

Absolute frequency measurement of the neutral ^{40}Ca optical frequency standard at 657 nm based on microkelvin atoms

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Abstract

We report an absolute frequency measurement of the optical clock transition at 657 nm in ^{40}Ca with a relative uncertainty of 7.5×10^{-15} , one of the most accurate frequency measurements of a neutral atom optical transition to date. The frequency ($455\,986\,240\,494\,135.8 \pm 3.4$) Hz was measured by stabilizing a diode laser system to a spectroscopic signal derived from an ensemble of 10^6 atoms cooled in two stages to a temperature of $10\ \mu\text{K}$. The measurement used a femtosecond-laser-based frequency comb to compare the Ca transition frequency with that of the single-ion $^{199}\text{Hg}^+$ optical frequency standard at NIST. The Hg^+ frequency was simultaneously calibrated relative to the NIST Cs fountain via the NIST time scale to yield an absolute value for the Ca transition frequency. The relative fractional instability between the two optical standards was 2×10^{-15} for 10 s of averaging time and 2×10^{-16} for 2000 s.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The revolution in optical frequency metrology that has come with the widespread implementation of femtosecond-laser frequency combs along with the advances of laser cooling has enabled contemplation of frequency metrology at the uncertainty level of 10^{-17} and below. Absolute frequency measurements of optical atomic transitions at this level could lead to interesting discoveries as we search for new physics in new domains. Of particular interest are tests of theories of atomic structure and QED [1], improved values for

fundamental constants [2] and even searches for drifts in the values of some fundamental constants [3–6].

Presently, many groups are investigating a variety of atomic, molecular and ionic transitions at higher and higher levels of stability and accuracy. Thus far, the most accurately measured optical frequencies belong to transitions in single trapped ions, due to the highly non-perturbative nature of their confinement [7]. Neutral atoms offer considerable benefits due to the high signal-to-noise ratios associated with the large number of atoms used and, in some cases, the simplicity of the lasers. Of particular significance are the accurate measurements (1 part in 10^{14}) of the 1s–2s transition in hydrogen [3], due to its theoretical simplicity and direct connection to *ab initio* QED calculations. Another transition that has been of considerable interest due to its narrow linewidth and relative experimental convenience is the intercombination transition in neutral calcium at 657 nm. This transition was first investigated in 1979 [8] and has since served as a test bed for many atom interferometry investigations

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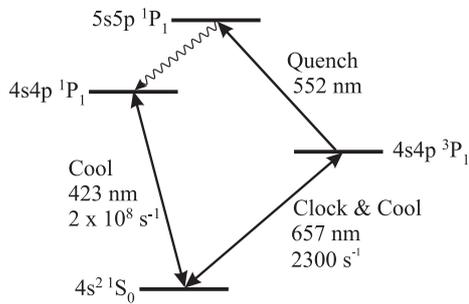


Figure 1. Partial energy level scheme for neutral ^{40}Ca , showing transitions relevant to the Ca optical frequency standard. Included are the Einstein A coefficients for the cooling and clock transitions.

[9–11]. One of its important applications is as an optical frequency standard. In fact, this intercombination line is one of only three optical transitions whose frequency has been measured with a fractional uncertainty of roughly 10^{-14} in multiple laboratories [1, 12–18].

In this paper we describe a new measurement of the 657 nm transition in Ca. This measurement takes advantage of a second stage of laser cooling and a new method for reducing laser probe beam imperfections to reduce the uncertainty of residual first-order Doppler shifts to less than 1 Hz. The resulting measurement has an uncertainty of 3.4 Hz and is limited mainly by technical effects (that could be considerably reduced in future investigations).

2. Experimental apparatus

The experimental setup has been described in detail in earlier publications [13, 19–21], but here we summarize the apparatus with particular emphasis on those aspects most relevant to the frequency measurement. The $^1\text{S}_0(m=0) \rightarrow ^3\text{P}_1(m=0)$ intercombination line at 657 nm (see figure 1) is well suited for an absolute frequency standard due to its narrow linewidth (375 Hz), convenient wavelength and inherent insensitivity to external perturbations. To reduce first-order Doppler uncertainties, we used two stages of laser cooling to reduce the temperature of the atomic sample to 10 μK before performing spectroscopy of the clock transition. The atoms were then excited with a diode laser whose frequency was pre-stabilized by locking it tightly to a narrow resonance of an environmentally isolated Fabry–Perot cavity. The resulting spectroscopic signals from the clock transition were used to fix the frequency of the diode laser on the centre of the atomic transition.

