Cs}}{m_p} = \left(\frac{m_{Cs}}{m_{H_3}} \right) \left(\frac{m_{H_3}}{m_p} \right) = \frac{m_{Cs}}{m_{H_3}} \times 3.001\,089\,196\,45(<9) \quad (3.8)$$

Cs^{45+} ions (Ne-like) can be produced in CRYISIS and would be a perfect mass doublet with H_3^+ ions which can be produced in the SMILIS ion source. The ions from this ion source uses a movable electrostatic deflector to steer the ions into the mass selecting 90° magnet before the pretrap. By increasing the number of scans measured before the ion species are switched it would be possible to directly compare the cyclotron frequencies of these two ion species.

3.6 The ^{198}Hg and ^{204}Hg masses

In 1980 all stable isotopes of mercury was measured by the MANITOBA group [45]. The mass difference between the isotopes was in good agreement with the values published in the mass tables obtained by a mass extrapolation method, while the atomic masses themselves differed significantly. In the latest mass evaluation the deviation between the extrapolated and directly measured values is 15 keV [46] with an uncertainty of 3 keV. In the 1993 edition of the mass tables [47] Audi and Wapstra expressed this mass dilemma as follows:

We do not feel happy about this situation and think that a re-measurement of the Hg masses combined with that of lighter elements ($Z = 73-77$), is the single most desirable experiment concerning masses near the line of β -stability.

The MANITOBA group indeed tried to re-measure the mass of the Hg isotopes. However, there were evidently troubles with their mass spectrometer. They got a difference between the masses of Hg and a lighter atom [48] that seemed to agree with that in the last mass evaluation by Audi and Wapstra [49]. In addition the estimated error in the new mass determinations was so large that they did not come near of solving the problem. Thus it was claimed that the new Hg mass values were preliminary and final values were never published. There were thus justified doubts whether the 1980 MANITOBA measurements were correct. It was therefore for a long time a need for more accurate mass measurements of a few Mercury isotopes. Due to experimental difficulties it was not before June of 2001 that the atomic masses of ^{198}Hg and ^{204}Hg could be measured during the last two days of two consecutive run weeks (Paper VIII). The mass values for the two measured isotopes agree with those measured by the MANITOBA group however with an uncertainty 3 times lower (table 3.3 and Figure 3.6).

From the experimental point the measurement was a success after several trials in vain, including a couple of unsuccessful run weeks with lead ions. The key to the success of the Hg measurements was the cooling of the highly charged ions with helium ions during the charge breeding in CRYISIS (see section 2.4). The mercury measurements were done with the heaviest ions and the highest charge state that have been used in SMILETRAP.

