DETERMINATION OF THE ATOMIC MASS CONSTANT BY ION ACCUMULATION

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Abstract − An experiment for the direct measurement of the atomic mass constant was performed by accumulating ions from an ion beam up to a weighable mass. The aim of this experiment was to develop an alternative approach for the redefinition of the SI unit kilogram. In a recent experiment a mass of about 300 mg bismuth was accumulated and the atomic mass unit could be determined with a relative uncertainty better than 1.0×10^{-4}.

Keywords: definition of SI-Unit Kilogram, mass accumulation, mass determination

1. INTRODUCTION

An experiment for the direct measurement of the atomic mass unit was proposed in [1]. The main idea is to produce a sufficiently intense heavy ion beam and to guide it into a collector where the total charge and mass of the accumulated ions can be measured. If the mass to charge ratio of a single atom, m_a / e, is assumed to be equal to the mass to charge ratio of all the collected ions, M / Q

$$\frac{m_a}{e} = \frac{M}{Q}, \text{ with } m_a = m_u \cdot A, A = \text{mass number}$$

one can determine the atomic mass unit, m_u, by means of a measurement of the total mass, M, and the ion current, I:

$$m_u = \frac{e \cdot M}{A \cdot Q} = \frac{e \cdot M}{A \cdot \int I(t)\, dt}$$

The charge, Q, can be obtained by the integration of the ion current over the accumulation time, t. This idea was realized with an experimental setup consisting of an ion source, followed by a magnet which separates the ions according to their mass to charge ratio, the ion collector and finally a high-sensitive balance.

2. EXPERIMENTAL SET UP

In order to accumulate a weighable mass in a manageable time ion currents of a few mA are needed. Therefore the so called **C**old or **Ho**t **R**eflex **D**ischarge **I**on **S**ource (CHORDIS) [2] was used, which can be operated in a gas-, a sputter- or in an oven version. For the production of singly charged bismuth ions the oven version was used, since due to the relatively low melting point of bismuth (271 °C) sufficient vapor pressures can be obtained already
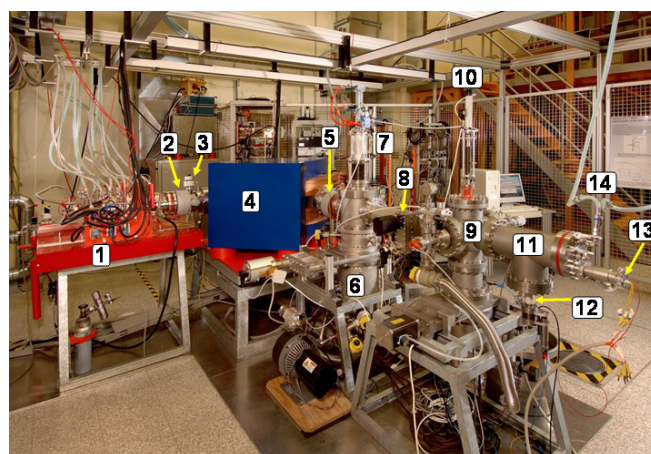

Fig.1. Experimental Set-up consisting of the ion source (1), seen on the red table. Just behind the source are two diagnostic instruments, a dc-current transformer (2) and a pressure sensor (3) respectively. The beam is bended by a double focusing dipole magnet (4). After the magnet a second current transformer (5) are seen followed by a vacuum chamber equipped with a turbo pump (6) at the lower part and a faraday cup (7) at the upper part. At point (8) a pneumatic valve is seen, which separates the ion source part of the set up from the collector part. Behind the valve a second vacuum chamber (9) is placed with another turbo-pump stage and a moveable wire monitor mounted above (10). The collector itself is housed in a T-shaped vacuum pipe (11) and is mounted on a table which seats on an isolator (12). The electron suppression electrode as well as the steering blend in front of the collector are cabled through the connector (13) at the very end of the system. The collector-bottom is cooled by a special water cooling system whereby the water is provided through an isolated flange (14).

at temperatures between 600 °C and 800 °C. For a continuous operation of the source several parameters has to be fulfilled simultaneously, which are mainly the cathode heating power, the oven heating power and temperature, the current from the cathode to the anode, the working gas pressure and the high voltage of the extraction system. The extraction voltage is usually between 20 kV and 30 kV.
To illustrate the extraction behaviour of the ion source an ion optical calculation using the IGUN-Code is shown in fig.2. The extraction system used in the CHORDIS ion source is a triode extraction system, which consist of an plasma electrode, a screening electrode and a ground

