

## Frequency Ratio of $\text{Al}^+$ and $\text{Hg}^+$ Single-Ion Optical Clocks; Metrology at the 17th Decimal Place

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**Time has always had a special status in physics because of its fundamental role in specifying the regularities of nature and because of the extraordinary precision with which it can be measured. This precision enables tests of fundamental physics and cosmology, as well as practical applications such as satellite navigation. Recently, a regime of operation for atomic clocks based on optical transitions has become possible, promising even higher performance. We report the frequency ratio of two optical atomic clocks with a fractional uncertainty of  $5.2 \times 10^{-17}$ . The ratio of aluminum and mercury single-ion optical clock frequencies  $\nu_{\text{Al}^+}/\nu_{\text{Hg}^+}$  is  $1.052871833148990438(55)$ , where the uncertainty comprises a statistical measurement uncertainty of  $4.3 \times 10^{-17}$ , and systematic uncertainties of  $1.9 \times 10^{-17}$  and  $2.3 \times 10^{-17}$  in the mercury and aluminum frequency standards, respectively. Repeated measurements during the past year yield a preliminary constraint on the temporal variation of the fine-structure constant  $\alpha$  of**

$$\dot{\alpha} / \alpha = (-1.6 \pm 2.3) \times 10^{-17} / \text{year}.$$

Time is the physical coordinate over which humans have the least control, and yet it is the most accurately realized fundamental unit. While any physical system that evolves predictably can serve as a time base, isolated atoms have long been recognized as near-ideal references for laboratory clocks, due to the abundance of identical copies, as well as their relative immunity to environmental changes. Because the forces within isolated atoms are many orders of magnitude larger than the external forces perturbing them, atomic resonance frequencies are affected only slightly by external fields. Yet even the small perturbations caused by external fields limit the accuracy of all atomic clocks.

In the work reported here, we combine recent advances in optical and atomic physics to construct atomic clocks based

on optical transitions in trapped  $^{199}\text{Hg}^+$  and  $^{27}\text{Al}^+$  ions, and measure their frequency ratio. Quantum-jump spectroscopy of single ions (1, 2) and subhertz lasers (3, 4) together with the femtosecond laser frequency comb (5, 6), allowed the first demonstration of an all-optical atomic clock (7), which was based on  $^{199}\text{Hg}^+$ . The development of quantum logic spectroscopy (8) has enabled the use of  $^{27}\text{Al}^+$  as a frequency standard (9), an ion that is highly immune to external field perturbations (10), but whose internal state is difficult to detect by conventional methods.

In each of the standards, the frequency that we attempt to produce in the laboratory is the resonance frequency of the unperturbed ion, at rest, and in the absence of background electric and magnetic fields. The deviations from this ideal condition produce shifts that are subtracted from the frequencies of two standards, to the degree that they are known (Table 1). The overall uncertainty in these shifts determines the final accuracy of each frequency standard. Although the specifics of the two standards are quite different, their respective systematic fractional frequency uncertainty is similar:  $1.9 \times 10^{-17}$  for  $^{199}\text{Hg}^+$ , and  $2.3 \times 10^{-17}$  for  $^{27}\text{Al}^+$ . Importantly, none of the current uncertainties are fundamental limits, and both standards can be improved substantially in the future, with a potential accuracy of  $10^{-18}$  or better (1, 11). The ratio of frequencies for the two optical clocks  $\nu_{\text{Al}^+}/\nu_{\text{Hg}^+}$  reported here marks an order-of-magnitude improvement in achievable measurement accuracy (12). As each of these clocks has an accuracy that exceeds current realizations of the SI-unit of time, we report the ratio of these optical frequencies, thereby avoiding the uncertainty ( $3.3 \times 10^{-16}$ ) of the currently realized SI-second (13).