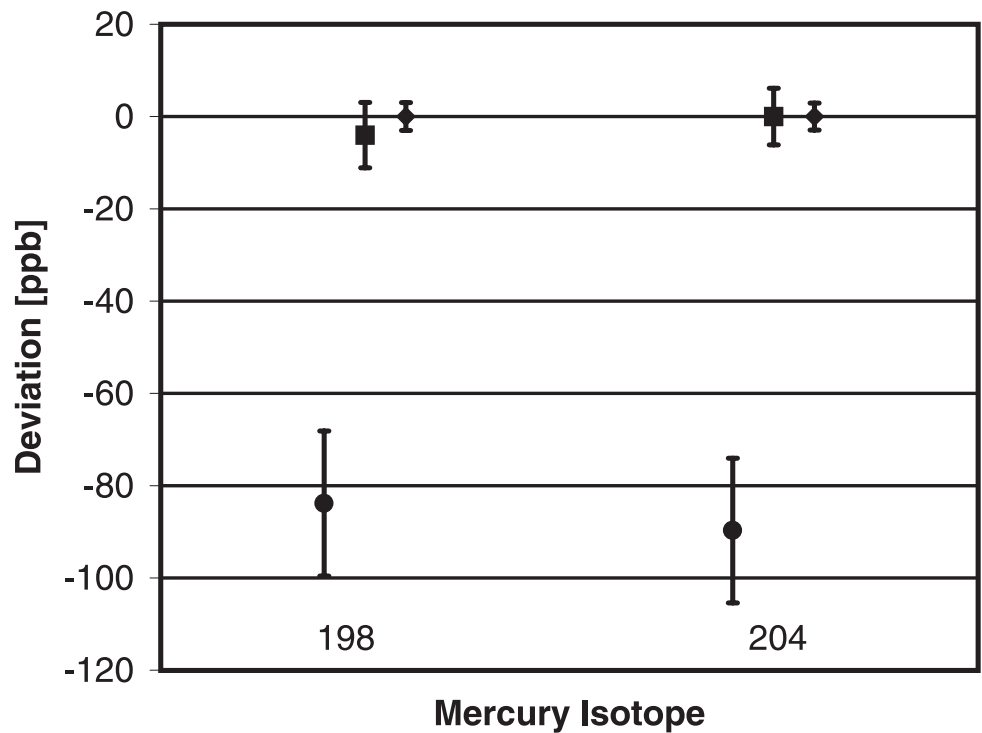


Figure 3.6: Comparison between our results and the previous mass determinations of the two isotopes ^{198}Hg and ^{204}Hg . From left to right in each group the values reported by Audi and Wapstra, the Manitoba group, and the Stockholm group are indicated. The direct mass measurements by the Stockholm group and the Manitoba group agree perfectly with each other and there is evidently something wrong with the Hg-mass values obtained by extrapolation from lighter and heavier atoms.

Table 3.3: Comparison of mass values of ^{198}Hg and ^{204}Hg obtained by three different groups.

	Mass of ^{198}Hg [u]	Mass of ^{204}Hg [u]
Audi&Wapstra	197.966 751 80(311)	203.973 475 90(320)
MANITOBA	197.966 767 60(141)	203.973 494 20(125)
SMILETRAP	197.966 768 44(43)	203.973 494 10(39)

The question still remains, why the mass extrapolation does not agree with the directly measured mass values. The answer requires accurate mass measurements of isotopes both heavier and lighter than Hg. Most likely, according to Wapstra [48], the error in the extrapolation procedure is due to wrong masses of heavy atoms. In the lower mass region there are some mass chains that also could be wrong and explain the deviation in the extrapolation from the lighter mass region. Wapstra therefore asked us to measure any of the masses of ^{235}U , ^{238}U or ^{232}Th . Lighter masses would probably also help solving the problem but new accurate mass values of the heaviest atoms should have priority.

In order to prepare for a mass measurement of the heaviest atoms a sputter target of enriched ($\sim 2\%$) uranium has been tested in the isotope separator. During this test run, ions with a charge state around $65+$ were extracted from CRYISIS at intensities enough to be used in SMILETRAP. Encouraged by this test a sputter target with 14 g of Th was prepared for the run week in February 2002. By using Th instead of U there would be less contamination of radioactivity in the isotope separator (Th has only one isotope and is less radioactive than ^{235}U). Unfortunately CRYISIS did not function in a stable manner during the run and after more than a week of tests CRYISIS was stopped. During the summer of 2002 CRYISIS has gone through a thorough maintainance, including a realigning of the optics and an upgrade of both the control system and the electronics. Hopefully this will lead to a more stable source. Therefore, it is planed to return to the measurement in the high mass region in the early part of 2003.

Chapter 4

Future improvements

Present limitations

It is of course of no use to drastically decrease the statistical uncertainty if the mass determinations still are hampered by some large systematic effects. The systematic effects that are present in SMILETRAP and their typical uncertainty are listed below.

- Reference mass 0.14 ppb using H_2 ions as reference, 0 ppb using $^{12}\text{C}^{q+}$
- Electron binding energies ~ 0.10 ppb
- Relativistic mass, usually from 0.10 to 0.20 ppb (can be as large as ~ 1 ppb). The uncertainty has somewhat arbitrarily been assumed to be 1/2 of the correction.
- Ion number dependence ~ 0.10 ppb
- q/A asymmetry, from 0.0 to 1 ppb
- Contaminate ions ~ 0.10 ppb
- Magnet field drift < 0.10 ppb
- B-field inhomogeneities and ion overlap¹ < 0.10 ppb

The present ability of SMILETRAP to measure masses with an uncertainty close to 10^{-10} using q/A doublets is partly due to the large number of scans used in a measurement. Instead of measuring one cold ion centered in the trap like the Seattle and MIT groups

¹The difference in B-field if the measured ion and the reference ion occupy different volumes

do we measure a large number of ions. The result in a typical measurement is based on more than 1000 scans with 21 frequency steps each scan with 1 or 2 new ions in the trap. Events with 0 or >2 ions are discarded in the data analysis. A normal measurement corresponds to about 20 000 ions of each species spread out over a measuring time of at least 20 hours. Each new ion is placed randomly in a rather small volume, $\sim 4 \text{ mm}^3$, in the center of the trap. Radially this volume is limited to 1 mm in diameter by the size of the entrance apperture of the trap. Axially the volume is limited by a boil off procedure that decreases the axial amplitude of the ions to $\sim 2.5 \text{ mm}$. This procedure averages out most of the imperfections and drifts in the E and B fields.

