A New Trapped Ion Clock Based On $^{201}Hg^{+}$

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Abstract—**There are two stable odd isotopes of mercury with singly ionized hyperfine structure suitable for a microwave clock: $^{199}Hg^{+}$ and $^{201}Hg^{+}$. Virtually all trapped mercury ion clocks to date have used the 199 isotope. We have begun to investigate the viability of a trapped ion clock based on $^{201}Hg^{+}$. We have measured the unperturbed frequency of the $^{2}S_{1/2}$ F=1, m_F=0 to $^{2}S_{1/2}$ F=2, m_F=0 clock transition to be 29.9543658211(2) GHz. In this paper we describe initial measurements with $^{201}Hg^{+}$ and new applications to clocks and fundamental physics.[1]**

***Index Terms*—Atomic clocks, ion traps, mercury**

I. Introduction

NASA Deep Space exploration activities require atomic clocks with 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 with these requirements in mind [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 are cooled to near room temperature with a neon buffer gas and probed with ~40.5 GHz (~30 GHz for $^{201}Hg^{+}$) radiation. State preparation and detection is carried out using a UV plasma discharge lamp.

To date virtually all trapped mercury ion clock development of this type has been performed with trapped $^{199}Hg^{+}$ ions and $^{202}Hg^{+}$ in a discharge lamp . The other stable odd isotope of mercury with hyperfine structure that can be used for a microwave clock is $^{201}Hg^{+}$. While early $^{201}Hg^{+}$ research led to initial measurements of some frequencies [3] most systematic sensitivities are largely unknown. Therefore we have begun an effort to measure these and determine the viability of $^{201}Hg^{+}$ for clock and fundamental physics applications.

The clock transition in $^{201}Hg^{+}$ is $^{2}S_{1/2}$ F=1, m_F=0 to $^{2}S_{1/2}$ F=2, m_F=0. State preparation into one the F=1 state is achieved with the discharge lamp since it is resonant with only the F=2 state. The lamp used to pump $^{201}Hg^{+}$ has better spectral overlap with $^{201}Hg^{+}$ than does the lamp used to pump $^{199}Hg^{+}$ (see Fig. 2) providing another motivation for studying $^{201}Hg^{+}$: the potential for faster optical pumping and a better state detection signal to noise ratio (SNR). In addition, $^{201}Hg^{+}$ has Δm_F=0 field-sensitive Zeeman lines that can be excited using the same polarization used to drive the clock transition enabling a simpler implementation to measure the magnetic field with improved precision. Finally, accurate knowledge of both $^{201}Hg^{+}$ and $^{199}Hg^{+}$ clock transition frequencies makes it possible to perform several measurements of interest in nuclear physics.

[1] This research was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under a contract with the National Aeronautics and Space Administration.

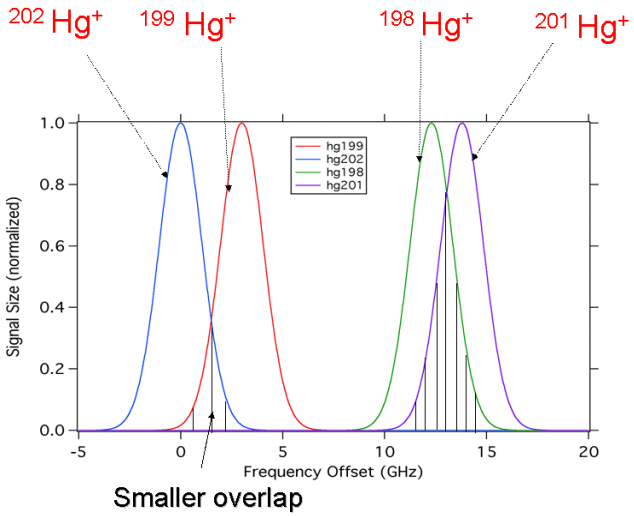

Figure 2. 194 nm UV spectrum of $^{199}Hg^{+}$ and $^{201}Hg^{+}$ and their respective lamps showing improved spectral overlap in the $^{201}Hg^{+}$ system.

II. $^{201}Hg^{+}$ Level Structure

$^{201}Hg^{+}$ and $^{199}Hg^{+}$ are the only stable isotopes of mercury possessing hyperfine structure suitable for a microwave atomic clock. One notable difference between them is their nuclear spin and magnetic moments, which are respectively I=1/2, μ=+0.506 μ_N for $^{199}Hg^{+}$ and I=3/2, μ=-0.567 μ_N for $^{201}Hg^{+}$ where μ_N is the nuclear magneton. Due to its nuclear spin of 3/2 $^{201}Hg^{+}$ has more energy levels than $^{199}Hg^{+}$. The negative nuclear magnetic moment of $^{201}Hg^{+}$ implies that its energy levels are reversed as shown in Fig. 3 (F=1 has higher energy than F=2).

