Frequency Comparison of Two High-Accuracy Al$^+$ Optical Clocks

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We have constructed an optical clock with a fractional frequency inaccuracy of $8.6 \times 10^{-18}$,\footnote{We describe this as the "inaccuracy" because the relative nature of frequency measurement is the most relevant property.} based on quantum logic spectroscopy of an Al$^+$ ion. A simultaneously trapped Mg$^+$ ion serves to sympathetically laser-cool the Al$^+$ ion and detect its quantum state. The frequency of the $^1S_0 \leftrightarrow ^3P_0$ clock transition is compared to that of a previously constructed Al$^+$ optical clock with a statistical measurement uncertainty of $7.0 \times 10^{-18}$. The two clocks exhibit a relative stability of $2.8 \times 10^{-15} \pm 1/2$, and a fractional frequency difference of $-1.8 \times 10^{-17}$, consistent with the accuracy limit of the older clock.

Optical clocks based on petahertz ($10^{15}$ Hz) transitions in isolated atoms have demonstrated significant improvements over the current cesium primary-frequency-standards at 9.2 GHz. They also shed light on fundamental physics, such as the possible variation of physical constants.\footnote{This is a significant aspect of the research.} While the merits of laser-cooled ion optical frequency standards were known,\footnote{This is important to understanding the context of the research.} further developments were required to permit their use. Sub-hertz frequency standards were known,\footnote{These are the foundational elements of the research.} and advancements in isolated atoms have demonstrated significant improvements over the current cesium primary-frequency-standard.\footnote{This highlights the significance of the research.}

For comparison, cesium standards that realize the SI second have reached an inaccuracy of $3 \times 10^{-16}$,\footnote{This is a comparison to a well-known standard.} and an optical lattice clock based on Sr atoms has been reported with an inaccuracy of $1.5 \times 10^{-16}$. Here we describe an Al$^+$ ion clock with an inaccuracy of $8.6 \times 10^{-18}$.

The $^1S_0 \leftrightarrow ^3P_0$ transition in Al$^+$ at 1.121 PHZ is of interest due to its low sensitivity to electromagnetic perturbations and its narrow natural linewidth of 8 mHz.\footnote{This transition is key to the research.} Al$^+$ has the smallest sensitivity to blackbody radiation\footnote{This is highlighted due to its significance.} among atomic species currently under consideration for clocks, thus relaxing the requirement on ambient temperature control. However, the absence of an accessible optical transition prevents the internal state of Al$^+$ ions from being detected by conventional methods, and the ion cannot currently be directly laser-cooled. Quantum logic spectroscopy (QLS)\footnote{QLS is a key technique in the research.} overcomes these difficulties by trapping a "logic ion" that can be directly laser-cooled together with the Al$^+$ clock ion. The coupled motion of the two ions allows for sympathetic cooling, as well as the transfer of the clock ion’s quantum state to the logic ion, where the state can be measured.

The clock described here shares features with our previously-constructed Al-Be clock,\footnote{This is a comparison to previous work.} but also includes many changes, making a comparison of the two clocks a valuable test of systematic errors. In the new clock, the $^9$Be$^+$ logic ion has been replaced by $^{25}$Mg$^+$, whose mass closely matches that of $^{27}$Al$^+$. Laser-cooling inefficiencies due to mass-mismatch are thus suppressed. The Al-Mg ion trap is a linear Paul trap built from tool-machined all-metal electrodes (Fig. 1). This construction differs from the Al-Be trap that was built from laser-machined and gold-coated alumina electrodes,\footnote{This is a significant detail.} and it exhibits reduced RF-micromotion-inducing electric fields.

QLS with a Mg$^+$ logic ion proceeds in the same way as with Be$^+$, but the ground-state-cooling process\footnote{This is a detailed aspect of the research.} has been modified to require only two lasers rather than three. This cools the out-of-phase axial motional mode to an average quantum number of $\bar{n} < 0.05$. It also enables quantum-non-demolition transfer (QNDT) of the Al$^+$-clock-ion state to Mg$^+$ with approximately 80 % fidelity in a single QNDT repetition and over 99 % fidelity after typically five QNDT repetitions.