From the specifications of the magnet this would correspond to $\Delta B/B = 0.4 \times 10^{-10}$ in the field occupied by the ions. This will not be a problem as long as the excitation time is less than about 0.5 minute but other obstacles presently limit the excitation times to values not much longer than 1 s.

Two of the limitations that now prevents a higher accuracy are the limit in linewidth of the cyclotron resonance and the initial absolute energy as well as the energy spread. The initial energy depends on how the ions are injected into the magnetic field of the trap. Ideally the ions should follow the field lines into the trap without any gain in radial energy.

4.1 Cyclotron resonance linewidth limitations

In a few measurements excitation times longer than one second have been used. However, in the last years the result have been very unsatisfactory the reason being that the measured line width has been larger than expected (Figure 4.1). Before the last improvements of the B-field an excitation time up to 4-5 second was tested with a measured line width close to the expected (Figure 4.2). These tests appear to have been done under extremely stable conditions outside our control and were not reproducible. The reason for increased linewidth is most likely due to secondary effects of the stabilization of the B-field (section 2.3). The temperature regulation typically results in a temperature oscillation $\pm 0.02^\circ\text{C}$ which corresponds to a 10 times larger frequency oscillation, for an ion with $q/A=0.5$ (Figure 2.3). This oscillation is damped by the active stabilization but apparently not enough, or perhaps too much. The effect that this instability in the B-field has on the FWHM of the cyclotron resonance is shown in Figure 4.3 for different field oscillations. It is not only the FWHM that changes due to the B-field oscillation. The shape of the resonance is also changed. Instead of a resonance close to a $\frac{\sin x}{x}$ shape a more Gaussian-like shape is observed Figure 4.4. This is not the only reason that the resonance measured in SMILETRAP always has a more Gaussian-like shape. Due to the initial spread in energy and that the excitation is not fully converting an initial pure magnetron motion into a

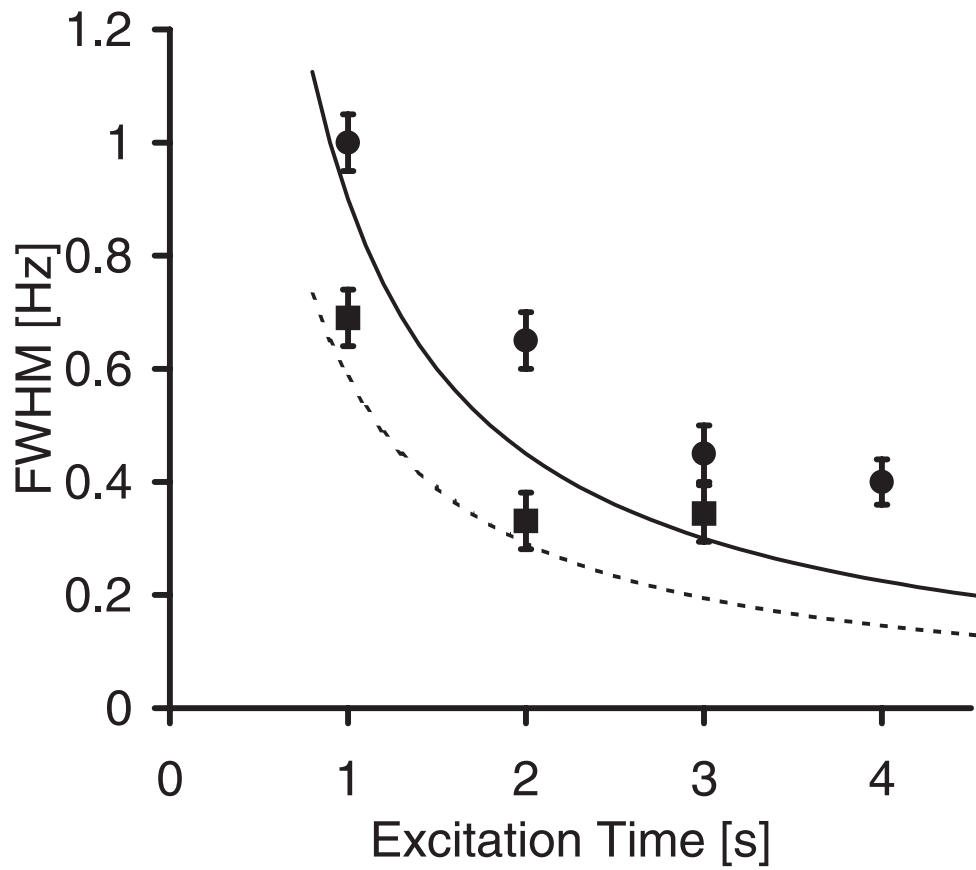


Figure 4.1: Measured FWHM for different excitation times. Circles are measured using the normal excitation scheme the solid line is the expected theoretical linewidth. The squares are the width of the central peak measured using a Ramsey excitation scheme the dashed line is the corresponding linewidth, see section 4.2.

