Is the time right for a redefinition of the second by optical atomic clocks?

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Abstract. Given the dramatic rate of progress in optical atomic clocks over the last decade, this paper presents the current state of play, and considers the possibilities, implications and timescales for a potential redefinition of the SI second in terms of an optical reference transition. In particular, the question of choice of a future standard is addressed, together with the requirements to accurately compare realisations of such standards, both for clocks local to, and remote from each other. Current performances of various optical clock systems are examined and possibilities for moving beyond potential limitations by alternative strategies are outlined.

1. Introduction

The microwave caesium atomic clock has been the basis for realisation of the SI second since 1967. From that time, there have been various clock technology advances, most significantly the advent of laser cooling and its application to cold caesium fountain clocks, the best of which now achieve systematic frequency uncertainties of ~ 1-2 x10^{-16} and report regularly to the BIPM. Fountain clocks provide the primary steering corrections for international atomic time (TAI) and universal co-ordinated time (UTC) [1, 2], with subsidiary sub-sets of commercial Cs clocks and hydrogen masers in national laboratories providing local dissemination of time and frequency for high technology and industrial applications. However, since the turn of the century, the pace of optical atomic clock R&D has quickened considerably, with result that several optical clock systems based on different atomic species now achieve lower systematic uncertainties than the best fountain clocks. Already fundamental science is benefitting in a number of cases from the lower uncertainties available from optical clocks, and the question of whether the time is right for a redefinition of the second becomes increasingly relevant. Whilst microwave fountain clocks will continue to underpin traceability for many microwave-based time and frequency applications in navigation, communications and synchronisation, limitations with these systems now arise due to the low microwave frequency and associated extended periods needed to reach 10^{-16} performance. Increasing development of optically-based platforms and architectures, however, can offer alternative and efficient routes and techniques for increased accuracy. Prime examples of this are recent demonstrations of femtosecond comb-based optical-to-microwave down-conversion to provide the microwave local oscillator for probing the Cs fountain [3-5].

2. State-of-the art optical clock performance

Those optical clock species now surpassing the Cs fountain capability include the \(^{27}\text{Al}^+, \quad ^{199}\text{Hg}^+, \quad ^{171}\text{Yb}^+\) and \(^{88}\text{Sr}^+\) trapped ion systems and \(^{87}\text{Sr}, \quad ^{199}\text{Hg}\) and \(^{171}\text{Yb}\) atoms in optical lattices, with reported systematic uncertainties in the 10^{-16} to 10^{-18} range. Benchmark frequency instabilities for single cold ions confined within rf traps, and multiple atom systems (~ 10^4 - 10^5 atoms) within optical lattices, are
Recent $^{87}$Sr lattice clock results report a relative clock systematic frequency uncertainty of $2.1 \times 10^{-18}$ [6], and statistical agreement between two cryogenic Sr clocks at $2 \times 10^{-18}$ [7].

| Atom / ion | Clock type          | Clock $\nu$ THz | Clock $\lambda$ nm | Lowest published clock systematic uncertainty | Uncertainty of CIPM $\nu$ value |
|------------|---------------------|-----------------|--------------------|---------------------------------------------|-------------------------------|
| $^{87}$Sr  | Lattice             | 429             | 698                | $2.1 \times 10^{-18}$ [6]                   | $5 \times 10^{-16}$           |
| $^{171}$Yb | Ion octopole        | 642             | 467                | $3.2 \times 10^{-18}$ [8]                   | $6 \times 10^{-16}$           |
| $^{29}$Al  | Ion, quantum logic  | 1121            | 267                | $8.6 \times 10^{-18}$ [9]                   | $1.9 \times 10^{-15}$         |
| $^{88}$Sr  | Ion quadrupole      | 445             | 674                | $1.2 \times 10^{-17}$ [10]                  | $1.6 \times 10^{-15}$         |
| $^{199}$Hg | Ion quadrupole      | 1065            | 282                | $1.9 \times 10^{-17}$ [11]                  | $1.9 \times 10^{-15}$         |
| $^{40}$Ca  | Ion quadrupole      | 411             | 729                | $3.4 \times 10^{-17}$ [12]                  | $1.2 \times 10^{-14}$         |
| $^{199}$Hg | Lattice             | 1129            | 266                | $7.2 \times 10^{-17}$ [13]                  | $6 \times 10^{-16}$           |
| $^{171}$Yb | Ion quadrupole      | 688             | 436                | $1.1 \times 10^{-16}$ [14]                  | $6 \times 10^{-16}$           |
| $^{131}$Yb | Lattice             | 518             | 578                | $3.4 \times 10^{-16}$ [15]                  | $2 \times 10^{-14}$           |
| $^1$H     | Cryogenic beam      | 1233            | 243                | $4.2 \times 10^{-15}$ [16]*                 | $9 \times 10^{-14}$           |