III. State Preparation

The initial $^{201}Hg^{+}$ clock state has three dark levels (m_F=-1,0,+1) when optically pumped by the discharge lamp. To get high SNR, we need to reduce this to one dark state (m_F=0). As shown in Fig. 4, this is achieved by adding two microwave fields to drive transitions from the m_F=±1 states in addition to the optical field that drives the electric dipole transition at 194 nm. These additional Zeeman lines can be driven with the same linear polarization that is used to drive the clock transition (all have Δm=0).

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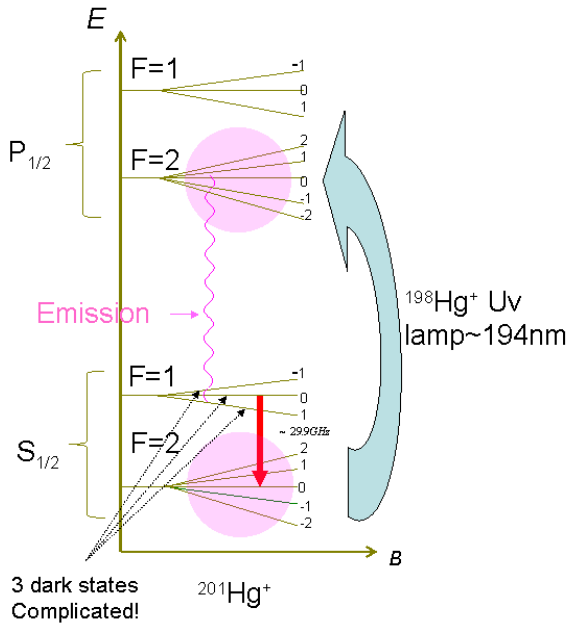

Figure 3. $^{201}Hg^{+}$ energy levels and optical pumping scheme with a $^{198}Hg^{+}$ UV lamp.

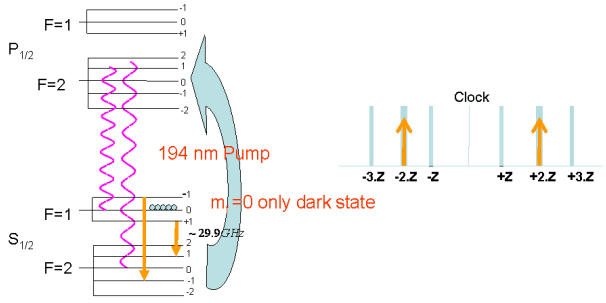

Figure 4. Microwave pumping scheme, by exciting the two Zeeman lines for which Δm=0, we empty the unwanted dark states.

IV. High Resolution $^{201}HG^{+}$ Spectroscopy

We performed high-resolution spectroscopy of $^{201}Hg^{+}$ and determined the unperturbed frequency of the $^{2}S_{1/2}$ F=1, m_F=0 to $^{2}S_{1/2}$ F=2, m_F=0 clock transition to be 29.9543658211(2) GHz [4], which is an improvement of 8 orders of magnitude from the previous known value. Fig. 5 shows spectroscopy for the clock transition using Rabi interrogation. A fit to the data gives mHz resolution on the shifted line center. The un-shifted frequency was determined by removing the largest systematic effects: second-order Zeeman shift, number- and temperature-dependent second-order Doppler shifts, background gas collision shifts, and the AC Stark (light) shift. In addition to performing spectroscopy on this transition, we have locked an LO to the clock transition and operated the instrument as a clock.

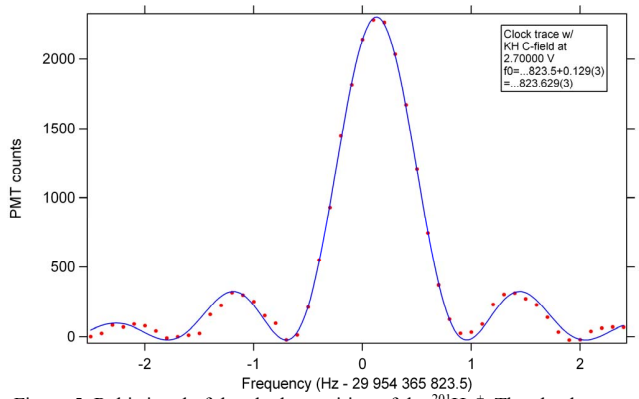

Figure 5. Rabi signal of the clock transition of the $^{201}Hg^{+}$. The absolute frequency after accounting for systematic shifts is 29.9543658211(2) GHz