The trap utilized in the new Al$^+$ clock has blade-shaped gold-coated beryllium-copper electrodes (Fig. 1) whose edges are approximately 400 µm from the ions. The $^{25}$Mg$^+$ logic ion ($I = 5/2$) is manipulated with light of 279.5 nm wavelength from two independent lasers. A frequency-doubled dye laser resonantly drives $^2S_{1/2} \rightarrow 2P^{3/2}$ cycling and repumping transitions, while a frequency-quadrupled fiber laser is detuned by 40-60
GHz to drive Raman transitions between the $^{25}\text{Mg}^+$ hyperfine qubit states. Both ions are created via multi-photon-ionization of neutral atoms from ovens. A 396 nm diode laser produces $^{27}\text{Al}^+$ [17], while a frequency-doubled dye laser at 285 nm creates $^{25}\text{Mg}^+$ [18]. Typically the clock transition is probed with 150 ms duration $\pi$-pulses with a duty cycle of approximately 65 %. The remaining 35 % is occupied with state preparation and state detection functions as well as interleaved experiments that allow real-time measurement and reduction of micromotion.

Systematic shifts of the Al-Mg clock are listed in Table I. Two types of residual motion cause time-dilation shifts: micromotion near the trap drive frequency of $\nu_{RF} = 59$ MHz, and harmonic-oscillator (secular) motion at the ion’s normal mode frequencies (Table II). In both cases the clock frequency shifts by $\Delta \nu = -\frac{f^2\sigma^2}{2\pi^2\nu_{RF}} (1 + \frac{2\omega_{RF}}{\sigma^2})$, where we add to the relativistic time-dilation $(\sigma^2)/(2c^2)$ a frequency-dependent term that corresponds to the Stark shift from the motion-inducing electric fields. Here $(\sigma^2)$ is the mean-squared $\text{Al}^+$-ion velocity and $f$ is the frequency of motion. For the highest motional frequencies the Stark shift correction is 2 %.

Excess micromotion (EMM) refers to the rapid ion motion at $\nu_{RF}$ [19]. It is caused by electric fields that force the ion away from the RF-minimum of the ion trap, or phase shifts between trap electrodes that cause the RF-fields to be non-zero at the pseudo-potential minimum. We measure the amplitude of this motion at $\nu_{RF}$ by observing the motional-sideband strength of the $\text{Al}^+ \ 1S_0 \rightarrow 3P_1$ transition in three orthogonal directions. For small amplitude of motion, the time dilation shift is $\Delta \nu/\nu = -\bar{\eta} \nu_{RF}/\nu_L^2 = -2.8 \times 10^{-15}\bar{\eta}^2$, where $\nu_L = 1.12$ PHz is the probe laser frequency, and $\bar{\eta} = (\eta_x, \eta_y, \eta_z)$ is the measured EMM Lamb-Dicke parameter (the ratio of sideband and carrier Rabi rates) for the three orthogonal directions. Typical values for $\eta_{1,2,3}$ are 0.01 to 0.04. We note that this EMM measurement method detects slow electric field fluctuations such as those caused by the migration of photo-electrons, as well as faster fluctuations caused by periodic line noise (50 to 60 Hz). Fluctuations that are shorter than the laser probe period of 0.05 to 0.1 ms will not be detected.

In a perfect linear Paul trap EMM along the trap axis does not occur, but imperfections in the ion-trap geometry can lead to axial EMM. For the Al-Mg trap there exists a sharp minimum of axial EMM at one spatial location. The $\text{Al}^+$ ion is held at this point, but random background-gas collisions cause the Al-Mg ion pair to spontaneously re-order, which causes the $\text{Al}^+$ ion to move by 3 $\mu$m every 200 s, on average. The $\text{Al}^+$ ion is maintained at the position of minimal micromotion by adjusting the electrode voltages every 10 s to force the ion-pair into the desired order. When the ions are in the wrong order (about 5 % of the time), the $\text{Al}^+$ ion experiences excess axial micromotion and a corresponding clock shift of $\Delta \nu/\nu = -2.7 \times 10^{-17}$. This additional shift is included in Table II.

