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Improved temperature regulation of Penning trap mass spectrometers

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ABSTRACT

With relative uncertainties better than $\delta m/m = 10^{-9}$ for mass measurements with Penning traps the temperature variation of the trap and surrounding materials must be kept within a few mK because temperature changes are known to shift the ion cyclotron frequency. In this paper we report a new temperature stabilization system recently installed at SMILETRAP II which regulates the flow of pressurized air that circulates around the trap enclosure. This system manages to keep the temperature fixed at the set point with a standard deviation of only 2.5 mK.

1. Introduction

Penning trap mass spectrometers (PTMS) are excellent devices for high-precision mass measurements [1]. In the trap ions are bound radially by a strong magnetic field, while the axial confinement is achieved with a set of electrodes creating a close to harmonic potential. Mass measurements with PTMS are based on the detection of the free-space cyclotron frequency ν_c which is related to the ionic mass through $\nu_c = qeB/(2\pi m)$, where qe is the charge and m the mass of the ion and B the magnetic field strength in the trap.

In a PTMS measurement the magnetic field strength is determined using a reference ion of well-known mass. By alternating between measurements of the cyclotron frequency of the reference ion ν_{ref} and the ion of interest ν_i a frequency ratio $R = \nu_i/\nu_{ref}$ can be derived from which the mass of the ion of interest can be calculated:

$$m_i = \frac{q_i m_{ref}}{q_{ref} R}. \quad (1)$$

From this the mass M of the neutral atom can be obtained by adding the mass of the missing electrons qm_e and their binding energies E_B:

$$M = m_i + qm_e - E_B/c^2. \quad (2)$$

Today several PTMS are being set up all over the world which aim to measure ionic masses with relative precisions of $\delta m/m = 10^{-9}$ (1 ppb) or better [1]. At these extreme precisions, several systematic effects have to be accounted for. In this paper we will discuss how the temperature instability of the trap will limit the precision of measurements with the commonly used time-of-flight ion cyclotron resonance (TOF-ICR) technique [2,3]. In Section 2 we will briefly describe the TOF-ICR technique as well as the temperature dependence of the measured frequency giving examples from SMILETRAP I [4] and ISOLTRAP [5]. Section 3 presents the temperature regulation system of the new SMILETRAP II setup. This system is unique as it regulates the flow of circulating air around the trap tube instead of regulating the temperature of the air. Finally we report the first results obtained with this system in Section 4 before we conclude.

2. Measuring ν_c with the TOF-ICR technique and precision limits

The TOF-ICR technique is thoroughly described in, for example, [1] and hence only a brief description will be given here. Using this technique the trapped ions are excited by an RF-field and then ejected from the trap. If the frequency of the applied field matches the cyclotron frequency, the ions gain kinetic energy. Measuring the mean TOF of ions from the trap to a detector as a function of the applied frequency, the cyclotron frequency appears as a well-pronounced minimum in the corresponding resonance spectrum (Fig. 1a). By fitting the theoretical resonance curve to the data, the cyclotron frequency can be obtained. The precision of

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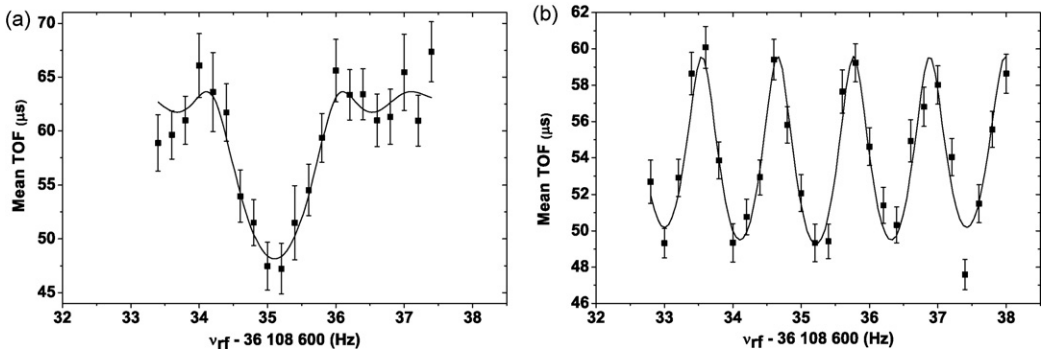

Fig. 1. Cyclotron frequency resonances in the case of single-pulse excitation (a) and two-pulse Ramsey excitation for D^+ ions [6].

