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Frequency combs for precision synthesis and characterization of optical atomic standards

To cite this article: Tara Fortier as a representative of the BACON collaboration 2024 *J. Phys.: Conf. Ser.* **2889** 012021

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Frequency combs for precision synthesis and characterization of optical atomic standards

Tara Fortier as a representative of the BACON collaboration

Time and Frequency Division, National Institute of Standards and Technology, 325
Broadway MS 847, Boulder CO 80305 USA

E-mail: tara.fortier@nist.gov

February 2024

Abstract. Over the past 20 years, optical frequency combs with atomic clocks, have been a powerful and enabling technology in the context of time and frequency measurement. Impressively, optical atomic clocks have yielded a 100 million-fold improvement in uncertainty in the past 30 years. These improvements are fueling a push toward redefinition of the SI second to optical atomic references, as well as application of atomic clocks to tests of fundamental physics and as relativistic gravitational sensors. Unfortunately, the long times needed to average down clock quantum projection noise and laser noise to reach a measurement stability at and beyond the 10^{-18} level limit the feasibility of next-generation applications. Here I describe the measurement advances in clock comparisons enabled by optical frequency combs and how differential measurement can improve the measurement stability. This paper will also include a discussion of optical frequency combs and their application to precision time/frequency metrology.

1. Introduction

Atomic clocks began their development in the mid 20th century and quickly became a mainstay of modern-day life; serving as the definition of the SI second since 1967 and being the time reference for global positioning system (GPS) starting in the early 1980's. To date, the best microwave atomic clocks based on a ground state hyperfine transition in ^{133}Cs , have demonstrated repeatability and measurement accuracy near 1 part in 10^{16} [1]. The latter requires impressive control, measurement and knowledge of how stray fields impact the 9.2 GHz carrier at the microhertz level. While there still exists some opportunity to improve the accuracy of existing microwave atomic standards, the development of clocks based on electronic optical transitions enables a natural route to much higher performance [2, 3, 4]. This is possible given that atomic clock accuracy and resolution scale inversely with the transition frequency. After more than two decades of development, optical atomic clocks have been evaluated to operate with uncertainties near, or even slightly below 10^{-18} [5, 6, 7] and with short-term instabilities approaching 10^{-16} at a 1-s averaging time, Fig.4. These gains in performance have provided a