Until recently, such an optical-frequency-ratio measurement (Fig. 1) would have required large and costly frequency-multiplication chains to translate between the microwave domain of electronic frequency counters and the

optical domain of the clock resonances. The development of tabletop femtosecond laser frequency combs (femtosecond combs) allows this translation to occur in a single, phase-coherent, convenient, and robust step. Here, the fourth harmonics of two clock lasers are locked to the mercury and aluminum clock transitions at 282 and 267 nm, respectively. An octave-spanning self-referenced Ti:Sapphire femtosecond comb (14) is phase-locked to one clock laser, and the heterodyne beat-note of the other clock laser with the nearest comb tooth is measured. The various beat-note and offset frequencies are combined to yield the unitless frequency ratio (12). In recent comparisons of the frequencies of the two optical clocks described here, a fiber laser femtosecond comb (15) has provided a second independent measure of the frequency ratio.

The  $^{27}\text{Al}^+ \ ^1\text{S}_0 \leftrightarrow \ ^3\text{P}_0$  standard, which uses quantum logic spectroscopy (8), has been described previously (9). One  $^{27}\text{Al}^+$  ion is trapped together with a  $^9\text{Be}^+$  ion, which provides sympathetic Doppler laser cooling as well as the means for internal-state detection of the  $^{27}\text{Al}^+$  ion's clock state ( $^1\text{S}_0$  or  $^3\text{P}_0$ ). The  $^{27}\text{Al}^+$  clock state is mapped to detectable states in  $^9\text{Be}^+$  repetitively through the ions' coupled motion, allowing for up to 99.94% clock state detection fidelity (16). With the ability to detect the clock state comes the ability to detect state transitions, whose probability depends on the clock laser frequency. The frequency of the clock laser is locked to the atomic transition by alternating between upper and lower slopes of the atomic resonance curve, and applying frequency-feedback to keep the transition rates equal. At the operating magnetic field of 0.1 mT, the Zeeman structure due to the nuclear spin of 5/2 is split by several kilohertz, and the individual Zeeman components are well resolved. The clock alternates every four seconds between  $\pi$ -polarized transitions with extreme states of opposite angular momentum ( $m_F = \pm 5/2$ ), which allows compensation of magnetic field shifts to first and second order (9, 17).

The accuracy of the aluminum standard is limited to  $2.3 \times 10^{-17}$  (Table 1), due primarily to uncertainties in the relativistic time dilation, or second-order Doppler shift, caused by microscopic movement of the ion in its trap, with rms velocities of  $v = 1$  to 2 m/s. According to special relativity, moving clocks are observed to run more slowly than stationary ones, with a fractional frequency shift of  $-v^2/(2c^2)$ . For ions confined in Paul traps, there are two types of motion: secular motion, which is the harmonic motion of the trapped particle, and micromotion, which occurs in part when the ion is displaced from the null of the RF confining field by slowly fluctuating electric fields. These quasistatic fields are monitored and nulled by interleaving micromotion test experiments with the clock interrogations. Tests are performed by measuring the strength of radial-to-axial coupling of certain normal modes via  $^9\text{Be}^+$  (18); nulling

is accomplished by applying compensation potentials at the ion trap to minimize this coupling. With real-time corrections, the stray electric fields are nulled to  $(0 \pm 10)$  V/m, allowing an estimate of the time dilation shift and uncertainty (19). The clock shift depends quadratically on the uncompensated electric field, and in the extreme case of a 10 V/m field along both radial directions, the fractional frequency shift is  $-3.2 \times 10^{-17}$ . We estimate the shift caused by such residual electric fields to be  $(-2 \pm 2) \times 10^{-17}$  when the clock is operating.