To perform coherent spectroscopy on the clock transition and to minimize systematic shifts due to cooling laser light and magnetic fields, we employed a measurement cycle as shown in figure 2. The cycle commenced with a loading period during which we filled a magneto-optic trap with atoms from a thermal calcium beam. This trap was based on the strong cooling transition at 423 nm and could load roughly 5×10^6 atoms in 15 ms [19]. The resulting atomic sample had a (near Doppler-limited) temperature of 2 mK. Second-stage cooling used the clock transition in conjunction with a quenching laser at 552 nm and produced a sample containing $\sim 10^6$ atoms with a temperature of 10 μK in 7 ms [20]. The

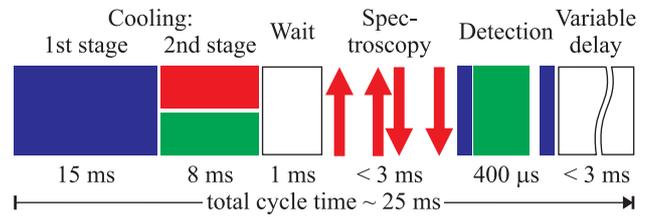


Figure 2. The cycle used to perform the spectroscopic measurements described in this work. First stage cooling based on 423 nm light is used to collect atoms and cool them to 2 mK. The second stage of cooling uses the narrow 657 nm transition in combination with the 552 nm quenching transition to reduce the atom temperature to $\sim 10 \mu\text{K}$. Then the four-pulse clock spectroscopy sequence occurs, followed by 423 nm pulses used in the normalized detection scheme. An optional delay can be added at the end to test for frequency shifts resulting from table vibrations or other effects that are synchronized with the measurement cycle.

atoms were released and expanded ballistically as they were probed by an atom interferometric method. This is based on a four-pulse Ramsey–Bordé sequence [22] where the atoms are first illuminated by a pair of pulses (separated in time by a duration, T) from one direction, then immediately excited by a second pair from the opposite direction. We used a combination of optical fibres and acousto-optic modulators (AOM) to generate the same pulse shape for all laser pulses and to provide spatial mode cleaning [21]. Finally, we used a normalized shelving detection scheme based on the strong cycling transition at 423 nm in conjunction with quenching light at 552 nm to measure the fraction of atoms excited by the sequence [20, 23]. This technique reduced the noise to near the atom shot-noise limit [24], thus enabling the high signal-to-noise ratio demonstrated in figure 3.

Ramsey–Bordé atom interferometry (AI) enables high resolution spectroscopy (resolution $\approx 1/4T$) while maintaining a high signal-to-noise ratio as shown in figure 3. The spectra were Fourier-transform limited, so all of the atoms in the sample contributed nearly equivalently to the signal. We note that the asymmetric envelope seen in figure 3(b) is a consequence of atomic recoil effects [25] and can lead to small shifts of the line centre [15, 21].

3. Stability of the calcium standard

Estimates based on the signal-to-noise ratio in the high-resolution spectra predict that a laser stabilized to the central fringe would have a fractional frequency instability of 4×10^{-15} for 1 s averaging time. We confirmed this level of instability by comparing it with a second stable standard, one based on a single trapped Hg^+ ion at 282 nm [7, 12, 26]. We bridged the 76 THz gap between the Ca frequency and half of the mercury frequency with a broadband femtosecond-laser frequency comb [27, 28]. Using the 2f–3f self-referencing technique, the comb’s carrier envelope offset frequency was locked to a stable microwave reference [29]. The comb’s remaining degree of freedom, the repetition rate ($f_{\text{rep}} \sim 1 \text{ GHz}$), was fixed by phase-locking a tooth of the comb near 563 nm (half the Hg^+ clock frequency) to light stabilized to the Hg^+ clock transition. In this way the stability of the Hg^+ probe laser was transferred to comb teeth spaced by 1 GHz throughout