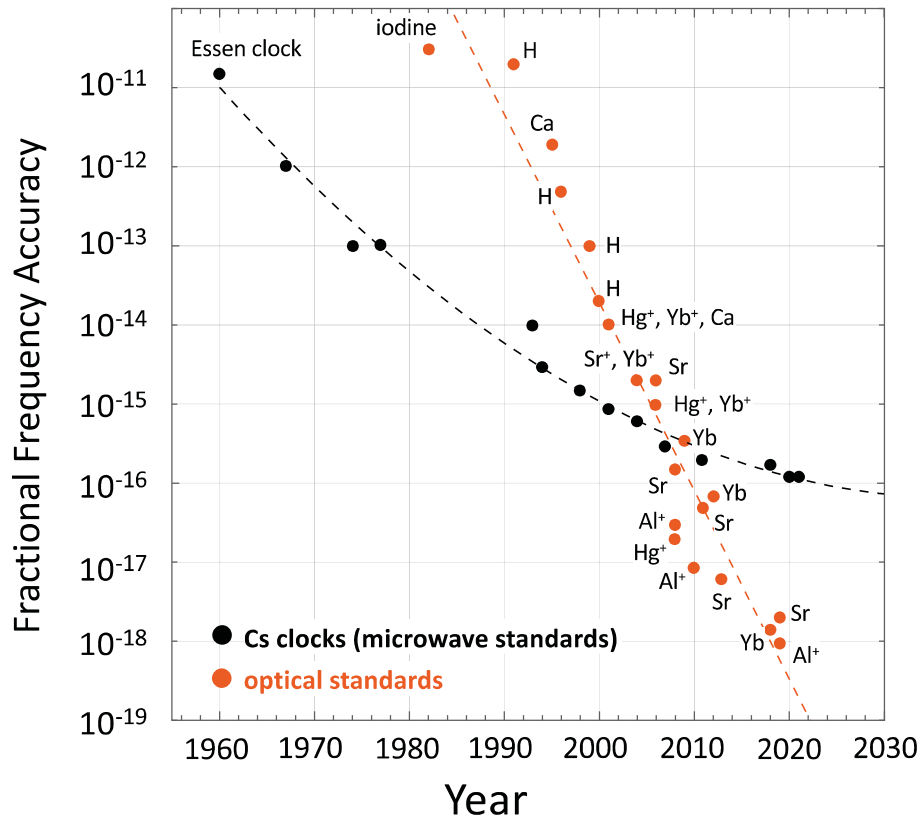


Figure 1: Historical progress of optical and microwave atomic references [4]. Optical clocks leverage their large transition frequencies to push past accuracy limitations experienced by microwave clocks near the 10^{-16} level. Since their first experimental demonstration in the 1980's, optical references have improved in performance with an average slope of 2.5 orders of magnitude per decade, or by a factor of 10 million since 1990! While their progress has yet to cease, optical clocks will encounter serious uncertainty limitations due to gravitational red shift as they near 10^{-19} .

compelling case for the international community to adopt a new definition of the SI second to an optical atomic transition with a fast-approaching target date of 2030 [8, 9, 10].

2. Frequency combs for clock synthesis and time/frequency applications

While optical atomic clocks have exceeded microwave clocks in performance, they are not without their challenges. Unlike microwave clocks, whose electronic carrier can be easily counted and conditioned using standard radio frequency electronics, optical carriers are far too fast to count using standard electronic counters and impossible to use as reference oscillators without the development of novel optical synthesis and

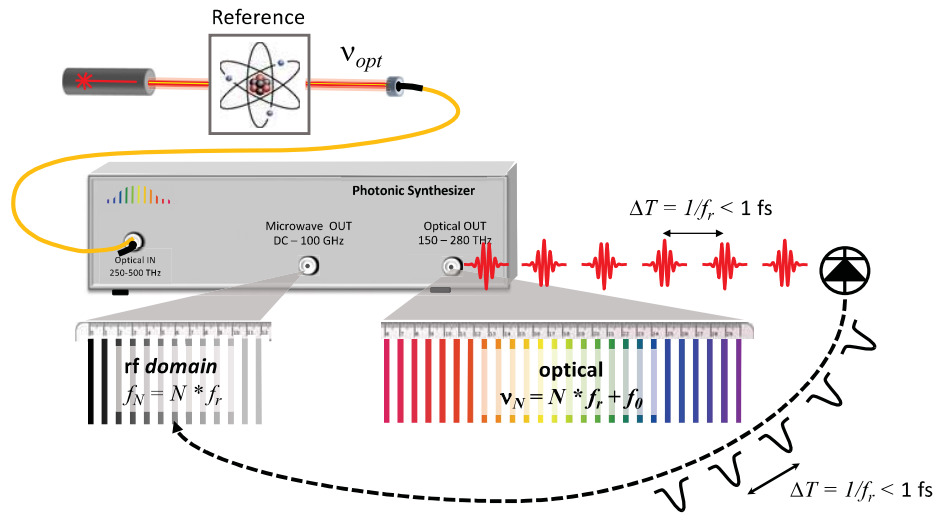


Figure 2: The frequency comb as a pulsed optical and microwave synthesizer. The frequency comb is based on a pulsed laser system. It outputs optical pulses with pulse-to-pulse period, T_r , and a corresponding optical spectrum of harmonically related modes with spacing, f_r frequency offset, f_0 , and optical mode numbers N . Here the mode number, $N \approx 300,000$, multiply f_r into the optical domain. The repetition rate, f_r , is the lowest order resonant mode of the laser cavity with a frequency that is usually ≈ 1 GHz. Photodetection of the optical pulse train derives an electronic pulse train with a corresponding rf spectrum represented by harmonics ($N = 1, 2, 3$, etc.) of f_r . The frequency comb can be stabilized to an optical reference using a self-referencing technique [11, 12], transferring the reference stability to the pulse-to-pulse timing, which in turn rigidly sets the frequency of the optical and rf modes of the comb spectrum with a deterministic frequency relationship to the reference frequency, ν_{opt} . In this way, the optical frequency comb takes a single frequency optical reference and broadly synthesizes optical and microwave modes that carry the references' stability.

conditioning techniques.