Secular mode heating (20, 21) causes deviations in the secular kinetic energy from the Doppler-cooling limit. We apply 313 nm  $^9\text{Be}^+$  Doppler-cooling light continuously during each 100 ms clock-transition interrogation, to keep the  $^{27}\text{Al}^+/^9\text{Be}^+$  ion pair as cold as possible. However, two poorly damped normal modes of motion heat the ion above the Doppler-cooling limit during this time (12), which leads to a second-order Doppler shift of  $(-1.6 \pm 0.8) \times 10^{-17}$ . Other important shifts are the blackbody radiation shift, which is very small in  $^{27}\text{Al}^+$  (10), and the DC quadratic Zeeman shift, which has been accurately calibrated by varying the magnetic field and measuring the shift in the  $^{27}\text{Al}^+/^199\text{Hg}^+$  frequency ratio together with the linear Zeeman splitting  $v_1$  between the ( $^1\text{S}_0 \ F = 5/2 \ m_F = \pm 5/2$ )  $\leftrightarrow$  ( $^3\text{P}_0 \ F = 5/2 \ m_F = \pm 5/2$ ) lines. The resulting shift is  $v_2 = -v_1^2 \times 1.0479(7) \times 10^{-8} \text{ Hz}^{-1} = -7.1988(48) \times 10^7 \text{ Hz/T}^2$ .

The  $^{199}\text{Hg}^+$  ion standard is based on the ( $^2\text{S}_{1/2} \ F=0$ )  $\rightarrow$  ( $^2\text{D}_{5/2} \ F=2 \ m_F = 0$ ) electric-quadrupole transition (22). A 194 nm laser cools the ion to the Doppler-cooling limit via the allowed  $^2\text{S}_{1/2} \rightarrow \ ^2\text{P}_{1/2}$  transition, and a fiber clock laser frequency-quadrupled to 282 nm excites the clock transition. The clock state of the  $^{199}\text{Hg}^+$  ion is measured directly via quantum jumps in the scattering fluorescence rate of the 194 nm laser (2). Systematic uncertainties in the  $^{199}\text{Hg}^+$  standard are listed in Table 1, and have been described previously (22, 23). The dominant uncertainties are due to the quadratic Zeeman effect and the electric quadrupole shift. The AC quadratic Zeeman uncertainty stems from possible unbalanced RF currents in the ion trap. The magnitude of this shift is conservatively estimated by assuming a worst-case asymmetry of 50% in the RF currents that flow in the nominally symmetric ion trap. Such an asymmetry would produce an rms field of approximately  $7.3 \times 10^{-7}$  T at the ion (12), causing a fractional frequency shift of  $-1.0 \times 10^{-17}$ . We use this value as an upper bound for the magnitude of the shift. The electric-quadrupole shift, which has previously limited the accuracy of the  $^{199}\text{Hg}^+$  standard, is constrained below  $10^{-17}$  by averaging over three orthogonal magnetic field directions (23, 24).

Second-order Doppler shifts for  $^{199}\text{Hg}^+$  are easier to control than for  $^{27}\text{Al}^+$ , because the heavy mercury ion moves less in response to ambient electric fields than the lighter aluminum ion does. Near the Doppler-cooling limit, the total

time-dilation shift due to secular motion is  $(-3 \pm 3) \times 10^{-18}$ . Micromotion is carefully compensated (19), leading to a shift of  $(-4 \pm 4) \times 10^{-18}$ . Thermal blackbody radiation has a negligible effect on the  $^{199}\text{Hg}^+$  ion, which operates in a cryogenic environment of 4 K(23). The quadratic Zeeman coefficient in  $^{199}\text{Hg}^+$  was calibrated in a way analogous to that of the  $^{27}\text{Al}^+$  standard. Here the shift is  $\nu_2 = -\nu_1^2 \times 4.9465(29) \times 10^{-11} \text{ Hz}^{-1} \approx -1.89 \times 10^{10} \text{ Hz/T}^2$ , where  $\nu_1$  is the linear Zeeman splitting between the ( $^2S_{1/2} F=0$ )  $\rightarrow$  ( $^2D_{5/2} F=2 m_F = \pm 1$ ) lines.

