When should we change the definition of the second?

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The microwave caesium (Cs) atomic clock has formed an enduring basis for the second in the International System of Units (SI) over the last few decades. The advent of laser cooling has underpinned the development of cold Cs fountain clocks, which now achieve frequency uncertainties of approximately $5 \times 10^{-16}$. Since 2000, optical atomic clock research has quickened considerably, and now challenges Cs fountain clock performance. This has been suitably shown by recent results for the aluminium Al$^+$ quantum logic clock, where a fractional frequency inaccuracy below $10^{-17}$ has been reported. A number of optical clock systems now achieve or exceed the performance of the Cs fountain primary standards used to realize the SI second, raising the issues of whether, how and when to redefine it. Optical clocks comprise frequency-stabilized lasers probing very weak absorptions either in a single cold ion confined in an electromagnetic trap or in an ensemble of cold atoms trapped within an optical lattice. In both cases, different species are under consideration as possible redefinition candidates. In this paper, I consider options for redefinition, contrast the performance of various trapped ion and optical lattice systems, and point to potential limiting environmental factors, such as magnetic, electric and light fields, collisions and gravity, together with the challenge of making remote comparisons of optical frequencies between standards laboratories worldwide.

Keywords: atomic clocks; optical clocks; redefinition of the SI second

1. Evolution of atomic clocks

The second in the International System of Units (Le Système International d’Unités, SI) was last redefined in 1967 in terms of the ground-state hyperfine frequency interval of the caesium-133 ($^{133}$Cs) atom. This followed about a decade after Essen’s first demonstration of a Cs atomic clock in 1955 [1] using a thermal Cs beam, and other advances in high-resolution microwave spectroscopy [2]. Aside from these improvements in spectroscopic resolution, the redefinition was predicated on the increasing inadequacy of the previous definitions of the second in terms of Universal Time (UT; based on the mean solar day) and Ephemeris Time (ET; based on the period of rotation of the Earth round the Sun). These inadequacies related to variations in the length of the day at the millisecond level due to polar wobble and seasonal variations in the case of UT, and to difficulties in calculating the Ephemeris second from astronomical data. As a result, the rapid take-up of Cs clock technology quickly led to the

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1967 redefinition, following measurements between atomic time and UT/ET to ensure no step change of international atomic time (TAI) relative to the previous definitions. The 1967 definition has endured for over four decades, partly because the Cs clock uncertainty of $10^{-10}$ at inception was a significant improvement over the previous definitions, but also because atomic clock technology has evolved enormously up until the present day. In particular, the arrival of laser cooling and trapping techniques [3–6] played a strong role in aiding this evolution, particularly in enabling the use of much longer Ramsey interrogation periods (giving greater spectral resolution) with slow atoms than was possible with the thermal atoms in a viable atomic beam length. These techniques led directly to the first demonstration of the atomic fountain in sodium in 1989 [7], and quickly led to the first Cs fountain clock by 1991 [8]. These fountain clocks are currently used to realize the second to better than 1 part in $10^{15}$ [9–13]. Microwave fountain clocks are discussed in more detail in the paper by Campbell & Phillips [14] in this issue.

The atomic clock frequency instability $\sigma$ can be described generically in terms of the expression $\sigma = \Delta f / (f(S/N)^{-1/2})$, where $f$ is the frequency of the resonance, $\Delta f$ is its spectral linewidth and $S/N$ is the signal-to-noise ratio. For a Ramsey interrogation, this can be written as