The frequency comb, developed at the turn of the century, helped fill this technological gap and simplified the measurement of, and vastly increased the application space of optical references [13, 14, 15]. Based on modelocked femtosecond lasers, optical frequency combs act like pulsed optical synthesizers with phase-coherent and harmonically related optical and microwave modes [11, 12, 16], see Fig. 2.

Since the early 2000's, frequency combs have been developed based on numerous fiber [17, 18, 19] and free-space laser platforms [20, 21, 22, 23, 24, 25, 26] enabling direct coverage of wavelengths spanning the visible to the mid IR [27, 28, 22, 23], see Fig. 3. When coupled with wavelength conversion techniques, such as difference frequency generation, second harmonic generation and high harmonic generation, frequency combs provide a convenient means to transfer the frequency and phase information

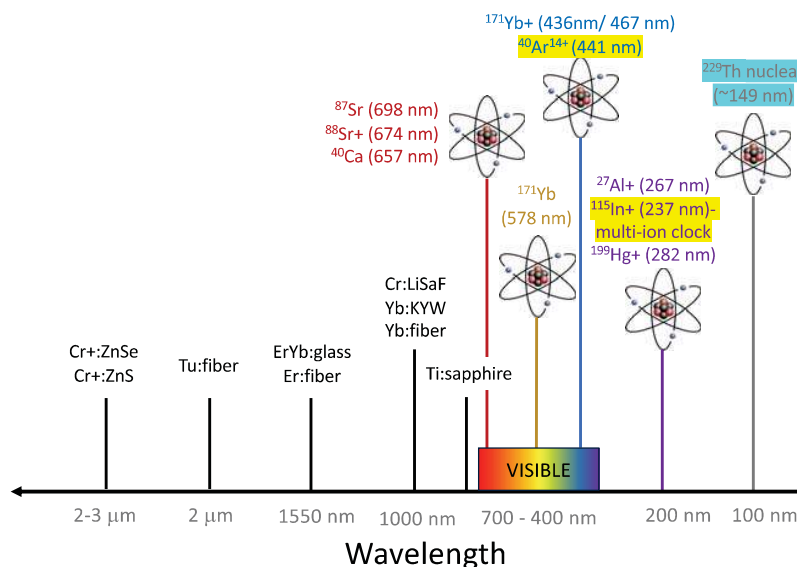


Figure 3: Frequency comb laser gain medium versus center wavelength. Also shown are a selection of optical atomic clock species versus transition wavelength. Highlighted are some of the newest species of optical clocks based on highly charged or multiple trapped ions (yellow), as well as a proposed frequency reference based on a nuclear transition in ^{229}Th (blue). For a list of recommended values of standard frequencies, see <https://www.bipm.org/en/publications/mises-en-pratique/standard-frequencies>.

of atomic references across much of the electromagnetic spectrum. Finally, a plethora of applications have been enabled by combining the stability and uncertainty of optical references with the synthesis abilities of optical frequency combs. These applications take advantage of the fs-level optical and microwave pulse timing stability, the optical and microwave frequency stability, and high phase coherence possible with optical frequency combs [14, 15].

2.1. Comb operation and frequency division of optical standards

Frequency combs were first developed to divide optical frequencies to the radio frequency domain to derive electronic signals from optical references. The derived signals are used to characterize the optical transitions against the ^{133}Cs primary standard [29] and to demonstrate continuity between the current and future definition of SI second to optical standards [8]. Moreover, the generation of microwave timing signals from optical atomic references will be central in future optical timescales used to define the SI second to optical atomic standards.