Table 1: Published systematic fractional frequency uncertainties of various optical clock species, and the fractional frequency uncertainties of the absolute frequency values adopted by the CIPM, following the 2015 Consultative Committee on Time & Frequency (CCTF); [16]*: the published uncertainty is that of the 2466 THz 1S-2S 2-photon transition, whereas the recommended CIPM value corresponds to the 1233 THz single photon frequency.

Table 1 shows the best published uncertainties (column 5) for various atom and ion clock systems. These data have resulted from the derivation of estimated uncertainty budgets for the different species under development at various national measurement and research institutes. This has been achieved by means of measured or theoretically-calculated sensitivities of the clock frequency to environmental perturbations (eg Stark shifts including black-body radiation and light shifts, Zeeman shifts, same-species density shifts and collisions with background gas, and for ions, electric quadrupole shifts) and cold atom/ion residual motional perturbations (eg 2nd order Doppler shifts). Most of these optical candidate-species for a redefinition have already been accepted (with the exception of $^1$H, $^{199}$Hg and $^{40}$Ca$^+$) as secondary representations of the second by the CIPM with a stated fractional frequency uncertainty, whereby associated operational clock data (stability, reproducibility and uncertainty) from these systems can now be reported to BIPM on a regular but preliminary basis in order to ascertain longer term performance prior to full contribution to UTC determination. Following the 2015 meeting of the CCTF, the fractional uncertainties shown in column 6 are now adopted by the CIPM. These latter uncertainties are larger than those published in the literature on two counts. First, the optical frequency absolute value has to be related to the current definition and its uncertainty, and, second, the Frequency Standards Working group (WGFS) of the CCTF adopts a more prudent approach when determining universal frequency values from very small data sets such as a measurement from a single laboratory, or a few laboratories, where a normal distribution of results is not available. In such cases, typically an enlargement factor of a few is applied, dependent on circumstances. The basis for such treatment is discussed in a publication in preparation [17].

With the current set of cold atom and ion species that surpass Cs fountain performance, the question of a redefinition of the second in terms of an optical clock species becomes increasingly pertinent. Of course, the caesium microwave standard has proved to be an enduring one over its ~ half-century reign, demonstrating improvements at the rate of a decade per decade during this time, and will undoubtedly continue to play a significant role for microwave applications going forward. However, the averaging
The time needed to achieve its current $10^{16}$ limiting uncertainty is unwieldy for many measurement applications, and with the perceived difficulties in further significantly reducing this uncertainty, these together clearly identify the higher frequencies of optical clock systems as the future way. The sub-set of species given in Table 1 is of course non-exclusive, and higher frequency options, such as the nuclear transition in $^{229}$Th$^+$, or VUV transitions accessed by high harmonic generation, may also come to the fore in future years. However, considering the current set of optical candidate species, there are some questions that need careful consideration in respect of a redefinition choice, and further ones in respect of when such a redefinition might occur. In the former case, should we decide on the candidate that demonstrates the lowest uncertainty, or alternatively choose the candidate that has collectively been studied most by the standards institutes? Right now, one observes that the candidate that fulfils both these criteria is the $^{87}$Sr optical lattice clock, but this may not remain the position going forward, which brings us to redefinition timescales. The progress in uncertainty reduction for many of the candidates is still fast improving, and we need to increase our knowledge of some perturbation sensitivities and limiting uncertainties for improved clock systems and operating algorithms. Further, it would seem unwise to make a redefinition decision until the current fast rate of improvement slows. Finally, it becomes most important to stress-test individual clock performance by real-time comparison of different clocks operated local to and remote from each other. One alternative suggestion to the selection of a “best choice” candidate has been the suggestion of a definition comprising a “universal” value determined from the matrix of individual best values for different species. However, in this scenario, the definition would not be based on a fundamental constant or a “constant of nature” (as in the current definition), but on an aggregation of values for different systems, with little physical significance. Further, this value would change every time individual values were upgraded, and change significantly with the introduction of a new species into the value matrix.