the determination depends heavily on the width of the resonance which in turn is inversely proportional to the excitation time. For a single-pulse excitation the full-width at half-maximum is FWHM $\approx 0.8/T_{excitation}$ [7]. Apart from increasing the excitation time the resonance width can also be decreased using multi-pulse Ramsey excitation [8,9] (Fig. 1b) for which recent studies have shown an improvement in the final precision of up to three times compared to single-pulse excitation [10,11]. In order to cover the resonance spectrum (Fig. 1) some 20–30 different excitation frequencies are included in a scan around the expected cyclotron frequency. To avoid ion–ion interaction which could shift the measured frequency it is in many cases necessary to optimize the capturing procedure to one or a few trapped ions [4]. However, because it is very difficult to control the exact number of ions captured, sometimes zero or too many ions are trapped and these events have to be discarded in the off-line analysis. Therefore, several scans of the frequency window have to be bunched together in the analysis in order to achieve enough statistics for the fitting procedure.

Furthermore, a temperature dependence of the cyclotron frequency has been observed. In the case of SMILETRAP I (Fig. 2) the temperature coefficient has been estimated to about 11 Hz/K for an ion with $q/A = 1/2$ or, relative to the cyclotron frequency, about 300 ppb/K [4]. A similar estimation at ISOLTRAP yielded a coefficient of 0.182 mHz/mK for $^{133}Cs^+$ ions [12], corresponding to a relative value of about 266 ppb/K. One of the major reasons for this dependence is believed to be non-zero temperature-dependent magnetic susceptibilities of the construction materials, although other effects could also be present. Since it is in most cases difficult

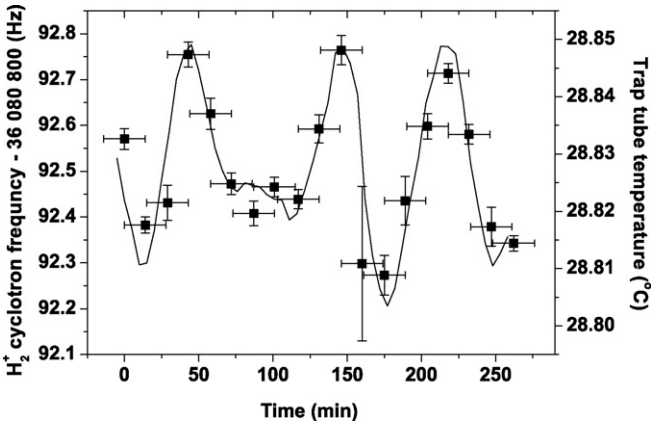

Fig. 2. Correlation between trap temperature (solid curve) and measured cyclotron frequency of H_2^+ ions (squares) at SMILETRAP I. The horizontal error bars of the cyclotron frequency data corresponds to the time interval during which the respective resonances were measured.

to regulate the temperature of the experimental hall to better than 100 mK, the question of temperature stability has to be addressed when aiming at relative precisions of 1 ppb or below.

At SMILETRAP I the laboratory room temperature was regulated within about 0.5 K peak to peak which corresponds to a variation in frequency of about 5 Hz or 150 ppb. In order to improve this, a temperature regulation system, consisting of a fan and a PID controlled heater, was installed. This system was, at optimal conditions, able to stabilize the temperature within 30 mK peak to peak. To further stabilize the magnetic field an active B-field regulation system was implemented. By applying a current through a warm correction coil proportional to the temperature of the tube of the magnet bore mostly larger temperature induced shifts of the magnetic field were canceled. However, a problem with such a system is adjusting the feedback to cancel the B-field variation and also there is a risk of introducing systematic errors from altering the B-field homogeneity. Even with these systems there still remained a slow oscillation around the set point [13] with an amplitude of about 0.3 Hz and a period of about 100 min induced by the temperature regulation system (Fig. 2).

In 2008 a temperature regulation system similar to the one at SMILETRAP I was installed at ISOLTRAP [12]. This system is, under normal running conditions with a room temperature stability of 0.4 K, able to regulate the temperature within 20 mK peak to peak. Under special conditions, when the room temperature has been stable to within 100 mK, the trap tube has been regulated to 5 mK. However, also this system shows an oscillation of the regulated temperature with a period of about 50 min.