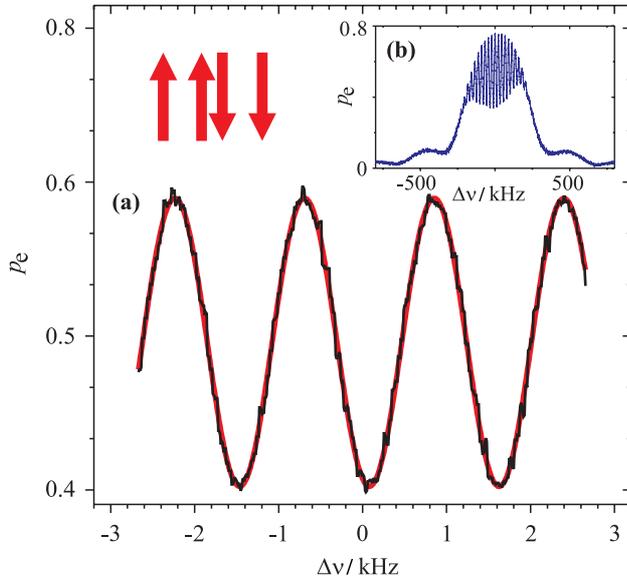


Figure 3. Four-pulse Ramsey–Bordé spectra taken at two different resolutions. The vertical axis shows the probability of excitation (measured by the fraction of atoms excited). A fringe resolution of 770 Hz is displayed in (a); this linewidth was used for much of the data taken and offered the highest performance (the scan shown required 4 s of data acquisition time). For reference, a sine wave of period 2×770 Hz is included. (b) A scan at lower resolution (11.55 kHz FWHM) displays the Fourier-transform limited envelope for the fringe structure. The asymmetry in the envelope is a consequence of atomic recoil and can lead to frequency shifts as described in the text.

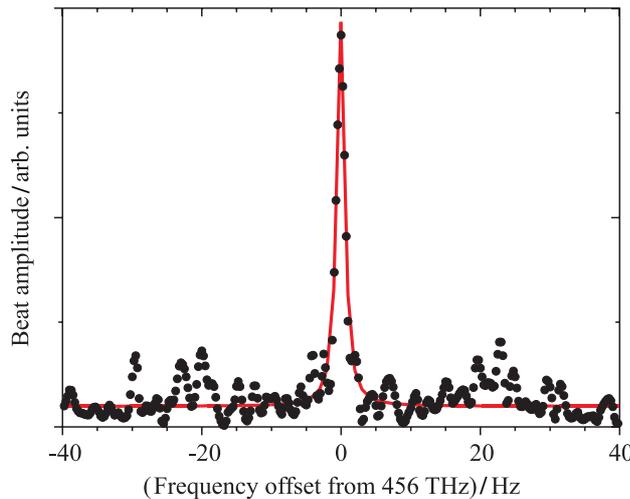


Figure 4. The beatnote between a tooth of the femtosecond-laser frequency comb around 657 nm and the Ca-stabilized light. The comb is self-referenced and locked to the Hg^+ laser system. The sweep time is 4 s with a resolution bandwidth of 1 Hz. The scale of the vertical axis is linear. A fit to the data with a pure Lorentzian yields a linewidth of less than 2 Hz.

the spectrum spanning from 580 nm to 1100 nm (273 THz to 517 THz). Figure 4 shows the beat between the Ca laser and a comb tooth near 657 nm.

Since light from both the Ca and Hg^+ lasers was sent to the comb through optical fibres, it was necessary to cancel the fibre-induced frequency noise to prevent degradation of

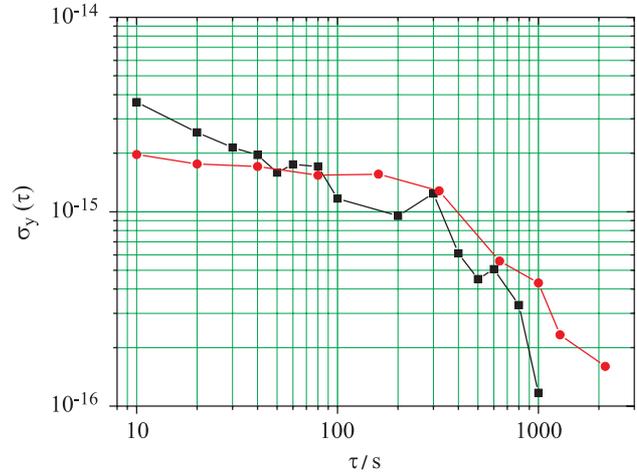


Figure 5. The Total Allan Deviation of the beatnote between the Ca and Hg^+ atomic clocks for averaging times from 10 s to 2000 s for two sets of measurements. The difference in stability at 10 s averaging time between the two curves results mainly from turning off exhaust fans in the room (circles = fans off). These data represent the instability of the entire system (the Ca and Hg^+ standards, the femtosecond-comb and both Doppler cancellation links).