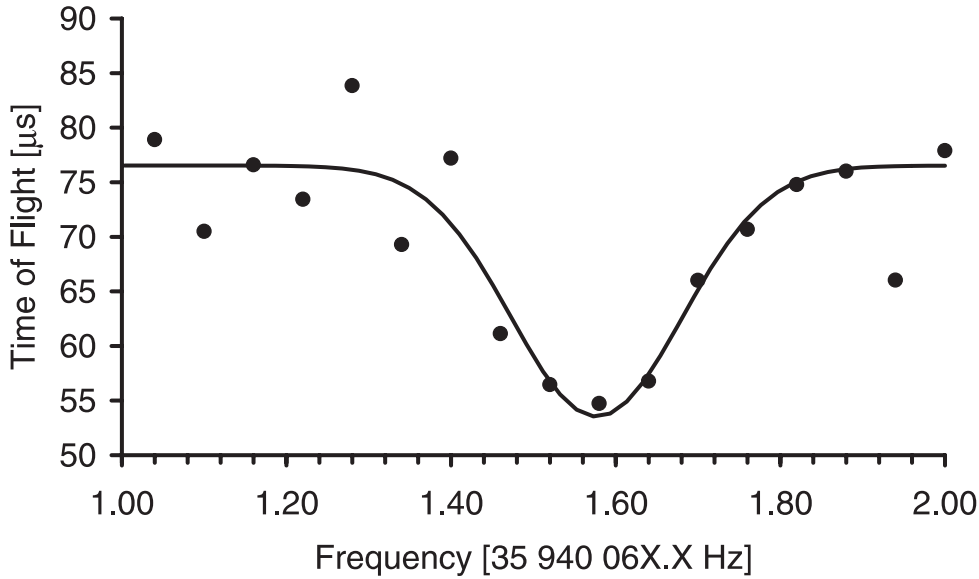


Figure 4.2: A H_2^+ cyclotron resonance using 4 s excitation time without B-field stabilization. The FWHM is 0.28 ± 0.04 Hz which is not too far from the expected value compared to more recent test with B-field stabilization giving $\text{FWHM} \sim 0.4$ Hz. Thus the B-field stabilization is associated with an oscillation causing a line width broadening which presently limits the excitation time to about 2 s.

pure cyclotron motion results in a line shape that is different from the ideal quadrupole resonance.

It will be possible to increase the excitation time by further improvements of the B-field stabilizations by adjusting the magnitude and the time constant of the active feedback. With the more narrow resonance thus achieved it might also be possible to detect inhomogeneities in the B-field and to tune it further with the shim coils.

However, when the excitation time is increased a new problem will appear. The amplitude of the exciting RF-field must be decreased. The problem is that the amplitude of the excitation field is already small. At an excitation time of 3 s the amplitude of the applied RF-field at the trap is in the order of a few mV which is the same level as the white noise from the combined electronics of the trap. If the amplitude is not decreased the relativistic mass increase will become too large which will both shift the center of the resonance and distort its shape. A solution to this problem may be to use Ramsey excitation, as discussed below.