Temperature oscillations pose a problem in Penning trap mass measurements. To avoid a shift of the frequency ratio, the measurement cycle has to be sufficiently short not to allow the magnetic field to change significantly between the measurement of ν_i and ν_{ref}. At the same time enough statistics has to be gathered to be able to fit the resonance curves. At SMILETRAP I this was solved by interchanging ion species often, usually every five scans (corresponding to about 2–3 min), while in the analysis 20–100 scans were bunched together. Although this approach reduced a potential measurement error from a shift of the frequency ratio, the temperature oscillation still limited the achievable precision. The effect of a short-time B-field oscillation on the resonance is illustrated in Fig. 3. Here the theoretical resonance curve in the case of a stable magnetic field is compared with the same curve time averaged over a full period of a typical temperature oscillation. The result is a broadening of the resonance width together with a smearing of the sidebands which reduces the precision of the frequency determination. The temperature-induced cyclotron frequency oscillation of 0.3 Hz is believed to be the main reason why the FWHM of a 1 s single-pulse excitation at SMILETRAP I was about 1 Hz rather than the expected Fourier limit of 0.8 Hz. Also, the resonance showed

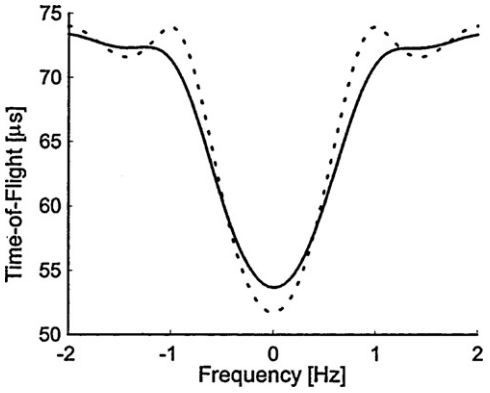

Fig. 3. An illustration of the smearing of the TOF resonance [13]. The dashed curve shows the theoretical TOF resonance curve of a single-pulse one-second excitation in the case of a stable magnetic field. The solid curve is obtained by time averaging the dashed curve over a full period of a typical temperature oscillation (Fig. 2).

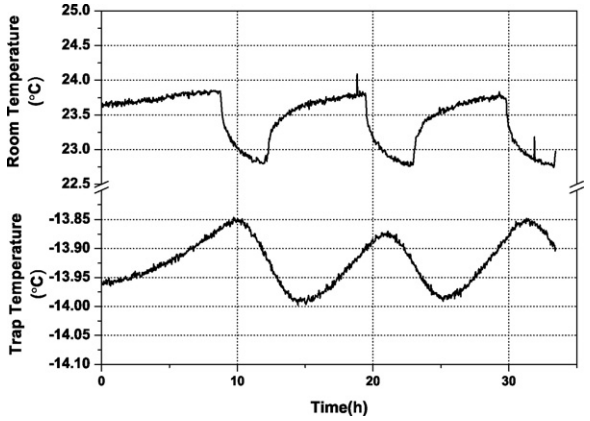

Fig. 5. Temperature of the experimental hall (upper curve) and the trap tube (lower curve).

a clear departure from the shape expected from a stable magnetic field, having less pronounced sidebands (Fig. 2). The smearing of the resonance becomes even more severe for longer excitation times or Ramsey excitation were the expected line width is smaller. This meant that measurements at SMILETRAP I were limited to excitations with resonances having a FWHM of about 0.6 Hz or larger, in turn limiting the achievable precision to about 0.2 ppb for a reasonable measurement time of a few days.

In the following section we describe the new regulation system that has been constructed for the high-precision mass spectrometer SMILETRAP II which is presently being set up at Stockholm University.

3. The temperature regulation system of SMILETRAP II

The schematic of the setup of the SMILETRAP II precision trap is shown in Fig. 4. The trap electrodes and holders are taken from SMILETRAP I and are made of oxygen-free high thermal conductivity (OFHC) copper, where the electrodes are in addition gold-plated. The trap is placed in the so-called trap tube which is situated inside a 5.8 T superconducting magnet. The tube itself consists of an outer and an inner tube made out of low-magnetic stainless steel, type 316 LN, identical to SMILETRAP I. The materials used inside the magnet were chosen to meet the requirements for ultra-high vacuum, high conductivities, low magnetic susceptibilities and other mechanical properties, respectively. A GaAlAs diode temperature sensor [14] is mounted to the outside of the outer trap tube approximately 20 cm from the precision trap to avoid any disturbance of the magnetic field homogeneity. Around this position the thermal contact to the cryostat makes the temperature of the trap region about −14 °C. As the trap and its holders are constructed out of OFHC copper the thermal connection between them and the cold surface of the trap tube is very good. The main heat transfer to the trap occurs along the stack of ion transport elements, as the inner trap tube is made of stainless steel and kept under ultra-high vacuum. However, the only part of the stack that connects to room temperature is a stainless steel potential mesh about 60 cm away from the trap at the end flange at the injection side. Thus, the heat transport from the outside is very low and the trap temperature is very close to the temperature at the thermal contact and the sensor position.