the beat signal [30, 31]. This was achieved by two-way transmission of the optical signals and an AOM-based servo system to cancel the fibre noise. With the Ca and Hg^+ lasers locked to their respective atomic references, we used a frequency counter to record the beatnote and measured the relative frequency instability between the sources as a function of averaging time. Under these conditions we measured a relative instability of less than 5×10^{-15} at 1 s, confirming the estimations we made based on signal-to-noise ratios found in four-pulse AI spectra. This level was roughly a factor of two larger than that of the probe laser stabilized only to the reference cavity and a factor of three larger than the amplitude noise measured on the peaks of the fringes in the Bordé–Ramsey spectra. We attribute the elevated noise level at 1 s to extra laser frequency noise aliased down via the optical Dick effect [32].

In figure 5 we show two instability measurements as a function of averaging time for the Ca– Hg^+ beat signal, displayed as the Total Allan Deviation [33]. These measurements were made six months apart, between which several noise sources (mainly cooling and exhaust fans) were removed in the calcium lab, although this degraded the temperature stability in the Ca lab as well. The hump at about 400 s could be related to the temperature fluctuations, although its source remains unidentified. We emphasize that the optical sources can reach extremely low levels of instability ($\sim 2 \times 10^{-16}$) within reasonable averaging times (~ 2000 s). We note that with the same comb setup we could also measure the ratio of the Ca and Hg^+ frequencies by recording the relevant beatnote signals. Alternatively, the comb could be locked to the calcium standard and the resultant repetition rate could be directly counted. This arrangement turns the calcium frequency standard into a clock, which can be used as a source of extremely stable microwave radiation [29, 34].

4. Systematic shifts of the calcium clock frequency

Before measuring the absolute frequency of the Ca transition, we thoroughly evaluated the potential frequency shifts for the clock transition (see [21] for a detailed description). This evaluation took advantage of the high stability of the optical clock signals, which enabled relative measurements with sub-hertz accuracy for averaging times of only a few minutes. These measurements were performed with an interlaced system that enabled comparisons between signals taken under two different spectroscopic conditions (e.g. different resolutions); by alternating quickly (every 25 ms) between the two conditions, the drift of the reference cavity was largely suppressed. When possible we temporarily amplified the shifts for easier reduction and/or evaluation.

An important advantage of this approach was the reduction of the first-order Doppler shift, which limited previous measurements with our system to an uncertainty of 20 Hz. The addition of second-stage cooling reduces this to the few hertz level, but additionally we introduced a new method for improving two key laser beam parameters (wavefront flatness and residual angle between the counter-propagating probe beams) [21]. By launching clouds of atoms to velocities of up to 2.5 m s^{-1} (about 50 times larger than the rms velocity of the atoms at $10 \mu\text{K}$), we amplified the effects of the Doppler shifts and were able to isolate the various contributions. Potentially this approach could reduce the Doppler contribution to the total fractional uncertainty to well below a part in 10^{16} . However, due to probe laser beam imperfections (even after spatial filtering of the probes with optical fibres), we set a conservative value of 0.4 Hz (10^{-15} , fractionally) for the residual Doppler uncertainty due to these factors in these measurements. Additionally, a non-orthogonality between the horizontal probe beam direction and the acceleration of gravity leads to a Doppler frequency shift as the atoms accelerate during the spectroscopy. This effect was measured by reversing the order (or equivalently, the directions) of the pulses in the red pulse sequence (see [21] for details) and was found to cause a shift of just less than 5 Hz for these measurements (corresponding to a residual angle of 0.5 mrad). This pulse reversal technique, however, has its own uncertainty (0.5 Hz) due to the residual angle between the two horizontal beams. Adding (linearly, rather than in quadrature, since the errors could be correlated) the gravity uncertainty to the uncertainty due to probe beam imperfections, we find a total Doppler uncertainty of 0.9 Hz .