First-order Doppler shifts from photon-recoil are suppressed in tightly bound atomic particles (25), due to the Mössbauer effect, where each photon-absorption event lasts for many motional-oscillation cycles. Trapped ions (1, 2) and neutral atoms in optical lattices (26) benefit from this effect, but first-order Doppler shifts may still occur, if the trap itself moves in a correlated fashion with the clock laser pulses. Possible causes for such a shift in ion traps might be stray charge buildup from photo-electrons, which are correlated with the interrogation pulses, or more generally, correlated mechanical motion caused by thermal transients or optical shutters. A correlated velocity of only  $10^{-8} \text{ m/s}$  would cause a first-order Doppler shift error of  $3 \times 10^{-17}$ . Both standards were evaluated for first-order Doppler shifts by illuminating the ions with counter-propagating clock-laser beams, allowing such motion to be both detected and averaged away. The probe direction is selected by shifting the opposite laser beam away from the ion's resonance, either spectrally by  $\pm 100 \text{ kHz}$ , in the case of  $^{27}\text{Al}^+$  (several times per second), or spatially by a few beam waists in the  $^{199}\text{Hg}^+$  clock (every hour). We have not observed a direction-dependent shift in either standard. However, the ratio reported here contains unequal statistical weights for the two probe directions in the  $^{199}\text{Hg}^+$  standard. This leads to an additional uncertainty of  $7 \times 10^{-18}$  (12), which we treat as a systematic uncertainty in the  $^{199}\text{Hg}^+$  standard (Table 1).

The  $^{199}\text{Hg}^+$  and  $^{27}\text{Al}^+$  atomic clocks were operated simultaneously, while the femtosecond combs recorded their frequency ratio every second. Figure 2A shows the Allan deviation (an estimate of the statistical measurement uncertainty vs. measurement duration) of a typical ratio measurement. For measurement durations  $\tau$  greater than 100 s, the deviation is given by  $3.9 \times 10^{-15} / \sqrt{\tau / \text{s}}$ . A departure from this slope at long measurement times, which would indicate fluctuating systematic shifts, has not been observed. We expect that both clocks contribute uncorrelated noise of approximately equal magnitude to the statistical measurement uncertainty, and derive a long-term stability of  $2.8 \times 10^{-15} / \sqrt{\tau / \text{s}}$  for each clock.

Figures 2B and 3A show the history of measurements of the  $\nu_{\text{Al}^+}/\nu_{\text{Hg}^+}$  ratio. While the first-order Doppler evaluation discussed in the previous paragraph was carried out fully only

for the last four data points of Fig. 2B (which, when combined, give the last point of Fig. 3A), the consistency of the earlier measurements provides confidence in the reproducibility of this result. These four points result in a weighted mean of  $\nu_{\text{Al}^+}/\nu_{\text{Hg}^+} = 1.052871833148990438(55)$ , where the statistical uncertainty of  $4.3 \times 10^{-17}$ , the  $^{27}\text{Al}^+$  systematic uncertainty  $2.3 \times 10^{-17}$ , and the  $^{199}\text{Hg}^+$  systematic uncertainty  $1.9 \times 10^{-17}$  have been added in quadrature, to yield a fractional ratio uncertainty of  $5.2 \times 10^{-17}$ . This ratio may be multiplied by the  $^{199}\text{Hg}^+$  absolute frequency, which was calibrated by the NIST-F1 primary cesium standard (27), to yield a frequency of 1121015393207857.4(7) Hz for the  $^{27}\text{Al}^+$  standard.