At the temperature of the trap of −14 °C, the sensitivity of the sensor is 2.8 mV/K. The sensor voltage is read with a high-precision National Instruments PCI 4351 card using the lowest voltage range with a resolution of 6.25 μV. In combination with the sensor this yields a total specified resolution of about 2.5 mK, also taking into account, for example, the stability of the current source and temperature effects on the read-out electronics.

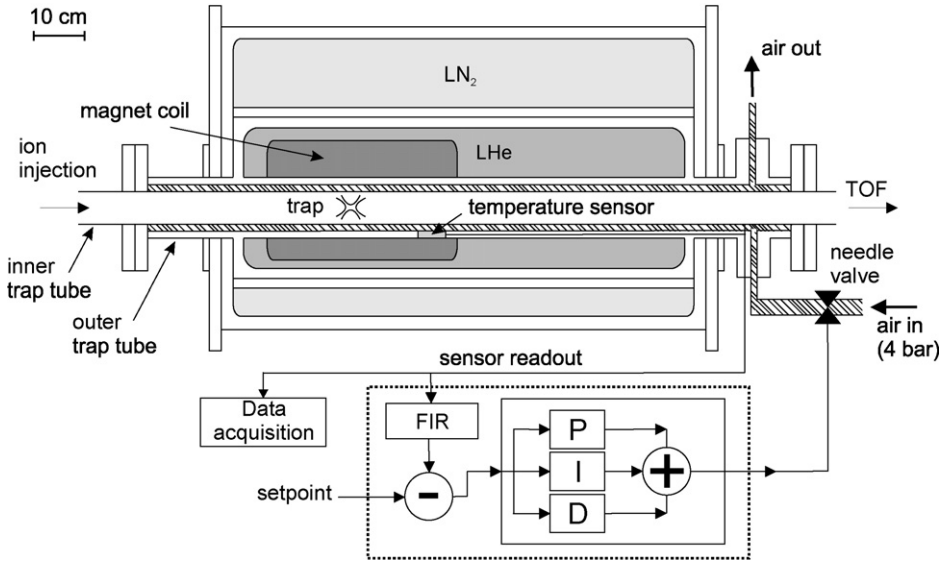

Fig. 4. Schematic layout of the SMILETRAP II Penning trap system with the temperature stabilization setup.

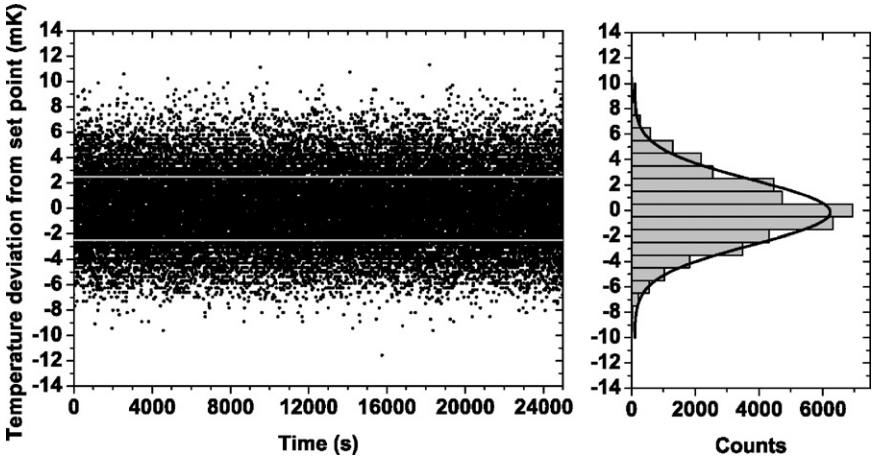

Fig. 6. Trap temperature deviation from the set point of −11.800 ° C over a period of 7 h. The horizontal gray lines represent the resolution of the temperature measurement. The full line in the distribution plot is a Gaussian fit to the data with a standard deviation of 2.5 mK.