electrode. In the simulation the electrodes where biased to 23 kV, -2.3 kV and 0 V for (a), (b) and (c) respectively. The particle density was chosen to $1.1 \cdot 10^{11}$ atoms/cm^3. The main acceleration of the positively charged ions are happens from the plasma- to the screening electrode. From the screening- to the ground electrode the ions will be slightly decelerated, that's why the extraction system is also called "accel-decel" system. The screening electrode is necessary to keep the plasma electrons in the plasma chamber and to hinder electrons from outside to drift in the plasma room.
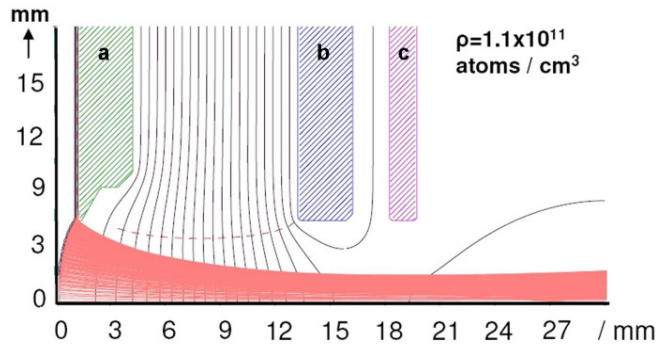

Fig.2 Ion optical simulation of the ion extraction from the ion source. Thereby (a) is the plasma electrode, (b) the screening electrode and (c) the ground electrode. The potential lines due to the voltage gradient are drawn between the electrodes. The curved shape at 0 mm position caused by the extracted ions is the plasma meniscus. Because of symmetry reasons the figure shows only the half extraction system. The same behavior would be obtained for the mirrored part on the x-axis. Red lines indicate certain ion trajectories.

Behind the source the ion current can be measured with a dc current transformer (Bergoz MCP). In addition the pressure at the working gas entry of the source and behind the source is controlled by pressure sensors (Pfeiffer-Vacuum PKR 256/251). The ion beam enters a double focussing dipole magnet 50 cm behind the source, where the ions are separated according their mass to charge ratio. At the entrance aperture of the magnet a four fold segmented stainless steel blend is mounted to check the horizontal and vertical ion beam position in respect to the magnet gap. In order to match the beam emittance in respect to the magnet acceptance we can vary the working gas pressure through a needle valve in the range of $0.5 - 4.3 \cdot 10^{-4}$ mbar measured behind the source. The pressure in the source influences strongly the so called plasma meniscus which mainly influences the ion optic through the extraction system, see also fig.2 and more details in [3].

Behind the magnet the beam current can again controlled via a second dc current transformer and a moveable water cooled faraday cup. The faraday cup is mounted on top of a chamber 50 cm behind the magnet. A turbo pump (Varian, 1800 l/s) is mounted on the bottom of the chamber and provides a vacuum of approximately $8 \cdot 10^{-8}$ mbar without beam and around a few 10^{-6} mbar with beam present. Thereby the vacuum is measured directly above the pump. After this chamber the beam line can be separated by a gate valve to isolate the collector part from the beam line. Behind the valve an another chamber is mounted equipped with a further turbo pump (Varian, 1000 l/s) on the bottom

and a wire monitor on the top. A mass spectrometer suitable up to 300 amu (Pfeiffer Vacuum, Prisma) is used to monitor possible desorption from the collector surface during the accumulation process. It serves in the set up also as residual gas analyzer and can be mounted on a side flange. The wire monitor is needed to adjust the magnetic field to a certain ion species. By varying the magnetic field and measuring the corresponding beam current on the wire a mass spectrum can be obtained from which the right magnet setting can be determined. After this second chamber a T-like beam pipe is mounted where the collector is housed.
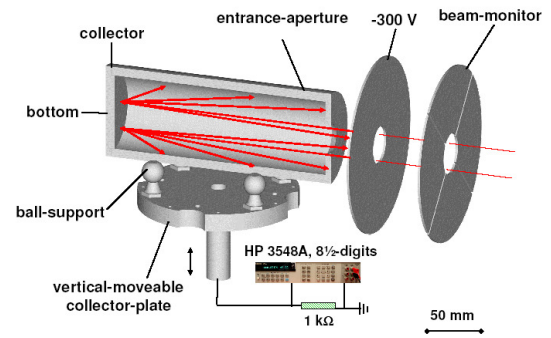

Fig.3. Collector Set-up consisting of collector cylinder, table holder, electron suppressor blend and segmented entry blend. In addition several ion trajectories are drawn.

Presently a cylindrical aluminum collector (Fig.3) with a length of 20 cm, a diameter of 8 cm and an entrance hole of 3 cm is used. The collector is placed on four stainless steel balls mounted on a special table in the vacuum chamber to minimize mass changes due to surface contacts. The table itself is electrically isolated from the surrounding vacuum chamber to guarantee correct current measurements. For adjustment purpose the table can be rotated and vertically moved. In front of the collector a segmented entry blend is mounted to steer the beam into the collector. Just after this blend a further blend is needed to suppress secondary electrons which will be produced by stopping the ions at the collector bottom. Figure 3 shows in addition some representative ion trajectories.