$$\sigma = \frac{1}{2\pi f \sqrt{(NT_{\text{int}} \tau)}} \quad (1.1)$$

where $N$ is the number of atoms, $T_{\text{int}}$ is the Ramsey interrogation time and $\tau$ is the averaging time. From this, we can see the advantage of gaining lower instability by making use of as high a frequency as possible, and this has driven the long-standing interest of the frequency metrology community in developing optical clocks. The optical analogues of the microwave clocks are based on nearly forbidden optical transitions in atoms or ions, where the optical frequency is close to $10^{15}$ Hz, and the transition natural linewidth is approximately 1 Hz or narrower. With optical frequencies approximately $10^5$ times higher than those of microwave clocks and similar linewidths to the microwave case, this offers a significant decrease in clock instability at equivalent averaging times over the microwave case. For example, while the best microwave fountain clocks require averaging of several hours to reach $10^{-15}$ relative frequency instability, this can be achieved in tens of seconds in the optical case. This increased clock stability not only provides for a better time resolution, but also facilitates the determination of the systematic variations of the clock frequency due to environmental perturbations such as electric, magnetic and light fields. This underpins the comparison of clock transitions in different ion or atom species in order to evaluate the individual species potential as a candidate for a future redefinition of the second, where clock insensitivity to perturbations is a desirable goal. The technical realities associated with observing such narrow optical transitions include not only the need for laser cooling to remove the broad Doppler linewidth (hundreds of megahertz) that obscures the clock natural linewidth, but also the need for a high-stability narrowlinewidth probe laser to interrogate the narrow optical transition without loss of resolution. The atoms/ions also need to be trapped and located in a small region of space in order that the interrogation times can be long enough (approx. 1 s) to avoid interrogation time spectral broadening. It can be seen from equation (1.1)
that the instability is proportional to the inverse of the square root of the number of particles. Here, there is a divergence in the experimental methodologies for realizing the optical clock, dependent on whether one uses atoms or ions.

The ion clocks are primarily based on a single laser-cooled ion confined within a radiofrequency (r.f.) electromagnetic Paul trap [15] that approximates to the ideal of a single particle at rest isolated from its environment. The drawback to this approach is the low level of detected signal from the ion as the clock transition is driven. However, with good solid angle collection efficiency, it is now routine to detect every time the clock transition is driven by monitoring quantum jumps in the cooling transition fluorescence [16]. The clock atomic lineshape is built from the statistics of the quantum jumps as the clock laser is tuned, and the laser subsequently servo-ed to the lineshape by stepping the laser between the lineshape half-intensity points and correcting for any quantum jump imbalance observed.

The atom optical clock approach is an advance on the microwave fountain technique. In the fountain, the interrogation time is limited, in practice, to the height of the fountain, with the standard 1 m height resulting in a typical 1 s Ramsey interval between the interrogation of the slowed atoms on their way up and as they fall back. In an atom-based optical clock, this difficulty is overcome through the use of an optical lattice to confine the atoms within the interrogation region [17]. The lattice comprises a standing wave in one, two or three dimensions, which provides for sub-wavelength optical trapping sites to hold many cold atoms. Ideally, the aim is to hold only one atom in a trapping site, in order to remove any possibility of collisional shift. The lattice trapping light field is a huge perturbation to the clock atoms, resulting in very significant AC Stark shifts of the upper and lower levels of the clock transition. However, a far off-resonant ‘magic’ wavelength for the lattice laser is used, whereby the AC Stark shift of the upper and lower levels can be set equal to good precision [17]. This allows many atoms to contribute to the clock signal, which should improve the signal-to-noise ratio (and hence stability) by a factor \( \sqrt{N} \), where \( N \) is in the range \( 10^4 \)–\( 10^6 \), compared with the single-ion case. At this point in time, the anticipated improvement has not yet been demonstrated, mainly due to other limitations associated with the local oscillator stability floor. Also, as discussed later, understanding of the frequency shifts associated with the lattice fields and collisional dynamics is still evolving. Currently, the systematic uncertainty for the leading ion system is about an order of magnitude lower than that for the neutral atom lattice clocks.

Figure 1 shows the evolution of frequency standards since the first Cs clock. The microwave clocks have shown steady improvement over the last half century to reach present-day uncertainties of approximately \( 5 \times 10^{-16} \). However, optical clock development has exhibited a significantly faster rate of improvement, particularly so over the last decade, owing to the invention of the femtosecond comb [18,19], which allows for fast and efficient optical–microwave frequency comparison, essential to relate the optical standards to Cs microwave standards. One can also see that, for some optical systems (e.g. \( \text{Al}^+ \), \( \text{Hg}^+ \) and Sr), the total uncertainty of the optical standard, derived in part from measurement of some systematic shifts, and estimation of others, is below that of the Cs primary standard. Since these optical standards can be intercompared using a frequency comb to well below the Cs uncertainty, this calls into question the usefulness of comparing these optical standards against the current primary standard. Such a situation is not ideal and forms the basis for the argument for a redefinition.