Previous tests for possible counting errors or systematic offsets in the optical frequency combs have shown that the techniques employed here support fractional uncertainties of order  $10^{-19}$  (28, 29). Further details are provided in the online materials (12). As an additional check, for many of the measurements of Fig. 2B an independent Er: fiber based comb (15) measured  $\nu_{\text{Al}^+}/\nu_{\text{Hg}^+}$  simultaneously with the Ti:sapphire femtosecond comb (14). During the last four measurements of Fig. 2B these measurements show agreement at the level of  $1.3 \times 10^{-17}$ , which is consistent with the non-overlapping dead-time fractions of 3% and 5% in the frequency counters used by the two independent femtosecond combs. This dead-time error is already included in the statistical measurement uncertainty of  $4.3 \times 10^{-17}$ .

The last entry in Table 1, the gravitational red shift uncertainty of the  $^{27}\text{Al}^+$  clock with respect to the  $^{199}\text{Hg}^+$  clock, is only  $10^{-18}$ , because the two standards are in adjacent laboratories, and their height difference was easily measured with 1 cm uncertainty. This uncertainty is typically much larger for greater separations, and for example contributes  $3 \times 10^{-17}$  uncertainty for intercontinental primary-standard comparisons between Boulder, Colorado, USA, and other locations (30). In the future, high-accuracy portable optical clocks may be available, which could be used to map the height of the geoid with an accuracy beyond that achievable from satellite-based geodesy. Such an endeavor would also require an improved frequency-transfer link between a stationary reference clock and the distant portable standard.

Besides their application to frequency metrology and precision time-keeping, accurate atomic clocks can also help address a fundamental question in physics: Are the constants of nature really constant, or do they change in time, or depend on the gravitational potential in which they are measured? Frequency ratio measurements of dissimilar atomic clocks can help answer these questions, because these ratios depend on the fine-structure constant  $\alpha$ . From the sequence of measurements in Fig. 3A one may extract a linear rate of change in the frequency ratio of  $(-5.3 \pm 7.9) \times 10^{-17} / \text{year}$ . A fractional change in  $\alpha$  of  $\delta$  leads to a fractional shift of

$-3.19\delta$  in the energy of the  $^{199}\text{Hg}^+$  clock transition when it is expressed in units of the Rydberg energy (31). For  $^{27}\text{Al}^+$  the fractional shift is  $0.008\delta$  (32). Thus the measured linear slope in the frequency ratio corresponds to

$$\dot{\alpha} / \alpha = (-1.6 \pm 2.3) \times 10^{-17} / \text{year}, \text{ consistent with no}$$

change (Fig. 3B). However, due to the absence of first-order Doppler shift tests in the first point (Fig. 3 caption), this result must be considered preliminary.

The uncertainties in the atomic clocks reported here occur at the exciting intersection of relativity, geodesy, and quantum physics, and the total uncertainty of  $5.2 \times 10^{-17}$  shows unprecedented sensitivity to gravitational effects and cosmological fluctuations. Future improvements in these atomic clocks will provide even more sensitive probes of nature.

## References and Notes

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## Supporting Online Material

[www.sciencemag.org/cgi/content/full/1154622/DC1](http://www.sciencemag.org/cgi/content/full/1154622/DC1)