3. ACCUMULATION EXPERIMENT

The main task of the accumulation process is to collect all singly charged bismuth ions without losses of mass and charge in the collector. On disturbing factor is the fact that ions hitting the collector bottom remove atoms from the aluminum surface. This process is called sputtering and is mainly dependent from the ions energy and mass. The sputter process is characterized by the sputter-yield which is the ratio between the numbers of removed particles from the surface to the number of incoming (primary) ions. The sputter yield of bismuth ions with a kinetic energy of 25 keV on aluminum is approximately 13. Most of the sputtered

particles leave the surface under an angle of 50°- 60°. If the collector is long enough most of the sputtered particles will be collected at the cylinder-wall and only a small amount is scattered into the direction of the entry hole. In order to get a handle of the losses through the entry hole a polished stainless steel ring is mounted on the backside of the entry hole during the accumulation experiment. On this ring a mass accretion caused by the sputtered atoms, m_{ring}, can be measured by weighting the ring before and after the accumulation. In addition the sputter distribution along the collector tube was measured and could be very well reproduced by Monte Carlo simulations. The ratio between the particles which will be lost through the entry hole to the atoms collected on the ring, k_{MC}, can be calculated using this Monte Carlo Model. In summary 4.1% of the whole accumulated mass was lost by sputtering.

Figure 4 shows a part of the bismuth ion current which was measured in the collector. In addition the temperature of the oven is shown as a function of time. If the oven starts at room temperature it needs usually several hours until the full beam current can be obtained. The oven pipe connected to the plasma chamber and the plasma chamber itself must damped with bismuth, then the plasma density is high enough to extract the maximum possible current. If enough bismuth was condensed on the walls it is possible to switch of the oven.
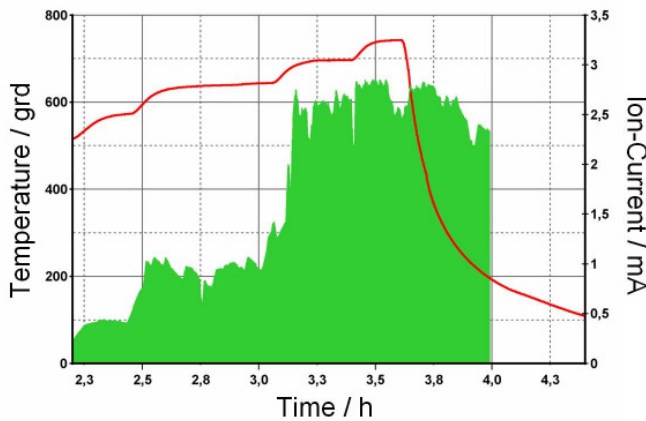

Fig.4. Oven Temperature (red line) and Ion-Current (histogram) as a function of time. The curves show only a part of the current to illustrate the current development as a function of the

After the accumulation process the collector was dismounted out of the vacuum chamber and placed manually in the special designed vacuum balance, which is located in a room very close to the experimental area.
From the mass difference of the collector before and after the accumulation the mass, M_{Coll}, of the collected bismuth ions can be determined, which was in the actual case an amount of 309.81 mg. Thereby the measurement uncertainty of the accumulated mass was 0.02 mg. A DC Bi$^+$ ion beam of 2.5-3.0 mA for about 25 hours must be produced for this mass.
The charge, Q, determination was done offline via current integration using the Simpson-Algorithm. Approximately every 60 ms a current was measured using a digital multi-meter (HP3458A) with 7½ digits. All together there was collected a charge of 149.19 As.

The uncertainty of the charge is determined through the sum of the individual integration samples in the Simpson formula which are depend from time interval between two current measurements and the uncertainty of the current itself. The uncertainty of the whole charge was determined to $1.4 \cdot 10^{-4}$ As.
The atomic mass can then be calculated using the discussed values in the following form:

$$m_u = \frac{e}{A} \cdot \frac{M_{Coll} + k_{MC} \cdot m_{Ring}}{Q}$$

which yields to:

$m_u = (1.660386 \pm 0.000156) \cdot 10^{-27}$ kg

This result has a relative deviation from the CODATA [4] value of $\approx 1 \cdot 10^{-4}$.
A more detailed discussion on uncertainties, including also more information on the determination of the sputter losses will be published.

4. CONCLUSION

An accumulation of a heavy ion bismuth beam up to a weighable mass in a collector was performed. DC bismuth ion currents up to 3 mA are sufficient to accumulate a mass of about 300 mg in a period of ≈ 20 h. With the described experiment we achieved a relative accuracy of better then 1×10^{-4} for the atomic mass unit.
This result is an improvement by nearly a factor 10 compared to our previously published result [5], which is mainly due to a complete new oven design for the ion source and a bigger magnet, with a gap nearly twice the size of the old one.

REFERENCES

[1] M. Gläser, Review Science Instruments Vol. 62, pp. 2493-2494, 1991.

[2] R. Keller, et al.: Nuclear Instruments and Methods Vol. B37/38, pp. 74-77, 1989.

[3] C. Schlegel, et al.: IEEE Trans. on Plasma Sci. Vol.36, pp 169-173, 2008.

[4] The NIST Reference on Constants, Units and Uncertainties, "2006 CODATA recommended values"

[5] C. Schlegel, et al: Metrologia Vol. 44, pp 24-28, 2007.