V. Fast Optical Pumping

We measured an optical pumping time of 0.47 s for $^{201}Hg^{+}$ as opposed to 1.46 s for $^{199}Hg^{+}$ as shown in Fig. 6. This shorter optical pumping time makes it possible to reduce "dead time" in the clock cycle thus decreasing the impact of local oscillator (LO) phase noise. Less dead time also means a shorter cycle time enabling a shorter time constant for the loop that locks the LO to the atomic reference, which further improves short-term stability.

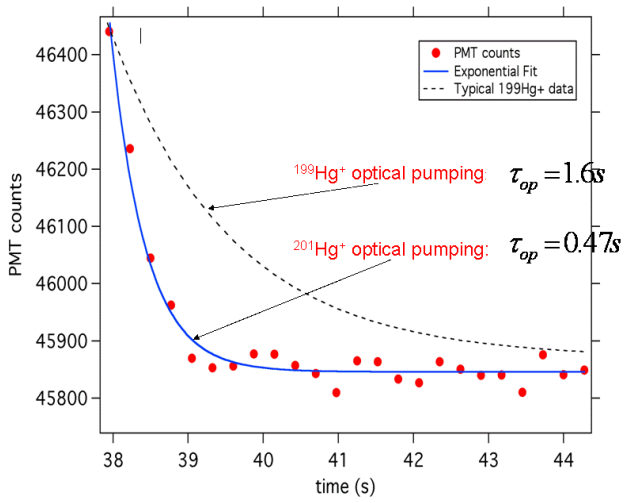

Figure 6. $^{201}Hg^{+}$ and $^{199}Hg^{+}$ optical pumping times.

VI. Zeeman Spectroscopy

As seen in Fig. 7, two of the Zeeman lines for which Δm_F=0 can be excited using the same polarization as the clock transition. Thus without modifying the microwave polarization we can detect these Zeeman lines with Hz resolution as opposed to kHz resolution in our previous $^{199}Hg^{+}$ measurements [5]. Consequently, we improve our knowledge of the frequency separating the Zeeman line from the clock transition by three orders of magnitude.

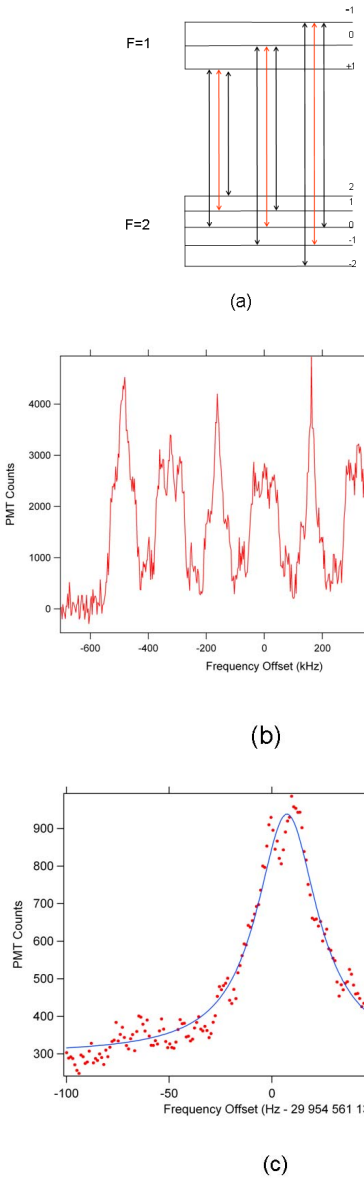

Figure 7. Zeeman spectroscopy of $^{201}Hg^{+}$. (a) The hyperfine energies transitions. (b) Power broadened Zeeman spectroscopy. (c) Spectroscopy of the Zeeman line (m_F=-1 to m_F=-1) with Hz resolution as opposed to previously measured kHz resolution for $^{199}Hg^{+}$ Zeeman spectroscopy.