In the time domain, the act of modelocking creates a massively amplitude modulated optical carrier via the coherent addition of the 100,000 to 10^6 harmonically related longitudinal optical modes in the laser cavity. The resulting optical field

manifests as short bursts of light, representing just a few cycles of radiation for each round trip in the cavity. The resulting low duty cycle pulse train (pulse width ~ 10 fs – 1 ps, and pulse-to-pulse period of 0.1 - 10 ns) acts like an optical metronome with a pulse repetition rate, f_r of 100 MHz to 10's of GHz, depending on the free spectral range of the laser cavity.

As seen in Fig. 2, direct photodetection of this optical pulse train with high linearity and high-power diodes [30] can permit the derivation of an electronic pulse train with a short-term timing stability and uncertainty matching that of the optical atomic reference, $\Delta T_r/T_r = \Delta\nu/\nu_{opt}$ [31]. Spectral decomposition of this pulsed electronic signal yields harmonics of the frequency comb laser pulse repetition rate that can be counted using an electronic counter referenced to a ^{133}Cs primary standard. The latter measurements enable evaluation of the divided optical standard against the definition of the Hertz. The low residual instabilities and inaccuracies in optical-to-microwave conversion have been achieved despite distortion of the recovered electronic signals due to saturation and nonlinear effects experienced at high photo-carrier concentration per pulse in the photodiode active region [32, 33, 30]. The technique of optical frequency division, beyond the synthesis of microwave timing signals from optical references, also enables the generation of harmonically related and exceptionally pure microwave tones for use as local oscillators in systems that require high stability and low phase noise [34, 33, 31].

2.2. Clock characterization beyond the definition of the Hertz

When performing an absolute optical frequency measurement against a ^{133}Cs primary standard, the measurement is presently limited by the realization of the Hz, fractionally near 10^{-16} . To evaluate optical atomic clocks beyond this limit requires a comparison against a reference with similar or lower uncertainty, namely another optical atomic clock. Optical frequency combs facilitate such measurements using their broad harmonic optical spectrum to cover bandwidths of 100's of THz in the optical domain, connecting optical references operating at disparate frequencies. As seen in Fig. 3, while optical frequency combs usually operate with center frequencies lower than that of many of the current optical references, nonlinear optical techniques can convert clock probe laser frequencies from the infrared to the visible to connect the frequency comb spectra to optical atomic clock transitions. For developing ^{229}Th nuclear atomic clocks [35], high harmonic generation via semi-ionization and recombination of electrons within a noble gas of Xenon or Argon using amplified pulses from the frequency comb can be used to coherently extend comb sources from the infrared and visible to frequencies near 2 PHz [36, 37].

Optical atomic clock comparisons are performed by using the comb as an optical frequency ruler. By measuring the comb mode spacing, f_r the number of modes separating two clocks, $M - N$, and by measuring the difference frequencies between the atomic references and the nearest optical mode of the comb, a relative frequency

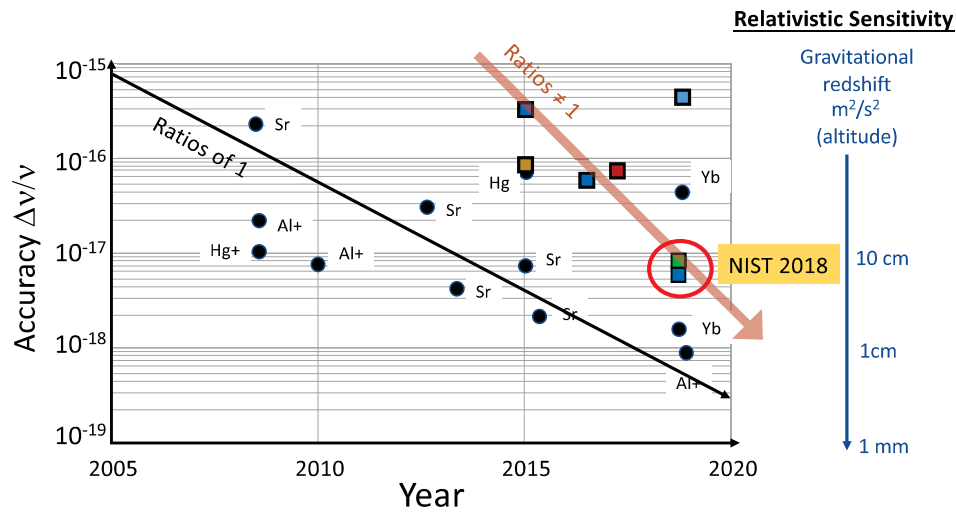


Figure 4: Selected results for clock self-evaluations, frequency ratios = 1, black circles, and inter-species clock comparisons, frequency ratios $\neq 1$, colored markers. Also shown is the relativistic sensitivity to earth’s gravitational potential in relative altitude difference.