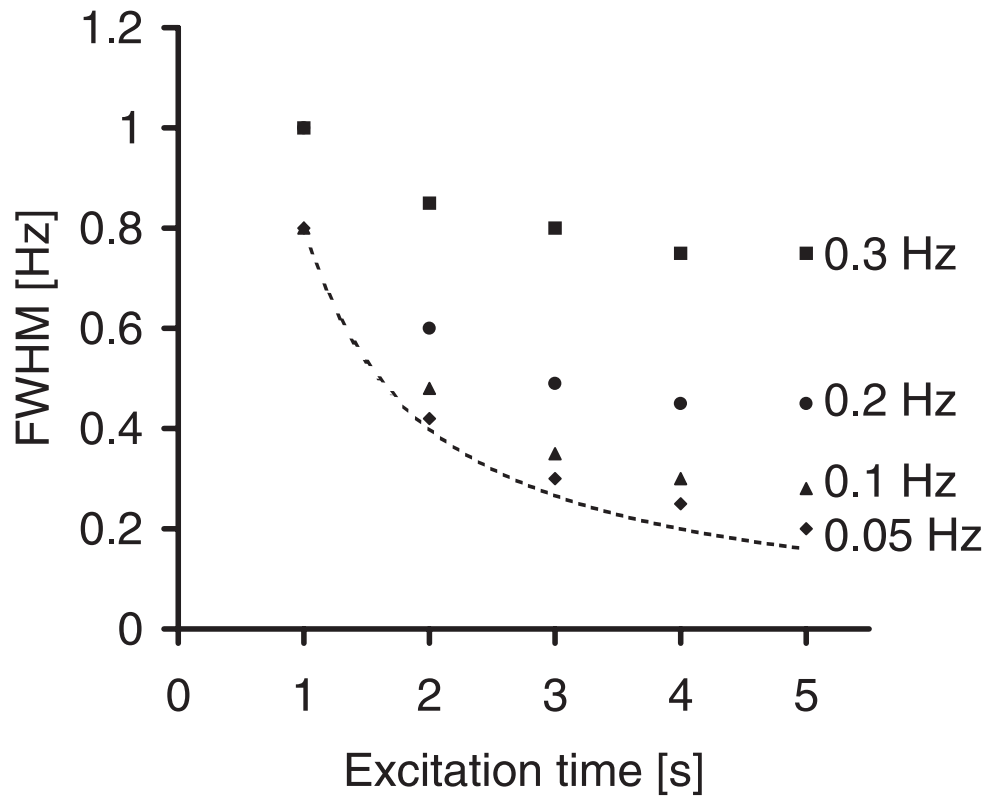


Figure 4.3: Simulated data showing the effect on the FWHM from four different B-field oscillations in Hz as compared to the expected line width the dashed line; 0.1 Hz corresponds roughly to 0.01°C for $q/A=0.5$ ions. Compare this plot with the experimental data in Figure 4.1.

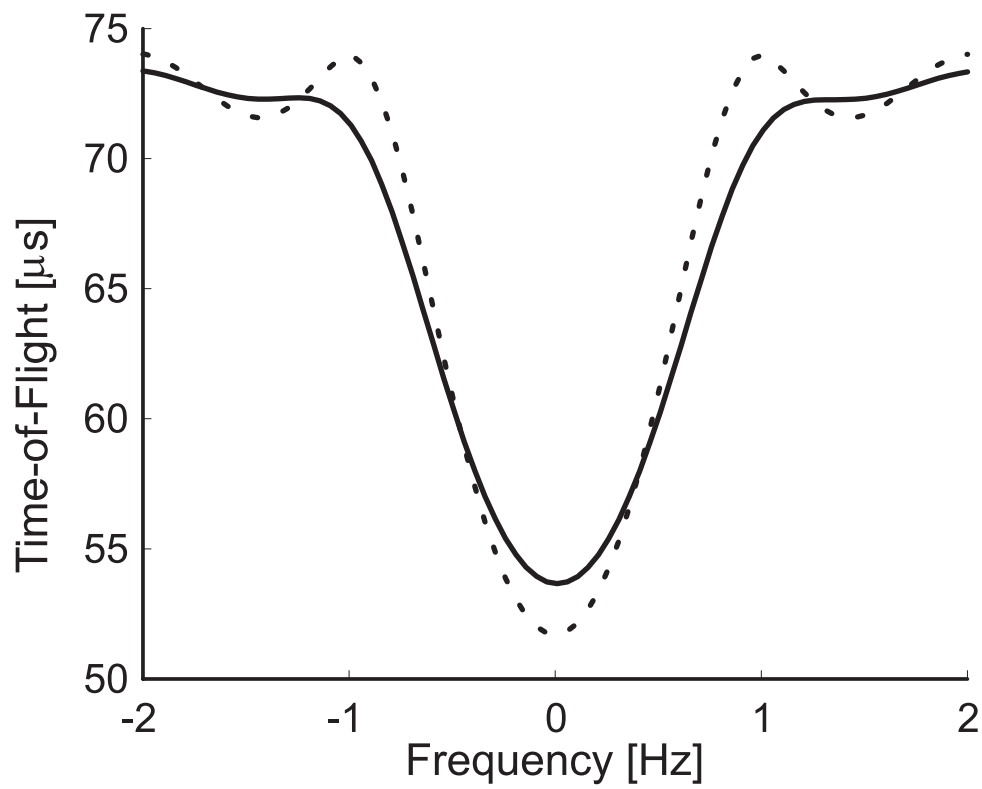


Figure 4.4: The shape of the cyclotron resonance changes when the B-field is unstable. The dashed curve is the expected line shape which becomes the solid curve with a slight oscillation in the field. As seen the line shape approaches a Gaussian.