VII. Fundamental Physics Measurements

A. Hyperfine anomaly

Hyperfine structure in atoms is primarily due to the energy of the nuclear magnetic dipole moment in the magnetic field generated by the electrons. If a point nucleus is assumed the ratio of hyperfine frequencies for two isotopes of the same atom (ion) can be calculated using the ratio of the nuclear magnetic moments, nuclear spins and total angular momenta. However, the nucleus has finite volume and the extent to which the measured frequency ratio differs from this idealized ratio is referred to as the hyperfine anomaly or Bohr-Weisskopf effect [6]. One of the advantages of having an improved knowledge of the ground state hyperfine frequencies of several mercury ion isotopes is that we can calculate the hyperfine anomaly of Hg^{+}. Until now knowledge of the hyperfine anomaly in Hg^{+} was limited by the knowledge of the $^{201}Hg^{+}$ hyperfine frequencies.

The hyperfine anomaly, Δ is defined by the following equation:

$$\frac{\Delta f_1}{\Delta f_2} = (1+\Delta)\,\frac{\mu_{I1}/I_1}{\mu_{I2}/I_2}\,\frac{F_1}{F_2}, \qquad (4)$$

in terms of the nuclear magnetic moments, μ_{Ii}, nuclear spins I_i and total angular momentum, F. Using the frequency ratio derived from the work presented in this paper and the clock frequency for $^{199}Hg^{+}$ [9] we find the Hg hyperfine anomaly:

$$\Delta\left(S_{1/2},^{199}Hg^{+},^{201}Hg^{+}\right) = -0.0016257(5). \qquad (5)$$

This new value for the mercury anomaly has a precision that is no longer limited by the frequency measurements, but instead on knowledge of the nuclear magnetic moment ratio [7],

$$\frac{\mu_{201}}{\mu_{199}} = -1.1074164(5)\ . \qquad (6)$$

B. Dual Ion clock.

It is possible that a comparison between the $^{201}Hg^{+}$ and $^{199}Hg^{+}$ hyperfine frequencies over a period of time might be able to place a limit on nuclear constant temporal variation [8]. We intend to operate a dual clock with both isotopes simultaneously interrogated in the same ion trap. Since the two species are located in the same trap they will experience the same perturbation. We expect similar systematic sensitivities for both isotopes (those for $^{199}Hg^{+}$ are well know and we are in the process of measuring those for $^{201}Hg^{+}$), thereby leading to further common mode rejection of systematic effects.

The ratio of the hyperfine frequencies of two atoms depend on the ratio of nuclear magnetic moments and the ratio of their Z values (leading to α dependence). If the two atoms are isotopes of the same atom, the α dependence drops out. Flambaum has shown that the magnetic moment ratio is proportional to the dimensionless nuclear constant m_q/Λ_{QCD} [10] where m_q is the mean light quark mass and Λ_{QCD} is the QCD scale factor. Thus a measurement between two isotopes of the same atom can yield a direct limit on m_q/Λ_{QCD}. The sensitivity to variations in the nuclear constant ratio can be expressed as:

$$\frac{\partial}{\partial t}\ln\frac{f_{201}}{f_{199}} = \frac{\partial}{\partial t}\ln\frac{\mu_{201}}{\mu_{199}} = [B_{201}-B_{199}]\frac{\partial}{\partial t}\ln\frac{m_q}{\Lambda_{QCD}}, \qquad (7)$$

where the B_i are sensitivity factors for each isotope. Using

Flambaum's method, Lea has calculated a sensitivity factor of 0.24 for mercury [11], which is significantly higher than other systems where this comparison has been made [12]. This higher sensitivity derived using this somewhat model-dependent approach is due to the large nuclear magnetic moment in Hg and that the moments have opposite sign in the two isotopes.

VIII. CONCLUSION

We have demonstrated a new ion clock using the 201 isotope of Hg^+. In addition, we carried out ground state Zeeman spectroscopy of $^{201}Hg^+$ and measured the clock transition with a resolution of 8 orders of magnitude better than previous measurements.

This new isotope distinguishes itself by its short optical pumping time and Zeeman transitions that can be excited with the same polarization that is required to drive the clock transition. The short optical pumping time enables a shorter cycle time, thus reducing the impact of LO phase noise. Our knowledge of the absolute ground state hyperfine transition frequencies for the two stable isotopes of Hg^+ has made it possible to derive a more precise value for the Hg^+ hyperfine anomaly, which was previously limited by the knowledge of the $^{201}Hg^+$ ground hyperfine absolute frequency.

$^{201}Hg^+$ and $^{199}Hg^+$ possess nuclear magnetic moments that have large magnitudes and opposite signs. This increases the sensitivity for a direct measurement of the nuclear constant ratio m_q/Λ_{QCD}. We plan to build a dual $^{201}Hg^+/^{199}Hg^+$ ion clock where a comparison between the two frequencies could be made with high common mode rejection of systematic effects. Such a measurement would place a stringent limit on temporal variation of nuclear constants.

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