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in terms of the frequency of an optical clock. The International Committee for Weights and Measures (Comité International des Poids et Mesures, CIPM), on advice from the Consultative Committee for Time and Frequency (CCTF), has already anticipated the potential need for a redefinition by introducing the concept of secondary representations of the second [20]. This allows for the setting up and study of other microwave and optical frequency standards whose uncertainties can be shown to be within an order of magnitude of the primary standard, and there are already four optical systems accepted. One should note that the absolute uncertainty of the measured frequency of any of these secondary representations can be no better than that of the current primary Cs-based realization of the unit hertz (Hz). There are now opportunities for the frequency data from these representations to be regularly reported to the International Bureau of Weights and Measures (Bureau International des Poids et Mesures, BIPM) for comparison, in the first instance, against the Cs primary standards.

2. Possible algorithms for a redefinition of the second

In this section, the different conceptual proposals for a redefinition of the second are briefly considered. The possibilities arising include a definition based on a fixed value for the Rydberg constant, the choice of a single optical clock transition, the combination of a single optical transition and a set of optical frequency ratios, a ‘virtual’ frequency based on a matrix of ‘best’ optical transitions, or a very high-frequency transition in the vacuum ultraviolet or X-ray regions of the spectrum. Each of these options is considered in turn.

The atomic energy level structure that can be calculated with best accuracy is that of the hydrogen atom, on account of its relatively simple combination of a single proton and single electron. A non-relativistic description of the hydrogen atom gives rise to energy levels $E_n$ that are described by

$$E_n \approx -\frac{hcR_\infty}{n^2},$$ (2.1)
Redefinition of the second

where \( n \) is the principal quantum number, \( c \) is the speed of light, \( h \) is Planck’s constant and \( R_\infty \) is the Rydberg constant, given by

\[
R_\infty = \frac{m_e e^4}{8\varepsilon_0^2 h^3 c},
\]

where \( m_e \) is the mass of the electron, \( e \) is its electronic charge and \( \varepsilon_0 \) is the permittivity of free space. The Dirac equation extends the analysis to a full relativistic description, but still requires further adjustment via quantum electrodynamics (QED) theory to account properly for the Lamb shift in order to agree with direct experimental measurements of the 1S–2S transition frequency. Currently, the measurement of this transition by laser spectroscopy, relative to the Cs primary standard, has an uncertainty of 1.4 parts in 10^{14} \[21\], and, when combined with measurements of other hydrogen transitions \[22\] to account for the Lamb shift, gives a Rydberg constant value with the lowest uncertainty of 6.6 parts in 10^{12}.

This offers the possibility to redefine the second by fixing the Rydberg at its measured value and computing the hydrogen 1S–2S transition, or other hydrogen transitions. The difficulty with this is loss of accuracy of the 1S–2S transition relative to current Cs performance, and that between the measured value of 1S–2S and the Rydberg constant, which arises due to uncertainties in QED theory corrections and knowledge of the proton size. This latter issue is the subject of the paper by Nez \[23\] in this issue.

As a result, this option for redefinition does not seem competitive with the much lower uncertainties now quoted for cold atom and ion clock frequencies, and requires significant improvement in knowledge of the QED corrections. In addition, hydrogen would need to be trapped and cooled, which is more difficult on account of its small mass, and has correspondingly large second-order Doppler shifts. Generating the cooling radiation at 121.5 nm is also difficult.