4.2 Cyclotron resonance measurement using Ramsey excitation

Instead of exciting the cyclotron motion with one long RF-pulse as is normally done it is possible to use the Ramsey excitation scheme first developed for molecular beam resonance measurements [50, 51]. The principle of this technique is to excite the ions with two or more short RF pulses with longer interval between them. Instead of observing one single resonance so called Ramsey fringes are then observed. Several advantages are obtained by this method. The width of the central peak will be narrower than the line width from a normal one pulse resonance (with two pulses 60%). In order to achieve the same final energy the amplitude has to be increased, more the shorter the pulses are. This would solve the problem that the amplitude of the excitation field goes to zero as the excitation time is increased. It also has the experimental advantage that the correct excitation amplitude can be determined when using a short excitation time and then only the length of the interval between the pulses are increased. It might even be possible to use the Ramsey technique to detect field inhomogeneities by using pulses that are phase shifted. If the phase of the RF fields are shifted $\pi/2$ the maxima and minima change place under the condition that the field is homogeneous .

The Ramsey technique was tested in SMILETRAP at an early stage but since the field was very unstable and the fringes are close to each other the resonance fringes became quite ugly. But after stabilizing the field it was checked if this measure could solve the problem using the small excitation amplitudes with three equidistant pulses. In this way the first, third etc. side bands are removed. This procedure has been checked with H_2 , $^{16}\text{O}^{8+}$ and $^{76}\text{Se}^{25+}$ ions leading to promising results. As an example a scheme with three 100 ms long excitation pulses separated by 350 ms (*i.e.* 1 s total cycle time) has been tested. This results in a central peak that is narrower (FWHM \sim 0.7 Hz) Three well separated peaks are observed in the chosen frequency window (Figure 4.5). To make use of the Ramsey fringes this method requires a slightly broader frequency window than the normal excitation scheme and therefore fewer frequency scans are finished during an equal time. The loss in statistics in each step of the resonance is small compared to the gain in the determination of the center of the cyclotron resonance. By fitting the theoretical line shape for dipole excitation the center of the measured resonance curve is determined. In the measurement a resonance using a single excitation pulse was also measured simultaneously. The two different techniques were measured under exactly the same length of time which means that the normal mode completed more scans since more steps are used in Ramsey mode. When comparing the two data sets the cyclotron resonance is determined 1.6 times more accurately using the Ramsey technique. It is worth mentioning that during the tests with Ramsey excitation we have noticed that the determination of the resonance center is improved more than expected from the line width decrease if only a narrow frequency window is scanned around the central peak and using

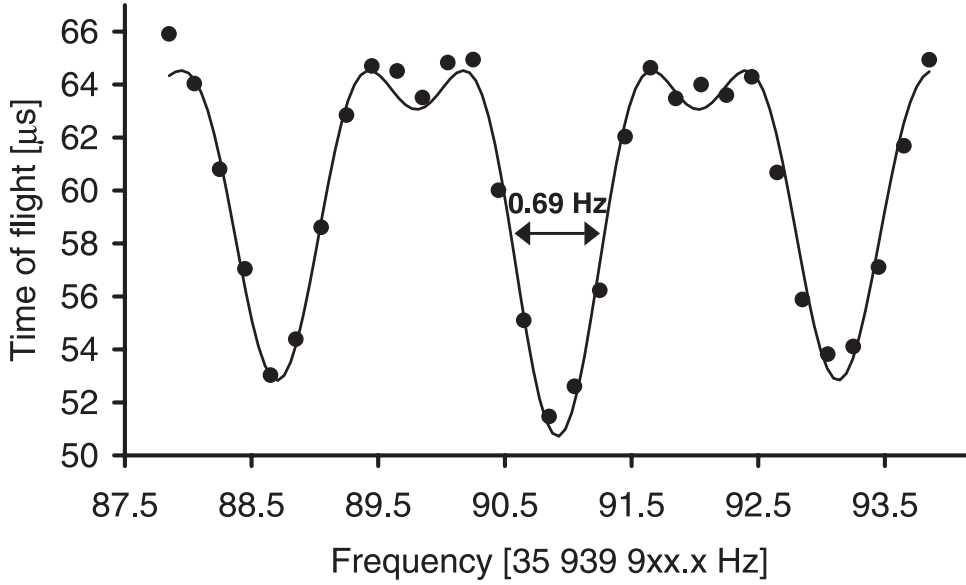


Figure 4.5: Three pulse Ramsey excitation test using H_2 ions. The total time of the excitation cycle is 1 s using three 100 ms long pulses. The center is determined to an accuracy of 2.2×10^{-10} .

the theoretical dipole shape for the fitting.