measurement can be performed. Here, the common mode noise in the measurement of f_r (primary standard limitations in the definition of the Hertz) cancel when taking the ratio of two optical frequencies. It is important to note that the frequency comb yields negligible errors in the comparison of clocks. More specifically, the additive instability of the frequency comb in optical synthesis has been demonstrated at levels near 10^{-18} at 1-s averaging time, and with additive fractional frequency errors below 10^{-20} [38, 39, 40].

3. Next generation applications of optical atomic clocks

The highest performance atomic clocks currently report fractional frequency systematic evaluations near 10^{-18} [5, 6, 7]. These “self” evaluations are often assessed by measuring ratios of 1 between two atomic clocks of the same species, and in the same laboratory. These evaluations predict the uncertainty of a clock based on the level of control achieved over stray fields, the measurement error of the classical field sensors, and the theoretical and experimental assessment of how stray fields impact the clock transition frequency. As seen in Fig. 4, historically, the comparison of dissimilar species has yielded ratio results (ratios $\neq 1$) with worse measurement uncertainty than self-evaluations (ratios of 1). The observed disagreement is likely the result of common mode noise shared between like systems, or unassessed “dark” systematics for a particular clock [41].

Despite, the disagreement in uncertainty, optical atomic clock comparisons have achieved remarkable performance. At the current uncertainty levels, optical clocks can discern the effects of time dilation due to changes in altitude relative to

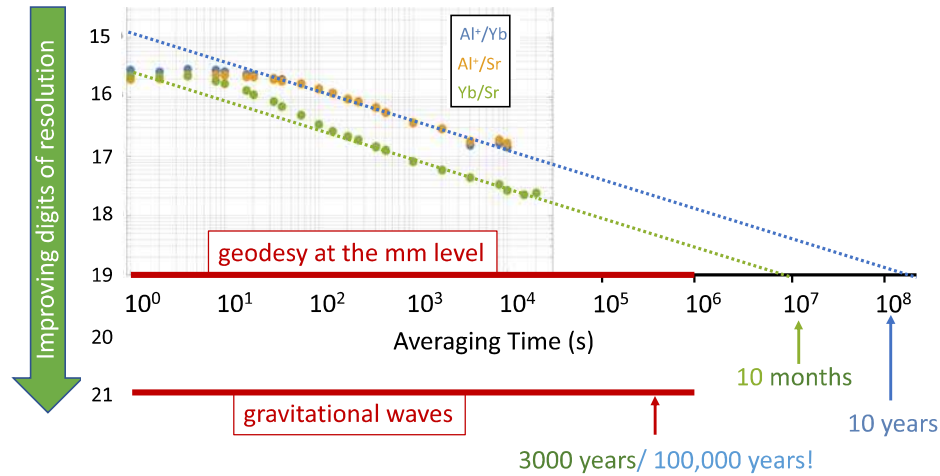


Figure 5: Allan deviation of the 2018 measurement instability for the ratios of the $^{27}Al^+$, ^{171}Yb and ^{87}Sr clocks at NIST [41]. Illustrated here are the averaging times needed to reach target resolutions for associated clock applications. Because optical atomic clocks exhibit Gaussian noise, the instability, or frequency resolution, improve as the square root of the averaging time. As seen in the blue, yellow and green data, a factor of 10 improvement in 1-s frequency instability results in a factor of 100 reduction in measurement time for resolution fractional frequency resolutions targets.

the earth’s gravitational potential at the cm-level [42, 43]. Because traditional measurements of the gravitational potential are currently limited near the mm-level (see https://geodesy.noaa.gov/library/pdfs/NOAA_TM_NOS_NGS_0077.pdf for the 2018 NIST geodesic survey results), earth-based optical clock comparisons will experience an uncertainty limit near 10^{-19} .

