Helium Pressure Shift of the Hyperfine Clock Transition in $^{201}Hg^+$

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***Abstract*— There are two stable odd isotopes of mercury with singly ionized hyperfine structure suitable for a microwave atomic clock: $^{199}Hg+$ and $^{201}Hg^+$. We are investigating the viability of a trapped ion clock based on $^{201}Hg^+$ in a configuration that uses a buffer gas to increase ion loading efficiency and counter ion heating from rf trapping fields. Traditionally, either helium or neon is used as the buffer gas at ~10^{-5} torr to confine mercury ions near room temperature. In addition to the buffer gas, other residual background gasses such as H_2O, N_2, O_2, CO, CO_2, and CH_4 may be present in trace quantities. Collisions between trapped ions and buffer gas or background gas atoms/molecules produce a momentary shift of the ion clock transition frequency and constitute one of the largest systematic effects in this type of clock. Here we report an initial measurement of the He pressure shift in $^{201}Hg^+$ and compare this to $^{199}Hg^+$.**

I. Introduction

NASA space exploration activities require atomic clocks capable of continuous operation, high stability, and high reliability. Over the past two decades the $^{199}Hg^+$ Linear Ion Trap Standard (LITS) has been developed at JPL to meet these requirements [1,2]. This technology is unique in that it delivers high stability on all time scales while operating at room temperature without the use of lasers, cryogenics, or microwave cavities. Ions confined in a rf trap are held near room temperature with a buffer gas and probed with microwave radiation. State preparation and detection is carried out using a UV plasma discharge lamp. To date virtually all trapped mercury ion clock developments of this type have been performed with trapped $^{199}Hg^+$ ions optically pumped with a $^{202}Hg^+$ discharge lamp.

The other stable odd isotope of mercury with hyperfine structure that can be used for a microwave clock is $^{201}Hg^+$. The $^{201}Hg^+$ isotope differs from $^{199}Hg^+$ not only in mass, but also in nuclear spin (3/2 compared to 1/2 for 199) and its nuclear magnetic moment which has a negative sign. Previously we measured the hyperfine clock transition frequency of the $^{201}Hg^+$ isotope [3]. In order to determine its viability for use in a clock and for possible application in searches for variations in fundamental nuclear constants [4,5], the systematic sensitivities of the $^{201}Hg^+$ isotope are being studied. Among these the largest are the second-order Zeeman shift, the second-order Doppler shift, and pressure shifts. The Zeeman shift for this isotope is well-known and easy to experimentally control. The second order Doppler shift can be well controlled in the laboratory using a compensated multi-pole ion trap [2]. The initial measurements of pressure shifts in $^{201}Hg^+$ reported here are taken in an older quadrupole ion trap design [1].

In this paper we will describe the experimental apparatus used to measure the He pressure shift in $^{201}Hg^+$, the systematic effects that we monitor and correct for, and the results. We will also describe future improvements that will enable better precision in the measurement.

II. Experimental Setup

A pressure shift experiment for a given background gas is conceptually straightforward: the $^{201}Hg^+$ clock frequency is monitored while the gas pressure is varied. For clock operation helium is commonly used as the buffer gas and is introduced through a heated quartz leak. The leak is pressurized to ~30 psi and heated sufficiently to diffuse helium and produce sufficient pressure. Our typical operating buffer gas pressure is ~1.3×10^{-6} torr as indicated on a calibrated Granville Phillips 370 ion gauge controller. (When correcting for gauge sensitivity the actual helium pressure is 5.56 times higher.) We change the helium pressure by varying the quartz leak rate. When measuring the pressure shift at low buffer gas pressure the non-linear pressure-dependent second-order Doppler shift dominates the observed frequency shift [6]. While the pressure shift is measured, other potential systematic shifts due to variations in the trapped ion number and other environmental changes must be monitored as well.

III. Systematic Uncertainties

The measurement shown in Figure 1 was performed in a quadrupole LITS [1,6]. In performing helium pressure shift measurements at the highest accuracy level, several systematic effects must be measured and corrected for. The trap resides in a magnetically shielded environment that has a bias magnetic field generated by a very stable current source. At the current level of precision reported here, the fractional frequency instability of the clock transition due to variations in the magnetic field is less than 10^{-15} and therefore inconsequential to the pressure shift measurements and no corrections are required.

The clock is currently in a laboratory environment and the ambient temperature can vary several degrees. We measured the end-to-end temperature coefficient with no active temperature compensation to be -2.3×10^{-14}/K. Under typical operating parameters we monitor temperature changes near the clock and, if necessary, correct for large thermal perturbations using this sensitivity during data analysis.

The measured clock frequency is referenced to a local hydrogen maser that is calibrated against UTC. It is only necessary that the maser stability remains smaller than our measurement error estimate over the measurement duration. The reference maser drifts at the level of $\sim 2 \times 10^{-15}$/day and our error estimate of the pressure shift measurement is at the 10^{-13} level. To further mitigate maser drift effects, the frequency at a given target pressure was always differenced from the frequency at a baseline pressure. Any frequency offset due to the maser was eliminated in the difference.