SOM Text

Figs. S1 and S2

References

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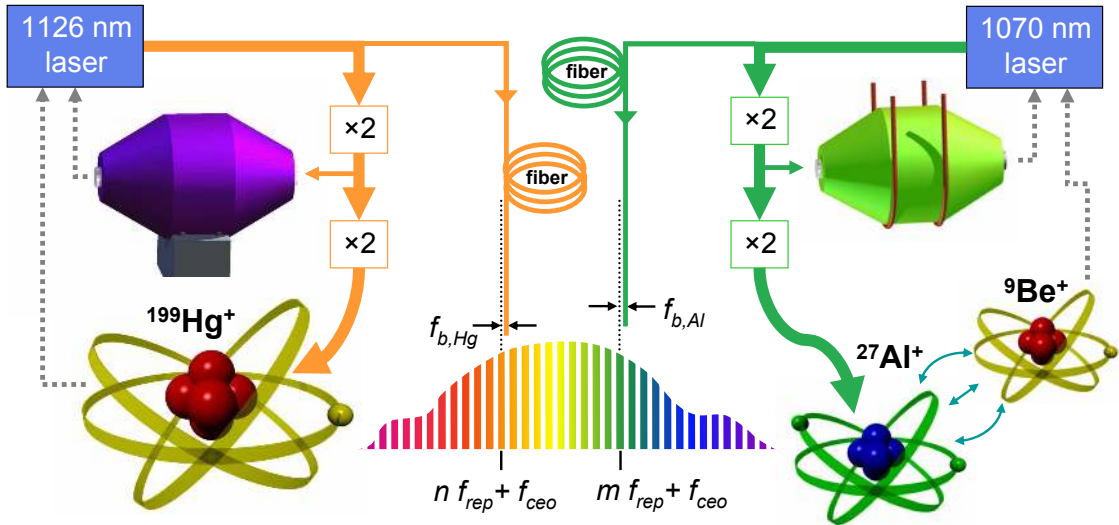
**Fig. 1.** Frequency ratio measurement system for the comparison of  $^{199}\text{Hg}^+$  and  $^{27}\text{Al}^+$  optical clock frequencies. On the left, the fourth harmonic of a 1126 nm wavelength infrared (IR) laser drives atomic state transitions in a  $^{199}\text{Hg}^+$  ion (40 ms probe time, 70% duty cycle). The transition rate yields an error signal to keep the laser frequency locked to the atomic resonance. On the right, a 1070 nm wavelength IR laser performs the same function for  $^{27}\text{Al}^+$  (100 ms probe time, 45% duty cycle), which is coupled to a nearby  $^9\text{Be}^+$  ion by their mutual Coulomb repulsion for the purposes of sympathetic cooling and internal state detection. Both lasers are pre-stabilized to ultra-low expansion glass Fabry-Perot cavities (purple and green ellipsoids), thereby narrowing their linewidth to about 1 Hz (4). Boxes marked “x2” are second-harmonic generation stages to convert IR light first to visible and then to ultra-violet wavelengths. The two laser frequencies are compared by means of a femtosecond comb (12), to which both clock laser systems are linked by 300 m lengths of actively phase-stabilized optical fiber.

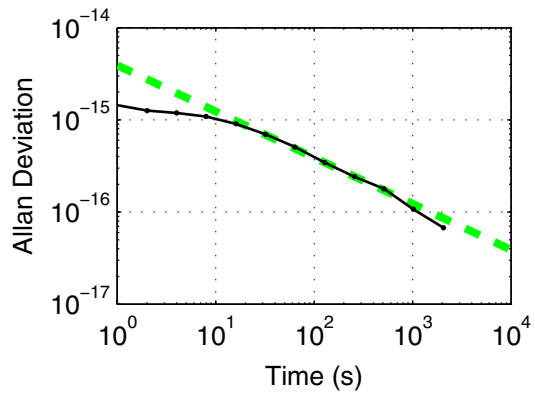
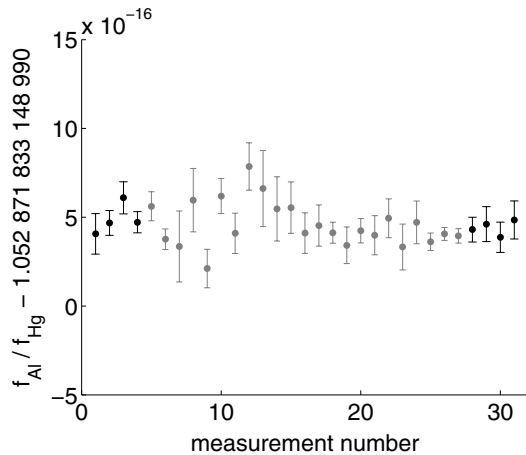
**Fig. 2. (A)** Allan deviation of a frequency comparison measurement (11000 s total). The dashed line represents a  $1/\sqrt{\tau}$  slope, beginning at  $3.9 \times 10^{-15}$  for 1 second. **(B)** History of frequency ratio measurements of the  $^{199}\text{Hg}^+$  and  $^{27}\text{Al}^+$  frequency standards. Error bars are statistical. Only the last four points are used in the ratio reported here (Fig. 3).