Due to the initial spread in the energy of the ions the baseline of the resonance is shifted down a little which decreases the depth of the resonance compared to the normal resonance. Ions with less initial energy could help in two ways, firstly the depth of the resonance would be enhanced and secondly the relativistic energy correction might be decreased.

Before using these techniques routinely, several tests have to be performed to detect any additional systematic error. The puzzling fact that the dipole shape so perfectly fits the fringe pattern as was already pointed out by Bollen *et al.* in [52] should also be understood.

4.3 Ion energies

As has been mentioned above the initial energies (0-1 eV) are slightly limiting the resolution of the measured resonance. No cooling technique is presently used in SMILETRAP. The coldest ions are rather selected in several steps (Paper I). The easiest way to decrease the initial energies would be to capture colder ions or to cool them in the pretrap. A smaller energy spread in the pretrap would be possible to conserve during the transport to the precision trap. The new pretrap electronics made it finally possible to test boil

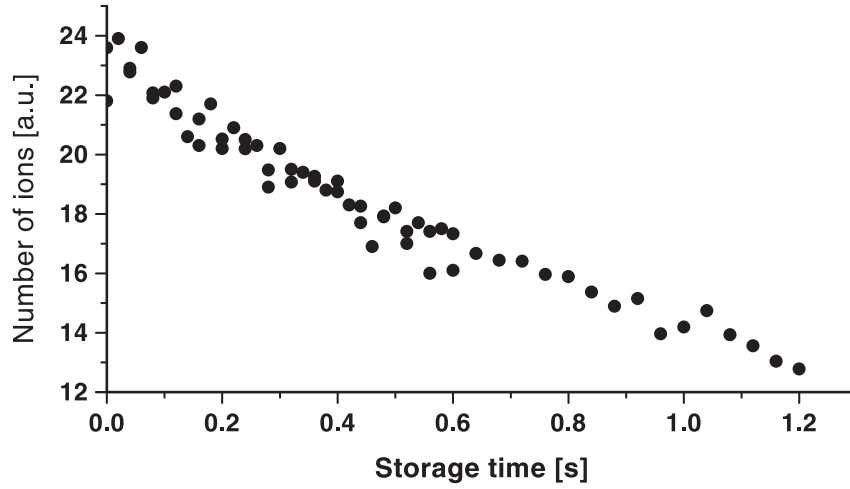


Figure 4.6: The number of stored $^{24}\text{Mg}^{11+}$ ions in the pretrap as a function of storage time.

off in the pretrap but without the decrease in axial energy spread that is observed in the precision trap.

It has to be admitted that the present pretrap has its limitations. For example it is not possible with the present setup to mechanically align the trap relative to the magnetic field direction. The storage lifetime of the ions in the pretrap is not at all impressive $T_{1/2} \simeq 1$ s (Figure 4.6). The lifetime is not limited by charge exchange due to poor vacuum but due to the weak field possibly combined with a misalignment. When the trap potential becomes too large compared to the B-field strength the trap becomes unstable due to the coupling between the different ion motions as was shown by G. Werths group [53]. This instability is further enhanced by misalignment and imperfections in the trapping fields. The short lifetime limits the possibilities to implement some cooling technique. For future measurements a new conventional magnet has been bought with a maximum magnetic field of 1.1 T as compared to 0.25 T with the present magnet. The stronger field and a new trap design that allows aligning of the E and B-fields will increase the lifetime in the pretrap and the possibilities to here cool the ions.

4.4 Determination and reduction of the q/A -effect

The main systematic limitation in the measurements, as is described in Paper I, is a possible frequency shift due to a q/A asymmetry effect. The uncertainty due to the relativistic mass increase can be as large as the q/A -effect but is decreased by optimizing the ion injection into the trap.