3.1. Clock comparisons for tests of fundamental physics

Beyond their sensitivity to relativistic geodesy [44], the electronic and nuclear spectra of atoms and ions reflect the value of our fundamental constants of nature and the physics internal to the atom. Shifts in the clocks transition frequencies reflect how external fields couple to these transitions. While clock comparisons have yet to reveal new physics beyond the standard model, they have provided constraints on the time variation of the fine structure constant at the 10^{-18} level and the proton-to-electron mass ratio below 10^{-17} [45]. Clocks have also been used to constrain violations of Local Position Invariance [45], and the coupling strength of ultralight dark matter to “normal” matter [41].

4. Differential laser spectroscopy of atomic clocks for improved measurement resolution

While much of the discussion regarding optical atomic clock is on improving their uncertainty, clock instabilities dictate the total measurement time needed to attain a target fractional frequency resolution for a specific application. In Fig. 5, we see that the traditional method (asynchronous frequency measurement) for comparing different optical clocks at NIST, would require between 3 weeks to 10 months of continuous averaging to reach a fractional measurement resolution of 10^{-19} !

4.1. Technical laser noise and its limitations to short-term clock instability

To address the measurement limitations in atomic clock comparisons, differential measurements can be used to reduce, or eliminate, noise from various technical sources. For instance, measurements at JILA have demonstrated differential frequency measurements of ^{87}Sr atoms within the same atom cloud. While this measurement achieved the highest instability measurement of the relative local geopotential [46], because the atoms shared the same environment and same laser, the measurement was highly common mode. While impressive, this level of common mode measurement is not possible for field tests of geodesy [43], or proposed gravitational wave detection over long baselines using remote clocks [47].

One of the primary limits to optical clock stability is the technical noise of the cavity stabilized probe laser. Laser noise is aliased onto the atomic measurement for clocks operating at probe duty cycles less than 100 % [48]. Additionally, drift of the optical reference cavity, used to pre-stabilize the probe laser, results in phase walk off between the probe laser and the atomic transition oscillations. The latter limits the maximum probe times, or free evolution time of the atoms, T_p , decreasing the measurement resolution for the atomic system.

To address this noise limitation, a common probe laser can be shared between atom clocks of the same species. Using this technique, two $^{27}\text{Al}^+$ clocks at NIST were compared, yielding close to a factor of 10 reduction in measurement time over two $^{27}\text{Al}^+$ clocks probed asynchronously with independent lasers [49]. This technique can be extended to dissimilar clocks by using an optical frequency comb to phase coherently transfer the probe laser frequency from one clock transition to another [50].

4.2. Differential spectroscopy of the $^{27}\text{Al}^+$ and Yb^{171} optical clocks

As seen in Fig. 6, the $^{27}\text{Al}^+$ and ^{171}Yb lattice clocks are synchronously compared at NIST by sharing the ^{171}Yb probe laser with $^{27}\text{Al}^+$ using an $\text{Er}/^{171}\text{Yb}$:glass laser frequency comb [51]. Because the quantum projection noise, and hence the stability of the $^{27}\text{Al}^+$ clock, is significantly limited by state detection from a single ion, the predominant route to higher stability is achieved via longer probe times. Using the $^{27}\text{Al}^+$ cavity stabilized laser, atom-laser decoherence limited the Al^+ probe time to 150