Table 1 lists the important potential systematic effects along with their associated shifts and uncertainties. While all of these effects are described in detail in [21], two were of particular importance during the absolute frequency measurements: frequency chirps caused by the AOMs (included in resolution-dependent phase shifts in table 1) and synchronized vibrations in the table. The frequency chirps result from ringing in the AOMs that make the short light pulses (duration = $3 \mu\text{s}$) used to excite the atoms [35]. Even though our setup employed a combination of AOMs and coupling into single mode fibres to produce the same pulse shape for all four laser pulses, the RF ringing in the AOMs still caused a fixed non-zero phase shift of the Ramsey fringe profiles. This in turn led to a frequency shift that was proportional to the fringe

Table 1. Uncertainty budget for the Ca standard for a spectroscopic resolution of 770 Hz (see text and [21] for details). $\Delta\nu_{\text{Ca}}$ is the shift of the frequency value due to a particular systematic effect, while $u(\Delta\nu_{\text{Ca}})$ is the uncertainty in this value (1σ values).

Effect	$\Delta\nu_{\text{Ca}}$ (Hz)	$u(\Delta\nu_{\text{Ca}})$ (Hz)
Residual Doppler	-4.7	0.9
Resolution-dependent phase shifts	1.6	1.3
Magnetic field	1.47	0.07
Quadratic Stark	-0.03	0.02
Black body (room)	-1.01	0.07
Black body (oven)	0	0.2
Ac Stark	0	0.1
Cold collisions	0	0.011
Thermal atoms (oven)	-0.9	2.2
Synchronized vibrations	0	1.1
Electronic offsets	0	0.8
Ca sub-total	-3.27	3.03
Frequency measurement statistics		0.01
Frequency measurement versus Hg		1.5
Measurement total	-3.27	3.4

spectral resolution. A second smaller uncertainty contribution to the resolution-dependent shift came from locking the laser to the asymmetric atom interferometric spectra (see figure 3). We measured the combined effects internally and evaluated them as well with absolute frequency measurements of the clock transition as a function of spectral resolution. Additionally, we estimated and corrected for the effect of the asymmetry (0.3 Hz with 1.0 Hz uncertainty) based on our knowledge of the actual velocity distribution of the atoms and laser intensity and shape as well as pulse parameters used for atom interferometry. The resulting correction for the combined resolution-dependent phase shift was 1.6 Hz with a 1.3 Hz uncertainty for a resolution of 770 Hz .

The second potential frequency shift of particular concern (synchronized vibrations) depended on the measurement cycle time. This shift was caused by offsets of the probe laser frequency due to vibrations in the optical table that were synchronized with the measurement cycle. More specifically, chopping of the current through our magnetic quadrupole field coils when switching between cooling and spectroscopy cycles induced vibrations in the optical table. These vibrations were synchronous with our measurement cycle and could lead to large shifts (tens of hertz!) of the average laser frequency depending on the length of the measurement cycle and the actual time at which the atoms were probed during the cycle (about 1 ms out of 25 ms , see figure 2). We suppressed these effects by suspending the vacuum system above the optical table, but as a check we also performed absolute frequency measurements for a variety of measurement cycle durations.

One other relevant shift was the gravitational redshift. The Ca optical frequency standard, the Hg^+ single-ion optical frequency standard, the hydrogen masers at NIST and the NIST Cs-fountain NIST-F1 are all located within a height difference of less than 8 m . The redshift is known to be slightly less than $1.1 \times 10^{-16} \text{ m}^{-1}$ [36]. This results in a systematic shift of less

than 0.5 Hz between all frequency sources. Since we measured the frequency of the Ca standard relative to that of Hg^+ , this correction and its uncertainty are effectively included in the value used for the Hg^+ standard (see table 1).