3. Derivation of high accuracy frequency ratios

Given the increasing number of optical clock species with systematic uncertainties lower than that for the Cs fountain clock, absolute determinations of their optical frequency values are at best constrained to that of the Cs fountain uncertainty. A more profitable approach is to directly compare optical frequencies ratios of the optical systems themselves. Here, all frequency comparisons can be quantified in terms of a matrix of frequency ratios, be they optical ratios, optical-microwave ratios (of which optical clocks measured relative to Cs are the most common case) or microwave ratios, and the WGFS of the 2012 CCTF opted to adopt this as a viable way forward to take advantage from the lower optical uncertainties. However, there are some interesting consequences arising. With a full matrix of frequency ratios, absolute frequency values for particular clock species may be derived by various routes eg from a single ratio or combinations of ratios, and this in effect over-constrains the determination of an optimised value for the clock transition frequency. A procedure to deal with this has been proposed recently [18], and is also reported in this Proceedings and should lead to a more statistically comprehensive approach. Nevertheless, at this time, we remain at the intermediate position of combining several Cs-related determinations with one or two all-optical ratios in order to determine values for some clock frequencies, with the Cs uncertainty providing the limitation. An example of this is the absolute value of the $^{27}$Al$^+$ clock transition, which is derived from both a Cs-related measurement of the $^{199}$Hg$^+$ transition and a subsequent $^{27}$Al$^+$/$^{199}$Hg$^+$ frequency ratio measurement. However, as the number of optical ratio determinations increase, one would expect that significance of the Cs-related values will be reduced within the matrix. It should be said it is still important to include the optical-to-caesium ratios in order to ensure that the matrix values remain consistent with the Cs-related values in order that a discontinuity larger than the Cs uncertainty does not occur at redefinition.

4. Optical clock comparisons

In preparation for a redefinition of the second, and given the range of high performance clocks of different species being researched in various labs internationally, it is highly desirable that individual clock systems are directly compared against each other in real time to provide independent validation,
and also to help determine whether there is a “best choice” redefinition candidate. Several NMIs are pursuing a number of clocks with different species and of course femtosecond combs now readily allow such local intercomparisons between different species clocks in the same institution, where the clock uncertainties are not compromised due to the comparison process, with comparison accuracies limited by the uncertainties of the optical clocks themselves eg.[19]. However, point-to-point direct intercomparison of remote clocks in different national laboratories remains a more rigorous goal allowing comparison of clocks of different designs, thereby reducing common mode effects that might arise in the design of clocks built in a single institution. There are a number of different techniques evolving for remote optical clock comparisons, and which are described in more detail in this Proceedings. These include microwave frequency transfer by satellite, optical frequency transfer via ground-to-satellite and satellite-to-satellite, portable optical clocks and optical frequency transfer by fibre using dark fibre or dark channels in internet-carrying fibre [20-22]. Microwave frequency transfer by satellite includes traditional 2-way satellite frequency transfer and GPS Precise Point Positioning (PPP) which are limited to comparison fractional accuracies of \( \sim 10^{-15} \) per day. A recent 2-way comparison using higher “chip” rates is discussed below, where comparison accuracies in the few x \( 10^{-16} \) are expected. With the launch of the ACES mission [23] in 2017, frequency comparison accuracies are expected to achieve \( 10^{-16} \) to \( 10^{-17} \) assuming lack of optical cycle slip over the 90-minute orbital period, which is increasingly demonstrated by high accuracy clock and comb combinations. On the optical side, free space optical transfer techniques between ground and satellite and satellite to satellite are still in their infancy, but proving experiments point towards accuracies of \( 10^{-16} \) per day. There is significant interest in building portable optical clocks for both ground and space operation. Here, there is generally a trade-off between highest clock accuracy and the compactness needed for portability, especially in space. On the ground, optical clock frequency variations due to clock height differences in the cm-range, due to gravitational red shifts, start to compromise clock evaluations. Finally, optical frequency transfer by fibre offers most promise, with frequency stability transfer at the \( 10^{-18} \) level in minutes already demonstrated over hundreds of km, dependent on link distance and the need to compensate for fibre phase fluctuations due to temperature and vibration. The main issues with fibre transfer is the point-to-point nature of the process, and the cost of accessing dark fibre or dedicated dark channels in fibre.