In Fig. 5 the unregulated temperature of the trap tube and the temperature in the experimental hall are shown. A temperature change in the laboratory of 1 K leads to a strongly correlated change in the trap temperature by roughly 0.15 K. A delay in the temperature change of the trap tube of about 1.5 h is also observed. Due to temperature variations in the experimental hall of about 1 K, and also because of other temperature variations induced by, e.g., electronics, people entering or leaving and so on, it is necessary to stabilize the trap temperature directly by an active regulation system.

The new temperature regulation system is designed to regulate the flow of air between the inner and outer trap tube. The flow of air at room temperature from a reservoir with a pressure of approximately 4 bar is dynamically adjusted through a needle valve. Because of the cold trap region due to the thermal contact, the trap temperature increases with the airflow as heat is introduced into the system. Although the cold trap region is a feature of the SMILETRAP II setup, the same principle is universally applicable as long as an appropriate temperature difference between the trap and the gas reservoir is maintained. From this point of view the principle of operation differs from the stabilization system of SMILETRAP I and ISOLTRAP where the flow is constant and the temperature of the air is regulated. The valve is controlled by an in house-developed LabView (National Instruments) program. After filtering the temperature sensor read-out through a fifth order finite impulse response (FIR) filter to reduce the noise, the signal is fed to a PID algorithm generating the response signal for the valve. The PID program also features a self-optimizing algorithm to set the PID constants. The response time of the regulation is determined by the sampling time of the temperature sensor of 0.6 s. It is thus much faster than heat-controlled systems with an estimated response time of several minutes.

4. Results and discussion

Although the temperature variation of the compressed air used for the regulation is 1 K or more, its influence is small and a good temperature stability has been achieved. In Fig. 6 the measured temperature distribution around the set value of −11.800 ° C is plotted for a test measurement lasting seven hours. During this time the temperature in the experimental hall changed by about 1 K. A normal distribution fit to the data yields a standard deviation of 2.5 mK which is identical to the total estimated resolution of the temperature measurement system as specified by the manufacturers. This resolution can therefore be assumed to set the limit of the presently achievable stability.

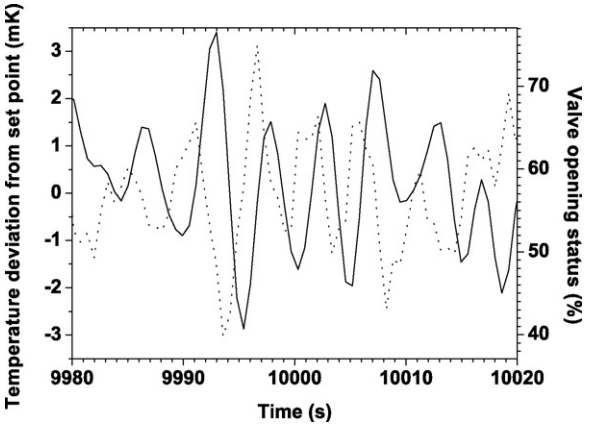

Fig. 7. Deviation of filtered trap temperature from the set point (full line) and valve position (dotted) over a period of 40 s.

A sample of the FIR-filtered signal together with the response of the valve is shown in Fig. 7. The response of the valve is lagging behind by a few seconds due to the filtering but the response is about three orders of magnitude faster compared to the heat-controlled regulating systems of SMILETRAP I and ISOLTRAP.

5. Conclusions

The new temperature regulation system at SMILETRAP II controls the gas flow around the trap region from a pressurized reservoir having a different temperature than the trap. This system keeps the temperature fixed at the set point with a standard deviation of only 2.5 mK or about 7 mK peak to peak even when the room temperature is changed by as much as 1 K. At SMILETRAP I this would have corresponded to a peak-to-peak frequency variation of about 80 mHz making it possible to resolve resonances below 0.2 Hz. Because the trap electrodes and holders are the same, as well as the materials of all other constructions, it is reasonable to assume that the temperature dependencies of the two systems will be close. Together with a higher magnetic field of the superconducting magnet of 5.8 T this implies that the temperature stability will not limit the precision of SMILETRAP II down to the level of 40 ppt (4×10^{-11}).

The present performance of the regulation system is limited by the resolution of the temperature measurement system. With a more precise temperature reading the stability could probably be improved. Also, stabilizing the temperature of the compressed air

to 0.1 K should be easy to implement and improve the stability even further.

Acknowledgments

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