5. Frequency measurement results

Having evaluated the various systematics, we were ready to perform an absolute measurement of the Ca transition frequency. To double-check two key systematics (those resulting from resolution-dependent phase shifts and those from residual synchronized vibrations), we took advantage of the higher stability of the optical standards (~ 1 Hz in 10 s averaging time) by measuring against the Hg^+ single-ion optical frequency standard rather than the NIST time scale. However, the absolute value was traceable through the Hg^+ standard, as it was simultaneously calibrated by the NIST time scale (and ultimately the Cs primary standard, NIST-F1). The statistical uncertainty of the Hg^+ ion transition frequency for these measurements corresponds to 1.2 Hz at 456 THz (the Ca transition frequency), while its systematic uncertainty was 0.9 Hz (0.43 Hz for Hg^+ and 0.82 Hz for the maser calibration), yielding a total uncertainty of 1.5 Hz during this measurement. Facilitating the connection between all of the frequency standards was the same NIST broadband frequency comb described above and in greater detail in [29]. The Hg^+ frequency was calibrated by measuring the repetition rate against two masers of the NIST time scale [7], while the Ca frequency was determined from the ratio of comb tooth number (after removing the offsets) between Ca and Hg^+ .

The measurement consisted of three distinct phases, which took place on the same day (in October 2004). During the first phase we operated under nominally optimal conditions with minimal dead-time and a resolution of 770 Hz. The bulk of the data (2260 s) were taken under these conditions, and the resulting scatter in the frequency values derived from the Ca- Hg^+ beatnote (measured in 10 s averages) is displayed in figure 6. These values were averaged to yield the first frequency point in figure 7. For the rest of the points shown in part (a) in figure 7, we kept the resolution fixed at 770 Hz, but changed the amount of dead-time added on to the end of the measurement cycle to quantify residual effects of synchronized vibrations. During the third phase (the points in (b) of figure 7), we performed measurements to check for resolution-dependent effects. The actual corrections that were applied for this shift were based on independent internal measurements made on a second day and were consistent with those made during this third phase.

We note that the frequency measurements shown in figure 7 are already corrected for the systematic shifts (based on the independent measurements). The error bars represent the systematic and statistical uncertainties added in quadrature for the corrected individual runs. The measurements agree well within the error bars; however, since some systematic shifts may have been present for the whole set of runs, we cannot average the measurements to reduce the final uncertainty. The net uncertainty for the Ca system was 3.03 Hz, which, when combined with the 1.5 Hz uncertainty of the absolute reference (the maser-calibrated Hg^+ system), yields a total uncertainty of 3.4 Hz for the frequency measurement. We derive the final

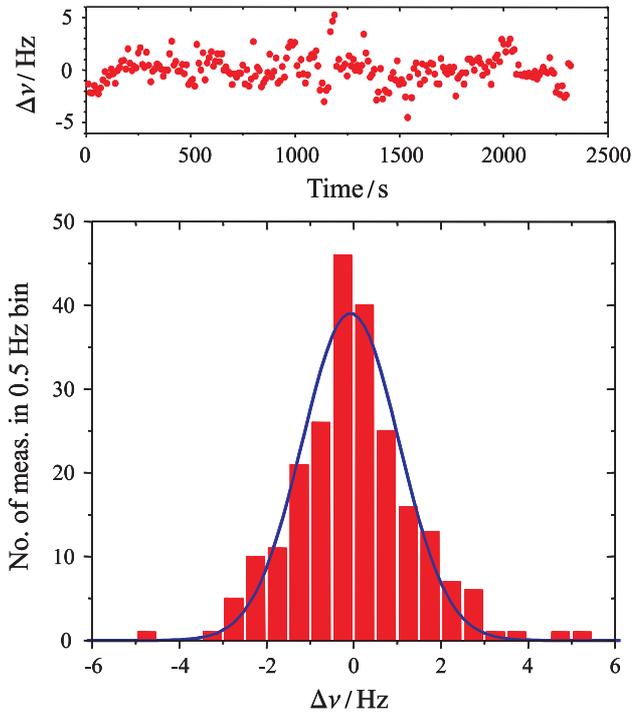


Figure 6. Top: A set of 10 s averages of the beatnote between Ca and Hg^+ taken over the course of one day. Bottom: A histogram containing 226 measurements of the Ca- Hg^+ beatnote taken at a resolution of 770 Hz with averaging times of 10 s. The solid curve shows a fit of a Gaussian to the histogram data.