![Figure 1: NPL \( ^{171}\text{Yb}^+ \) octupole / \( ^{87}\text{Sr} \) frequency ratio measurement (colour on-line)](image_url)

Over the past couple of years, international real-time remote optical frequency comparisons have started in earnest. During late 2014, a frequency comparison of the quadrupole transition in single \( ^{171}\text{Yb}^+ \) ions clocks at PTB and NPL was performed using GPS PPP, with both clocks related to local hydrogen masers via femtosecond combs [24]. The level of agreement achieved was -1.3 (1.2) \( \times 10^{-15} \). During summer 2015, a major month-long intercomparison activity between NPL, PTB, SYRTE and INRIM was carried out within the Euramet project on international timescales with optical clocks (ITOC). Several trapped ion and optical lattice clocks at the labs contributed data, with all clocks referenced by comb to H-masers and data relayed via a geostationary SES Astra 3B satellite [25]. During the campaign,
opportunity was also taken to undertake local comparisons such as the $^{171}\text{Yb}^+$ E3 – $^{87}\text{Sr}$ ratio determination at PTB and NPL (figure 1). The campaign itself was a proving ground for the demonstration of optical clock “up-times” which reached in excess of 80% over the month for some clocks, although more work is needed to fully automate clock and comb operation. A large amount of intercomparison data was gathered, with final results expected in spring 2016.

In parallel with the satellite comparisons, international fibre links have been established between PTB and SYRTE, and NPL and LPL / SYRTE. Comparison of the Sr lattice clocks at PTB and SYRTE via the PTB-SYRTE fibre link has recently been reported [26], demonstrating $5 \times 10^{-17}$ agreement, and comparison of the NPL Sr and $^{171}\text{Yb}^+$ clocks with the SYRTE Sr clock is expected shortly, followed by PTB – NPL Sr and $^{171}\text{Yb}^+$ clock comparisons via the SYRTE / LPL fibre link. Figure 2 shows the recently-established NPL – SYRTE fibre link.

5. Next generation local oscillators and clocks
Here I briefly consider some potential developments that may enable better clock stability and reduced uncertainty beyond the current state of the art. On the local oscillator (LO) front, environmental isolation enhancements have led to optical reference cavities with ULE spacers and ultra-high finesse multi-layer dielectric mirror coatings on fused silica substrates achieving thermal noise limited frequency stabilities ~ few $10^{-16}$. Over the past few years, thermal noise has been pushed even lower by two different strategies. These include the use of single crystal silicon cavities operating at 124 K and achieving sub-40 mHz linewidths at 1.5 $\mu$m [27], and the use of semiconductor crystalline mirror coatings [28], both of which point to $< 10^{-16}$ thermal noise floor. Additionally, we have seen the introduction of the universal synthesiser concept, whereby a master oscillators locked to a very well isolated long ULE cavity (~ 50 cm – 100 cm) is used to anchor a fibre comb, eg [29], from which ultra-stable light from relevant comb modes can be selected to probe clock transitions in different cold atom and ion species. However, cavity, mirror substrate and coating designs look to be approaching limiting thermal noise conditions.

There are other optical local oscillator concepts also under consideration. One of these is spectral hole burning (SHB), for example in the absorption spectrum of a Eu$^{3+}$ doped Y$_2$SiO$_5$ crystal at 4.5 K. A series of spectral holes can be burnt with ~ 50 kHz separation and linewidths as low as 1 kHz with a very compact prototype cryogenic SHB module [30]. These holes are of course limited to particular absorption wavelengths in the crystal, and early research had to contend with the holes broadening and weakening over time, requiring a re-write process. However, a stable writing / probing process has now been established [31], with projected signal-to-noise limited stability reaching below $10^{-16}$ in 1 s.