Next, I consider the definition based on an optical clock transition in a cooled atom or ion. From §1, it is seen that \( ^{27}\text{Al}^+ \), \( ^{199}\text{Hg}^+ \) and \( ^{171}\text{Yb}^+ \) single trapped ion systems \[24–26\] and \( ^{87}\text{Sr} \) and \( ^{171}\text{Yb} \) atoms in a lattice \[27,28\] have already demonstrated systematic uncertainties below that of primary Cs. In the \( ^{27}\text{Al}^+ \) case, a relative frequency uncertainty of approximately \( 10^{-17} \) has been achieved, together with an uncertainty of \( 10^{-16} \) for \( ^{87}\text{Sr} \) atoms in a lattice. However, it must be acknowledged that there are a number of additional ions and atom species that are expected to demonstrate similar uncertainties in the near future. In view of this, there arises the question of how to choose the ‘best’ transition, and what criteria should be used to inform this choice. Should the selection of the species be based on the clock exhibiting the lowest uncertainty, which may be that of a species requiring more technically challenging operating arrangements, such as the \( ^{27}\text{Al}^+ \) quantum logic clock? This system makes use of quantum information techniques to operate as a clock, and this has only been achieved in one laboratory to date. Alternatively, the outcome from selection could be the choice of the \( ^{87}\text{Sr} \) optical lattice clock, on the basis that it is under development in some seven laboratories at this time.

It is likely that there will be a number of ion and atom clocks with uncertainties within a factor of 10 of each other. In this scenario, there would be no difficulty in selecting a lowest-uncertainty single ion or atom primary optical standard,
together with a set of secondary representations of the second based on other cold ion and atom species with uncertainties within a factor of 10 of the primary choice. Given the demonstrated capability of frequency combs to intercompare optical frequencies in the same laboratory at the $10^{-19}$ level or below [29], traceability to the definition could be readily realized via this comb transfer process. This would allow either the primary standard or any one of the secondary representations to be used to disseminate time and frequency within the major frequency standards laboratories, and to secondary laboratories and research institutes. This option would appear to provide the most accurate and efficient metrology infrastructure, provided suitable methods for high-accuracy remote clock comparison between national measurement institutes can be introduced (see §6). On this basis, further discussion is delayed until the following sections.

An equivalent optical clock definition would be to define a set of measured frequency ratio values between the major optical clock species. Again, this relies on the femtosecond comb intercomparison capability to relate the different species clock transitions. This option is equivalent to the primary/secondary case above, provided the respective ratios are anchored to an optical ‘primary’ standard. It becomes more problematical if the Cs microwavestandard is retained, as there arises the potential for a disconnect between data derived via the ratio value and Cs-related measurements.

An alternative option for definition is via a ‘matrix’ value algorithm. Here, the most accurately measured values for individual species would be combined to provide a set of frequency ratios and a weighted mean value of the frequency ‘matrix’ at redefinition. A typical set of high-accuracy optical standards is shown in figure 2. Subsequently, individual values could be updated for bias corrections and improved uncertainties at appropriate times. The drawback to this idea is that the matrix value would have no physical significance. It also would require regular updating, and acquire significantly different values with the introduction of new frequencies.

One final area for consideration is that of very high-frequency transitions in the vacuum ultraviolet (VUV) or X-ray/extreme ultraviolet (XUV) regions. This consideration is predicated on the push to even higher frequencies, giving rise to yet higher line $Q$ factors. Against this, of course, must be considered the technical difficulties in accessing these regions, and developing narrow-linewidth sources capable of operation there. Aside from the spectroscopic routes developed to probe the H $1S–2S$ transition, recent research has demonstrated high harmonic generation of visible frequency combs [30,31] as a possible way of comparing frequencies in the VUV and XUV regions with optical frequencies. The precision with which this might be carried out, and the experimental logistics of operating in these regions, are still a matter of study.