**Fig. 3. (A)** History of frequency ratio measurements grouped by month. The line connects the first point to the last one with a slope of  $(-5.3 \pm 7.9) \times 10^{-17}$  / year. Lightly shaded points represent measurements where various systematic shifts were not verified to be at the level stated in Table 1 at the time of the measurements. In the first point, the first-order Doppler shift was not monitored, but all other shifts were well controlled. We do not expect this point to have an error due to this effect, because neither apparatus has ever shown a first-order Doppler shift. For the last point, all shifts listed in Table 1 were well controlled. Error bars are a combination of the statistical measurement uncertainty, and the systematic uncertainties listed in Table 1. **(B)** Preliminary constraint on temporal variation of fine-structure constant  $\alpha$  from this measurement (vertical bar). The horizontal axis corresponds to variation of the fine-structure constant, and the vertical axis corresponds to variation of the cesium nuclear magnetic moment  $\mu = \mu_{\text{Cs}}/\mu_B$  in units of the Bohr magneton  $\mu_B$ , which is not constrained by this measurement. Also shown are laboratory constraints due to  $^{199}\text{Hg}^+$  vs. Cs (33) and  $^{171}\text{Yb}^+$  vs. Cs (34) measurements. The striped ellipse represents the standard uncertainty for the temporal variation of  $\alpha$  and  $\mu$  due to these previous comparisons of atomic clocks, and the smaller white ellipse shows the reduced uncertainty when the present result is combined with the earlier data. This ellipse constrains  $\dot{\mu}/\mu$  to  $(-1.9 \pm 4.0) \times 10^{-16}$ /year.

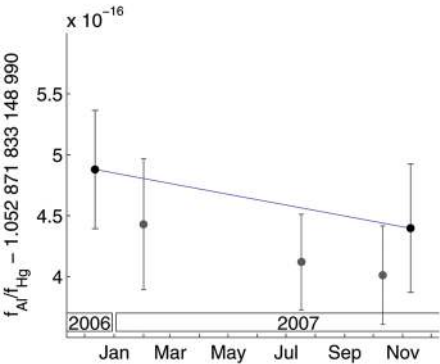
**Table 1.**  $^{27}\text{Al}^+ \ ^1\text{S}_0 \leftrightarrow \ ^3\text{P}_0$  and  $^{199}\text{Hg}^+ \ ^2\text{S}_{1/2} \rightarrow \ ^2\text{D}_{5/2}$  clock shifts ( $\Delta\nu$ ), and uncertainties ( $\sigma$ ) in units of  $10^{-18}$  of fractional frequency.

Shift	$\Delta\nu_{\text{Al}}$	$\sigma_{\text{Al}}$	$\Delta\nu_{\text{Hg}}$	$\sigma_{\text{Hg}}$	Limitation
Micromotion	-20	20	-4	4	Static electric fields
Secular motion	-16	8	-3	3	Doppler cooling
Blackbody rad.	-12	5	0	0	DC polarizability
313 nm Stark	-7	2	-	-	Polarizability, intensity
DC quad. Zeeman	-453	0.5	-1130	5	B-field calibration
AC quad. Zeeman	0	1	0	10	Trap RF B-fields
Electric quadrupole	0	0.5	0	10	B-field orientation
First order Doppler	0	1	0	7	Statistical imbalance
Bkg. gas collisions	0	0.5	0	4	Collision model
AOM phase chirp	0	0.1	0	6	RF power
Grav. red shift	-5	1	-	-	Clock height
TOTAL	-513	23	-1137	19	



**A****B**



**A****B**