Another interesting concept yet to be realised is that of a millihertz-linewidth super-radiant laser based on clock transition photons emitted from lattice-trapped cold atoms within a high-Q optical cavity.
The idea is to operate within the “bad-cavity” limit, where the cavity decay rate $\kappa$ is much greater than the atom spontaneous decay rate $\gamma$, giving rise to an atomic decay linewidth $\Delta \nu$ (atoms) much narrower than the laser cavity decay linewidth $\Delta \nu$ (laser). In this arrangement, energy is effectively stored within the atoms, rather than in the cavity, with result that the clock atoms should become insensitive to cavity length changes by a factor $\Delta \nu$ (atoms) / $\Delta \nu$ (laser). It is projected that super-radiant laser output powers of picowatts from $\sim 10^6$ atoms within the cavity should be enough for an atomic phase lock, whilst also avoiding significant cavity pulling effects.

On the cold atom clock front, there are also opportunities for improved clock stability. The first of these is already under study, and targets non-destructive phase measurement of the clock signal by retaining fractions of the cold atom sample through the Ramsey measurement process [33], rather than losing all the atoms during each measurement and having to cool, load and prepare a new sample for the next interrogation cycle. This should allow a reduction in interrogation dead time, thereby reducing the Dick effect. Alternative strategies include an intermediate stage of locking a cavity-stabilised local oscillator to a high signal-to-noise (SNR) cold atom sample in order to further discipline the LO before the high-accuracy probe of the clock atoms in the lattice, or extending this disciplining by a series of nested locks to a number of high SNR samples. One can also consider simultaneous probing of a number (m) of groups of N cold atoms, where one should expect the stability to reduce as a function of $1 / N^{m/2}$ [34]. Finally, as entanglement techniques become more robust, it can be shown that simultaneous probing of N entangled atoms [35, 36] allows one to surpass the standard quantum limit, and approach the Heisenberg uncertainty limit whereby the instability will scale as $(1/N)$ instead of the usual $(1/N^{1/2})$.

6. Conclusions
I have outlined the significant rate of progress currently occurring in the development of optical atomic clocks, and at this point there is no obvious sign of a slow-down in this rate of progress, with published fractional frequency uncertainty budgets ranging between $10^{-16}$ and $10^{-18}$. However, it is work in progress, with more national measurement laboratories and other research institutes are engaging in the development of such clocks. As clock stability improves for the various clock species, so we expect to see estimation of systematic uncertainties of frequency shifts due to perturbing fields also further reduce. This, together with improving methodologies of real-time local and remote clock comparisons and the adoption and generation of a clock frequency ratio matrix with increased data input, certainly brings us closer to the point where a redefinition of the second looks viable, but this is still at least some years away. One must also recognise that field perturbations will increasingly impinge upon a reducing uncertainty budget. As discussed, the earth’s gravitational field at “ground level” will give rise to frequency shifts of order $10^{-18}$ per cm height difference between clocks, and it’s variation both in terms of time and space are beginning to complicate matters. Experiments on Al$^+$ ion clocks [37] were able to show a gravitational red shift in the laboratory when one clock was raised some 30 cm. As we move to the $10^{-18}$ domain, are we able to discriminate shifts at this level as due to clock frequency drift, or are they local gravity variations? One solution is the idea of one or more master clocks in space orbit where gravitational shift is reduced. There is space agency interest in this possibility, but it will be – a decade before robust space-qualified optical clocks are available. Notwithstanding gravity, environmental control of the clocks to achieve frequency shift uncertainties below $10^{-19}$ due to the various electric, magnetic and collisional effects will get difficult and need both passive and active control strategies.

So is there a best clock in respect of the choice of atom or ion species? Quite probably the limiting uncertainties for different species will all be within an order of magnitude. Should we go for the best performing clock, or select the species that has seen the most research, or is easier to realise? Does it matter that some national laboratories do not operate a chosen “best” clock species? With local femtosecond comb comparisons and remote optical comparison by fibre, currently we have the high accuracy clock comparison capability that surpasses the uncertainties of the clocks themselves, so the traceability routes between a “primary” clock and other “secondary” clocks would not increase the inaccuracy of the latter.
Finally, the Cs fountain microwave clock will continue to provide traceability at appropriate levels for applications in the microwave domain for near future. However, one can point both to high science applications that can take advantage from lower optical clock uncertainties, and to emerging optical platform architectures that would benefit from a more direct linkage with optical clock capability. This can be seen from the industrial interest in optically-based systems as demonstrated by agencies such as ESA, NASA, DARPA, DSTL and the establishment of national and EU quantum technology initiatives to translate quantum research into quantum-based industries.

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