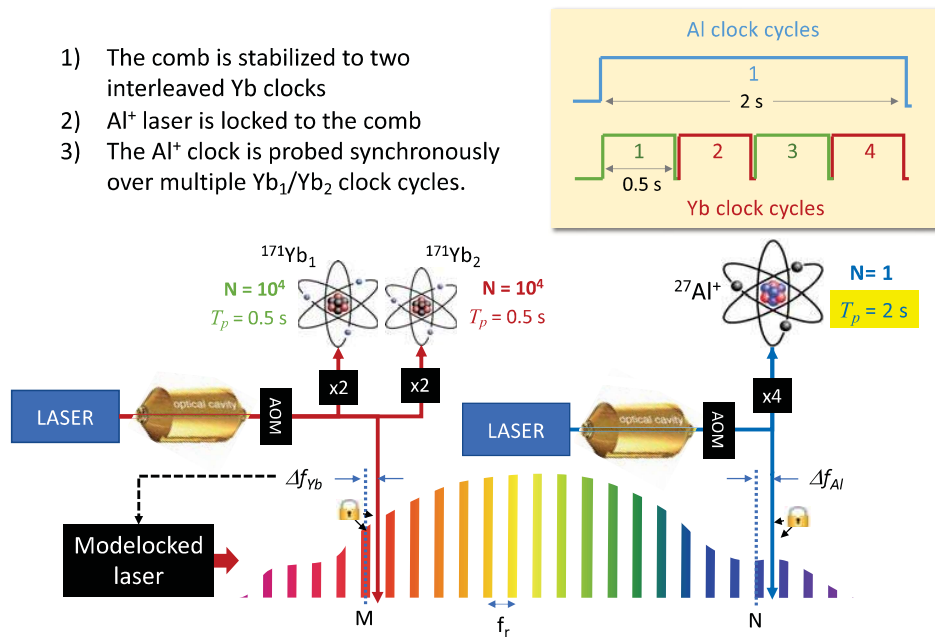


Figure 6: Experimental set up for realization of differential spectroscopy between two ¹⁷¹Yb lattice clocks, run in a zero-dead time configuration, and a single ion ²⁷Al⁺ clock. In this measurement, the Yb clock laser was coherently transferred to the ²⁷Al⁺ clock transition frequency using an optical frequency comb. The clocks were synchronously probed, with 4 Yb clock cycles combined to increase the Al⁺ probe cycle from 0.15 s to close to 2 s.

ms. Lower drift in the Yb cavity stabilized laser enabled a maximum atomic probe time for ¹⁷¹Yb of close to 0.5 s. While the latter represents a factor 3 improvement in probe time over spectroscopy using the cavity stabilized Al⁺ optical cavity, the upper state lifetime of ²⁷Al⁺ is 20.6 s [52].

To increase the ²⁷Al⁺ probe time beyond 0.5 s, two ¹⁷¹Yb clock clocks can be run in a zero-dead time configuration. By interleaving the operation of two ¹⁷¹Yb clocks, the phase walk-off of their cavity stabilized laser can be corrected every 0.5 s. Using this configuration, multiple cycles of the two ¹⁷¹Yb clocks can be combined to increase the ²⁷Al⁺ probe time to arbitrarily long times. However, the lowest observed 1-s measurement instability between the ²⁷Al⁺ and ¹⁷¹Yb clocks was achieved for an Al⁺ probe time of 1.7 s. One possible limiting noise source is phase walk-off between the ¹⁷¹Yb and ²⁷Al⁺ atom clouds induced by noise in the optical network, comprised of 10's of meters of fiber and free-space links, connecting the two atomic systems. While the stability of the links was established between cavity stabilized lasers as averaging down as $10^{-17} t^{-1/2}$, the cancellation of laser noise between systems requires low total integrated phase noise, not just high frequency stability.