The q/A -effect can be avoided by always using a proper charge state of a carbon ion as a mass reference as to achieve mass doublets. For example in the measurement of the mass of an ion with mass number 200 and charge 50 this ion should be compared to an ion with $q/A \sim 0.25$. $^{12}\text{C}^{3+}$ has the same q/A value and could be used to achieve a perfect mass doublet measurement. The mass of the investigated highly charged ion then becomes:

$$M = \frac{\nu_{C^{3+}}}{\nu_1} \frac{q_1}{3} m_{C^{3+}} + q_1 m_e - \frac{E_B}{c^2}, \quad (4.1)$$

with

$$\frac{\nu_{C^{3+}}}{\nu_1} = \frac{\nu_{C^{3+}}}{\nu_2} \times \frac{\nu_2}{\nu_1}. \quad (4.2)$$

The highly charged ion and the H_2 ion are denoted with subscripts 1 and 2 respectively. Thus, in high-statistics measurements (*i.e.* a statistical uncertainties in the order of 0.2 ppb) the use of $^{12}\text{C}^{q+}$ ions with the proper charge between 3 and 6 would drastically reduce the magnitude of the q/A asymmetry effect. At the same time these measurements will allow a better evaluation of the actual effect. It is a justified question why $^{12}\text{C}^{3+}$ has not already been used in SMILETRAP. Through several tests we have so far the experience that it is not an easy task to produce C^{3+} ions in CRYISIS with properties suitable for SMILETRAP.

A way out of this problem is the ongoing measurements on singly charged ^3He and ^4He using H_2 ions as mass reference. When these measurements are completed they will give valuable input to the size of the q/A asymmetry effect. Since both these ions are already measured using doubly charged ions with low statistical uncertainties (0.2 ppb) the difference in the obtained masses using the two charge states of each ion will determine the slope of the q/A effect over a the range from 0.25 to 0.67. To really pin down the size of the effect it is also possible to accurately remeasure the frequency ratio between $\text{H}_3^+/\text{H}_2^+$ and to compare this ratio to that of H^+/H_2^+ . This was in fact one of the first measurements I did and presented in my undergraduate thesis.

Chapter 5

Conclusion

The Penning trap mass spectrometer SMILETRAP in combination with the ion sources CRYISIS/CHORDIS can with the present technique measure masses of almost any stable element. My work has proved that it is possible to determine masses at an uncertainty of ~ 2 ppb up to mass 204. Already 21 atomic masses using >20 charge states have been measured in SMILETRAP to this accuracy or better. In a few cases using q/A doublets and gathering high statistics an accuracy closer to 10^{-10} has been reached.

I have used SMILETRAP to determine masses that are useful for various fields in physics, *e.g.* the mass and Q -value of ^3H and ^{76}Ge that are useful in the search for a neutrino mass. In the case of ^{76}Ge the Q -value was not only useful but indispensable in the analysis of the β -spectra of this decay.

In the future it should be possible to reach an uncertainty close to 10^{-10} also for the heaviest ions using longer excitation times in combination with the Ramsey excitation technique and after further improvements of the B-field stabilization by adjusting the active feedback system.

With these improvements implemented we aim at testing the accuracy gain by measuring the mass of ^{28}Si . In case of a convincing result the masses of ^{29}Si and ^{30}Si should be measured with the aim of reaching an uncertainty close to 10^{-10} . All these three masses are related to the determination of the Avogadro constant and a new definition of the kilogram [27].

In the mass determination of the two even-even Mg isotopes (Paper IV) it was shown that there is a need for accurate mass measurements for the determination of the bound g-factor in H-like ions. After the determination of the g-factor in Mg the Mainz group plan to measure the g-factor of $^{40}\text{Ca}^{19+}$ which is about the heaviest ion that they can produce as H-like with the present setup. With an improved setup it will be possible to produce heavier H-like ions which implies a further need for accurate mass measurement.

The next generation of measurements of h/m_{Cs} by the group of Steven Chu [42] justifies a return to the measurements of the m_{Cs}/m_p mass ratio which is likely to be improved with one order of magnitude.

It is also justified to return to mass measurements of atoms lighter and heavier than Hg to find out what went wrong in the mass extrapolation by Audi and Wapstra.

As shown in this work there is no limit to which ion species that can be produced with CRYISIS/CHORDIS but it would be interesting if the maximum charge state could be further increased. If all elements up to Uranium could be produced as Ne-like ions it would both increase our accuracy and open up for interesting measurements of the electron binding energy in the heavy ions.

Like in any other field of physics it is hard to make a reliable predictions for future measurements, but it is clear to me that SMILETRAP has a very promising future.

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¹With his inspiring "såhär bra har det aldrig varit förut" or "bättre än någonsin tidigare"

²Somewhere here I also could have added Jan Weimer. But as you know Jan "någonstans måste vi dra gränsen".

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