During each $\text{Al}^+$ clock interrogation pulse, the $\text{Mg}^+$ ion is simultaneously Doppler cooled by a laser that is tuned 21 MHz below the $|2S_{1/2}, F = 3, m = -3\rangle \rightarrow |2P_{3/2}, F = 4, m = -4\rangle$ cycling transition. The amplitude of secular motion (corresponding to the motional temperature) is extracted from the ratio of amplitudes for the red- and blue-sidebands of a Raman transition for all six normal modes [14]. Measured values are shown in Table II together with values calculated from laser-cooling theory for a single ion [20]. The single-$\text{Mg}^+$-ion Doppler cooling limit is also valid when applied to each of the six normal modes of the Al-Mg ion pair. The measured and calculated motional quantum numbers agree within the measurement uncertainty, and we consider the stated 30 % uncertainty for this shift to be a conservative limit.

The $\text{Mg}^+$ Doppler cooling laser beam maintains the
Al-Mg ion pair at a constant motional temperature during the clock interrogation pulse, but because it also overlaps the Al\(^+\) ion, it causes an AC Stark shift by coupling off-resonantly to allowed transitions that connect to the ground \((3s^2)^1S_0\) and excited \((3s3p)^3P_0\) clock states. Following the evaluation of the blackbody radiation shift \(14\), we estimate the differential clock polarizability at 279.5 nm as \(\Delta \nu/\nu = (-3.5 \pm 0.6) \times 10^{-17} S\), where \(S = I/I_S\) is the saturation parameter for \(^{25}\)Mg\(^+\) \((I_S \approx 2470 \text{ W/m}^2\)). The intensity \(I\) of the Mg\(^+\) Doppler cooling laser is estimated from the rate at which this laser repumps the \(|^2S_1/2, F = 2, m = -2\rangle\) dark hyperfine ground state. The ion fluorescence photo-multiplier counts \(F(t)\) collected in a duration \(t\) due to repumping of the dark state may be written as \(F(t) = b(t+\tau(e^{-t/\tau}-1))\) where \(\tau = (0.217/S)\) ms and \(b\) is the bright-state counting rate. We extract \(S\) by fitting \(F(t)\) to the observed ion fluorescence. Typically, we measure \(\tau = 2.1 \pm 0.8\) ms, and find \(\Delta \nu/\nu = (-3.6 \pm 1.5) \times 10^{-18}\).

Another Stark shift is caused by thermal blackbody radiation. The temperature of the Al-Mg ion trap is measured with two platinum sensors at opposite ends of the trap structure which are expected to be at temperature extremes. Heat is removed primarily through thermal conduction at one end of the trap where we measure a temperature of 35 °C. At the higher thermal resistance trap-end the temperature is 40 °C, and the thermal radiation field impinging upon the ion is bounded by this maximum temperature and the laboratory room temperature of 22 °C: \(T_{\text{ion}} = (31 \pm 9)\) °C.

During operation, the Al\(^+\) clock servo alternates between probing of the \(|^1S_0, m_F = 5/2\rangle \leftrightarrow |^3P_0, m_F = 5/2\rangle\) and \(|^1S_0, m_F = -5/2\rangle \leftrightarrow |^3P_0, m_F = -5/2\rangle\) transitions every 5 s, and the apparatus synthesizes an average of \(10^7\) counts per second. We observe a suppression factor of \(2.8 \times 10^{-15}\) and \(10^7\) counts per second. Each transition’s resonance is probed several times at the high- and low-frequency half-maximum points to derive a frequency-correction signal. The frequency-difference between the transitions is proportional to the mean magnetic field \(\langle B\rangle\), which allows an accurate estimate of the quadratic Zeeman shift due to the quasi-static quantization field of typically \(\langle B\rangle = 0.1\) mT. However, the quadratic Zeeman shift is proportional to \(\langle B^2 \rangle = (\langle B\rangle^2 + B_{A\text{C}}^2\rangle\) where \(B_{A\text{C}}^2\) is the variance of the magnetic field about its mean. The dominant sources of varying magnetic fields are currents at \(\nu_{RF}\) in conductors near the ion. We vary the trap RF drive power \(P\) and measure the frequency of the hyperfine clock transition in \(^{25}\)Mg\(^+\) \(|F = 3, m_F = 0\rangle \leftrightarrow |F = 2, m_F = 0\rangle\) near 1.789 GHz, which has a strong quadratic dependence on the magnetic field, to find \(B_{A\text{C}}^2 = 1.45 \times 10^{-12}(P/W)\) T\(^2\). For Al\(^+\) clock operation \(P = 15\) W, and \(B_{A\text{C}}^2 = 2.17 \times 10^{-11}\) T\(^2\), which alters the quadratic Zeeman shift by \(\Delta \nu/\nu = (-1.4 \pm 0.3) \times 10^{-18}\).