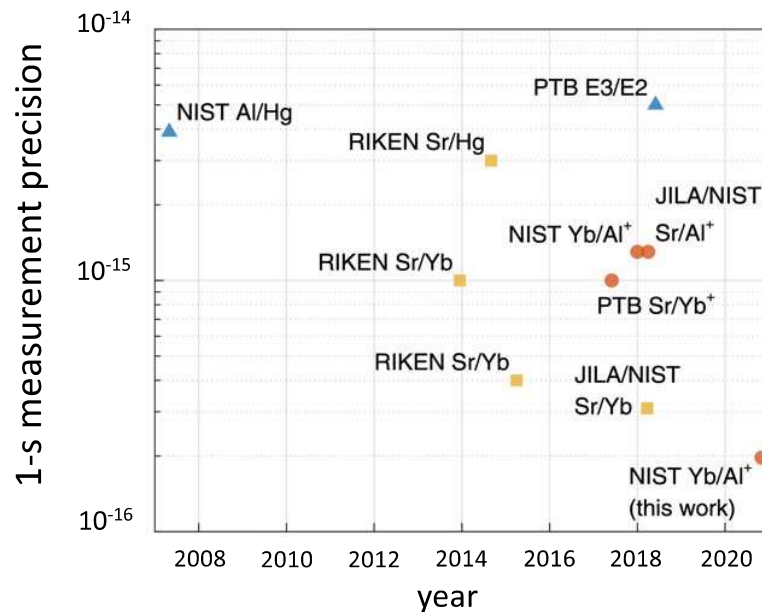


Figure 7: Selected 1-s measurement precision from NIST’s differential spectroscopy results as compared with previous inter-species optical atomic clock comparisons. Image credit: M. Kim *et al.* Nature Photonics **19** (2021) [50].

4.3. Results

Finally, in this measurement the clocks were run synchronously with close to 100% probe duty cycle, minimizing the effect of laser noise aliasing in the comparison of the two clock systems. The combination of increased $^{27}\text{Al}^+$ probe time (0.15 to 1.7 s) and the minimization of laser noise aliasing enabled nearly a factor of 10 improvement in the relative measurement instability of the $^{27}\text{Al}^+$ and ^{171}Yb clock near 2×10^{-16} [50], see Fig. 7. Aside from enabling the lowest instability inter-species clock comparison to date, differential spectroscopy permits a $100 \times$ speed up in total measurement time improving the feasibility of next generation atomic clock applications.

5. Conclusion and future outlook: the next 7 years.....?

Based on the current trends in optical atomic clock performance, Fig.4, one might expect researchers to report uncertainty results in 2030 near or below 10^{-19} . At this point, local clock comparisons performed over relatively short baselines will experience limitations due to the current uncertainty of traditional geodesic measurement techniques. Comparisons performed over continental differences will suffer from accumulated errors due to laser leveling and GPS uncertainty, currently at the 10^{-16} level.

The lack of access to dark fiber networks constitutes a significant barrier for the remote comparison of optical atomic clocks within North America and between North

America and the rest of the world. Transportable optical atomic clocks [53, ?, 54] will hopefully provide a means to connect continents and circumvent gravitational uncertainties due to earth's geoid. Additionally, ground to satellite free-space optical time/frequency transfer [55, 56, 43] will provide a means to disseminate signals at the level with an uncertainty better than 10^{-18} , which is not currently possible using GPS-based microwave time transfer methods.

Differential spectroscopy of clocks over longer fiber-based or free-space baselines can provide a means for improved measurement stability to ensure reasonable measurement times for upcoming applications in relativistic geodesy [43] and clock comparisons for gravitational wave detection. Such a measurement will encounter potential challenges in maintaining phase coherence $\Delta\phi_{rms} < \pi/2$ over the total measurement time for common mode rejection of laser technical noise over long links.

Waxing more speculatively, at the next Frequency Symposium will we see the beginnings of high-accuracy clock comparisons performed over quantum networks [57]? Finally, I look forward to seeing how well-established time and frequency techniques will be applied to problems in quantum technology.

6. Acknowledgements

I would like to acknowledge all the various contributors to the Boulder Atomic Clock Network collaboration (BACON) from the NIST timescale group, the precision photonic synthesis group, the Al⁺, Yb and Sr clock groups. I would like to specifically acknowledge the work of May E. Kim, William F. McGrew, Nicholas V. Nardelli, Ethan R. Clements, Youssef S. Hassan, Xiaogang Zhang, Jose L. Valencia, Holly Leopardi, David B. Hume, Andrew D. Ludlow and David R. Leibbrandt on the differential spectroscopy work. I would like to also thank Nick Nardelli who helped with Figures 1 and 5. Thank you also to Esther Baumann, Ladan Arissian and William McGeehee for the careful reading of this manuscript.

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