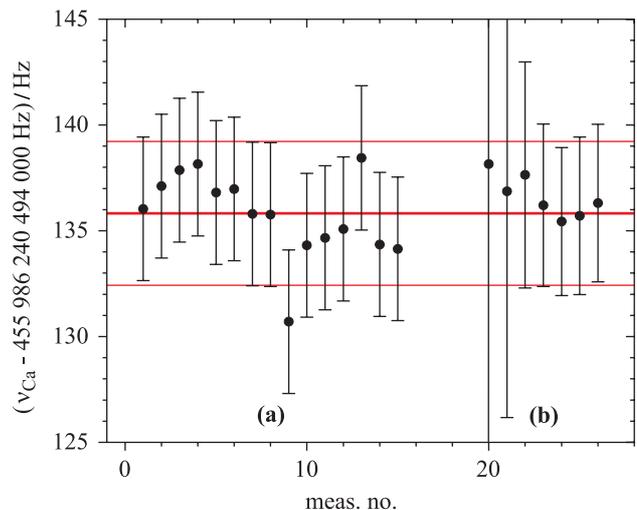


Figure 7. The set of frequency measurements performed for the Ca clock transition with corrections already included. The first fifteen measurements (labelled (a)) were taken at a resolution of 770 Hz. The second group (labelled (b)) were taken with resolutions ranging from 11.55 kHz to 385 Hz, as we checked for a resolution-dependent frequency shift. The larger error bars correspond to lower resolution measurements.

frequency value as a weighted mean (weighted by the error bars shown in figure 7) and assign the systematic uncertainty derived for the 770 Hz resolution measurements. The resulting value is $\nu_{\text{Ca}} = 455\,986\,240\,494\,135.8$ Hz \pm 3.4 Hz, i.e. a fractional uncertainty of 7.5×10^{-15} . Table 1 summarizes the systematic effects and their contributions to the final uncertainty (all reported uncertainties are 1σ).

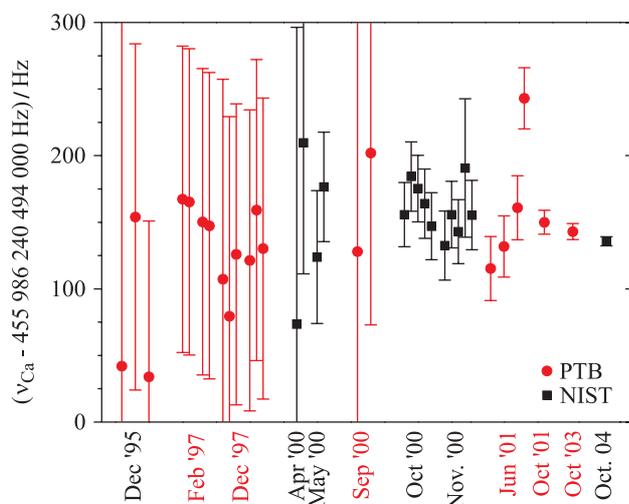


Figure 8. A historical plot of the absolute frequency measurements for the Ca clock transition at PTB (circles) and NIST (squares).

6. Conclusions

We have presented an absolute frequency measurement of the Ca clock transition at 657 nm relative to the Hg^+ reference, which was simultaneously calibrated by the NIST time scale. This frequency measurement achieved an uncertainty of 3.4 Hz, corresponding to a fractional uncertainty of 7.5×10^{-15} . Figure 8 shows the history of Ca frequency measurements at NIST and Physikalisch Technische Bundesanstalt (PTB), demonstrating excellent agreement through the years. We note, however, that the last two frequency measurements differ by 8 Hz [15], roughly the sum of the 1σ error bars for the two measurements, so more precise determinations of this transition frequency would be valuable. Since the dominant uncertainties in table 1 are technical in nature, future measurements with an uncertainty below 10^{-15} certainly seem feasible. However, much more accurate measurements with neutral atoms will more likely come from lattice-based systems, which can achieve extremely narrow spectroscopic linewidths and largely suppress Doppler effects [14].

Additionally, the short measurement cycle period (25 ms) led to good stability for the Ca standard. In measurements between the Ca and Hg^+ frequency standards, the combined instability of the two systems was 2×10^{-15} at 10 s averaging time, and 2×10^{-16} at 2000 s. Better pre-stabilization of the probe laser (e.g. to an improved reference cavity) could lead to lower instability for the Ca standard, since its present performance is limited by laser frequency noise and is still more than an order of magnitude higher than the calculated atom shot-noise limit [37].

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