Changes in ion number change the clock frequency in several ways. Space charge repulsion expands the ions axially and radially as the ion number increases. Due to the quadratic trapping potential the ions experience a larger trap rf field as the ion cloud radius increases. This leads to an increase in ion micromotion and temperature, resulting in a change of the second-order Doppler shift. Figure 1 shows how the clock frequency changes with ion signal size. In order to correct for on number fluctuations, we operate the clock in the linear regime where signal size is less than 3000 PMT counts and correct the clock frequency as a function of signal size using the sensitivity shown in Figure 1. (Note: if measured in a compensated multipole linear ion trap, this sensitivity would vanish [2].)

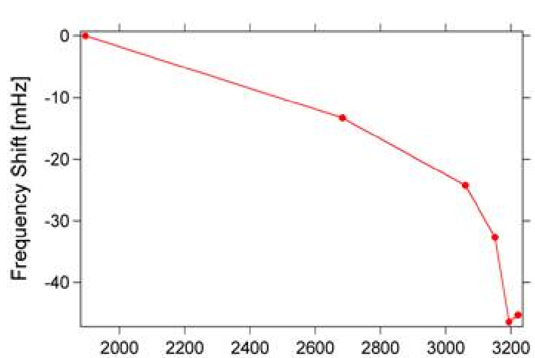

Figure 1: Clock frequency shift versus signal size change resulting from a variation of the trapped ions number in a quadrupole linear ion trap.

Varying the temperature of the quartz leak can potentially cause clock frequency changes due to other effects. We verified that the change in magnetic field due to quartz leak heater current is too small to be of concern. Also the leak itself is well-aged and extremely clean so that the relatively small changes in its temperature do not result in local outgassing rate changes for other gases.

We place an upper bound on fluctuations of all background gasses by measuring the vaccum chamber base pressure with the mercury oven and helium leak turned off. With the base pressure monitored over several days we observed pressure changes of less than 10^{-9} Torr. We expect the largest pressure shift to come from CH_4 [7,8]. In the worst case, if we assume the entire 10^{-9} Torr change to be due to CH_4, then the associated upper bound on frequency shift due to variations in the background gasses is expected to be $< 5 \times 10^{-14}$.

We measure the pressure using a NIST-calibrated Granville Phillips 370 ion gauge and controller. The manufacturer assigns an accuracy of 4% in the range of pressure we use in the experiment, which we include in the error estimate for the pressure shift measurement.

IV. Pressure Shift Measurement

In order to minimize ion number dependent shifts from the pressure shift, the ion number is held nearly constant throughout the buffer gas pressure test range (small changes are corrected for as described above As seen in Figure 2, even with constant ion number, changes in ion temperature at low helium pressure, resulting in a non-linear second-order Doppler shift change, can be much larger than the pressure shift in this trap [6]. The total shift due to changes in helium pressure is:

$$\Delta f = \Delta f_{SOD} + k_{He} \Delta P_{He},$$

where Δf_{SOD} is the second-order Doppler shift, k_{He} is the helium pressure shift constant, and ΔP_{He} is the change in helium pressure. As the buffer gas pressure increases, the ion temperature asymptotically approaches a minimum when the ions are in thermal equilibrium with the vacuum walls. At this point further increases in helium pressure no longer result in significant changes to the second-order Doppler shift. In this range where the frequency changes linearly with pressure, a straight line fit yields the pressure shift constant k_{He}. Future measurements will be performed in a multipole ion trap, which is nearly insensitive to Doppler related heating [6,2]. To further mitigate time-dependent systematic effects (usually diurnal), all frequency shift data was differenced from a clock frequency measurement taken at a baseline pressure both before and after the target pressure measurement. Figure 2 shows the measure clock frequency shift as a function of Helium buffer gas pressure changes.

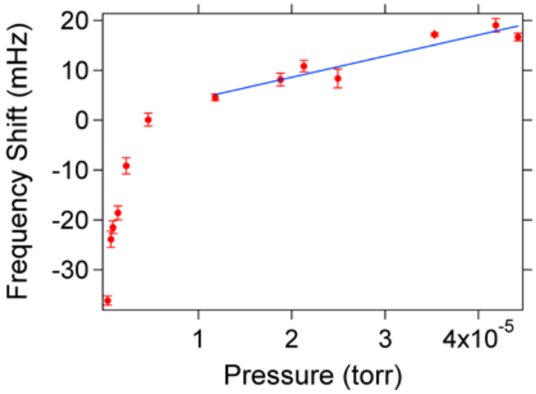

Figure 2: Frequency shift for $^{201}Hg^+$ versus helium pressure. A straight line fit is made to the data only in the high-pressure linear regime

The fractional frequency pressure shift for $^{201}Hg^+$ determined by a straight line fit to the data shown in Figure 2 gives:

$$\left(\frac{\Delta f}{f_0}\right)_{He}^{201} = 1.4(0.2)(0.5)\times 10^{-8}\ \text{torr}^{-1},$$

The first uncertainty is the rms scatter in the straight line fit. The second uncertainty incorporates our best estimate of the IG controller accuracy, and worst case perturbations due to background gases. Within the error estimate, this result is the same as the helium pressure shift in $^{199}Hg^+$ [6], consistent with theoretical expectations at this level of precision [9].

We next plan to measure the shifts of trace background gases by introducing them at a fixed helium buffer gas pressure [7]. Future experiments will also be performed in a compensated multipole linear ion trap, which will allow easy differentiation between Doppler heating effects and pressure effects.

Acknowledgements

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