Other potential systematic shifts are listed in Table [11]. When stabilizing the clock laser to the ion we probe the clock transition alternately with counter-propagating laser beams to observe and cancel potential first-order Doppler shifts [15]. For the clock comparison described below, we observe a differential shift for the two probe directions of \((1.2 \pm 0.7) \times 10^{-17}\) but this effect is suppressed by taking the average. The suppression factor is limited because the atomic line-shape and hence the servo gains differ slightly for the two probe directions. During clock operation the mean fractional gain imbalance was 1.5 %, thereby reducing the possible first order Doppler shift to \(3 \times 10^{-19}\).

We have looked for Stark shifts due to the clock pulse itself by raising the intensity of the probe beam in one clock, and comparing the frequency to that produced by the other Al\(^+\) clock. With an increase of 40 dB in the clock probe intensity, we observe no statistically significant frequency difference with a fractional uncertainty of \(2 \times 10^{-15}\), constraining any effect that scales linearly with this intensity to \(2 \times 10^{-19}\). We observe a rate of background gas collisions similar to the Al-Be clock and assign a \(5 \times 10^{-10}\) uncertainty to this potential shift [16]. Thermally induced frequency errors from beam switching AOMs (Fig. [10]) have been evaluated previously [18], and this uncertainty is reduced to \(2 \times 10^{-19}\) by operating the AOMs at less than 1 mW.

We directly compared the Al-Mg clock with the Al-Be clock to perform independent tests that could reveal unaccounted-for clock shifts. The Al-Be clock was evaluated with an accuracy of \(2.3 \times 10^{-17}\) [16], and this evaluation remained valid during the two-clock comparison. The two clocks were compared in 56 separate measurements, each of duration 1000 – 11000 s. As shown in Fig. [11] the frequency of the \(^1S_0 \leftrightarrow ^3P_0\) probe laser was actively steered to the Al\(^+\) ion in the Al-Mg apparatus.
Future frequency ratio measurements of the Al$^+$ clock and the NIST Hg$^+$ optical clock would enable improved constraints on present-era changes in the fine-structure constant $^\text{[2]}$.

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\[ \Delta \nu / \nu = \frac{\nu_{\text{AlBe}} - \nu_{\text{AlMg}}}{\nu_{\text{AlMg}}} = (1.8 \pm 0.7) \times 10^{-17} \]

In summary, we have built an Al$^+$ ion clock with a fractional frequency accuracy of $8.6 \times 10^{-18}$. Its frequency is compared to that of a previously constructed Al$^+$ clock, and the measured fractional frequency difference of $(-1.8 \pm 0.7) \times 10^{-17}$ is consistent with the inaccuracy of the previous clock. Significantly, the statistical uncertainty in the frequency comparison of $7.0 \times 10^{-18}$ is smaller than the inaccuracy of either clock, and the average measurement stability was $2.8 \times 10^{-15} \tau^{-1/2}$ (total measurement duration: 164 967 s). This result may be compared to other direct same-species atomic clock comparisons, where two Cs microwave clocks have reached an agreement of $(4 \pm 3) \times 10^{-16}$ $^\text{[23]}$, and two Yb$^+$ single ion clocks showed agreement of $(3.8 \pm 6.1) \times 10^{-16}$ $^\text{[24]}$. Future frequency ratio measurements of the Al$^+$ clock and the NIST Hg$^+$ optical clock would enable improved constraints on present-era changes in the fine-structure constant $^\text{[2]}$.

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