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In such high-energy regions, there arise possibilities to excite nuclear γ-ray transitions in addition to electronic X-ray transitions. Such nuclear transitions should have very low natural linewidth in the microhertz range, potentially giving rise to line $Q$ factors of $10^{20}$, although these can be difficult to observe on account of the recoil process in free atoms and the large associated Doppler shifts. This problem has been avoided in the case of Mossbauer transitions [32], whereby the transition is observed between nuclei in a solid such that the recoil is absorbed by the lattice structure. Over the last several years, interest has arisen in the possibility of using the thorium radio-isotope $^{229}$Th as a nuclear clock. $^{229}$Th has an unusually low-level nuclear isomer state, which is only approximately $7.6 \pm 0.5$ eV above the ground state [33], corresponding to a wavelength of 163 nm or 1840 ($\pm 120$) THz. There are proposals to drive the nuclear transition in $^{229}$Th$^{3+}$, both in a solid-state crystal [34] and in an isolated ion confined within an r.f. trap [35]. In general, it is expected that the nuclear transition is better shielded than electronic transitions from environmental effects. In the former case, recoil would be reduced via the solid lattice, but account would need to be taken of the strong crystal field coupling. For the isolated trapped ion case, the ion could be laser-cooled to remove the Doppler broadening, and many of the normal shifts encountered with electronic transitions are much reduced or not present. However, the main issue at present is the search to locate the transition. Currently, there is a large 242 THz region of frequency space to scan with a narrow-linewidth UV source. Double-resonance quantum jump techniques have been proposed to detect the nuclear transition [36], whereby a strongly allowed electronic dipole transition is probed while searching for the nuclear transition. As the nuclear transition is driven, there will be a change in nuclear moment and spin, resulting in changes to the electronic hyperfine splittings and angular momentum, and subsequent loss of electronic resonance. Taking this option to maturity will take time, even once the transition is located.

3. Optical clock architectures and candidates

The basic elements of an optical clock comprise a high-stability narrow-linewidth laser acting as a local oscillator (LO), which interrogates a narrow absorption in an atomic reference, and a femtosecond comb that acts as the frequency counter. A simplified diagram of the clock component inter-relation is shown in figure 3.

The laser local oscillator comprises a diode, fibre or solid-state laser that is pre-stabilized to an ultra-stable reference cavity using the Pound–Drever–Hall (PDH) technique [37]. The cavity makes use of an ultra-low-expansion (ULE) glass spacer that, close to room temperature, has a zero coefficient of thermal expansion, to which high-finesse ULE or silica mirrors are optically contacted. The cavity is temperature controlled within a vacuum enclosure with further isolation via vibration isolation platforms and acoustic damping. Recently, reference cavity designs have evolved to minimize vibration sensitivity [38–43]. Laser linewidths approximately 0.2–1 Hz and stability floors in the region of $2 \times 10^{-16}$ to $10^{-15}$ at time scales of 1–10 s are achieved, with the limit primarily due to thermal noise.

There are a significant number of optical atomic reference transitions currently in the frame, distributed among the two generic categories of electromagnetically trapped single ions and multiple atoms trapped in optical lattices. Once
Figure 3. Optical clock architecture showing the main components. EOM, electro-optic modulator; DBM, double-balanced mixer. The end-cap trap is just one example of an r.f. ion trap. (Online version in colour.)

Table 1. Distribution of ion (left) and atom lattice (right) clock species between the various national measurement institutes (NMIs) and research groups: NIST, National Institute of Standards and Technology, USA; NPL, National Physical Laboratory, UK; NRC, National Research Council Canada; MIKES, Mittatekniikan Keskus (Centre for Metrology and Accreditation), Finland; PTB, Physikalisch-Technische Bundesanstalt, Germany; UIbk, University of Innsbruck, Austria; NICT, National Institute of Information and Communications Technology, Japan; UProv, University of Provence, France; JILA, University of Colorado and NIST Joint Institute for Laboratory Astrophysics, USA; SYRTE, Systèmes de Référence Temps-Espace. France; Tokyo, University of Tokyo, Japan; NIM, National Institute of Metrology, China; UFlo, University of Florence, Italy; NMIJ, National Metrology Institute of Japan; KRISS, Korea Research Institute of Standards and Science; INRIM, Istituto Nazionale di Ricerca Metrologica, Italy; UDuess, University of Düsseldorf, Germany.

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<th>$^{40}$Ca$^+$</th>
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Doppler broadening has been removed by laser cooling, and suitable control of environmental fields achieved, cold ion or atom experimental linewidths in the region of 1–10 Hz can be observed. Natural linewidths for these systems...
range from 20 Hz to 1 mHz (with one octupole transition with a theoretical nanohertz linewidth). However, the effective linewidth is generally determined by the transform limit of the LO probe time (typically in the range 0.05–1 s).

Table 1 shows the various ion and atom candidate species under investigation at the various standard laboratories and research institutes. Research is very much work in progress and covers several ion species, but is concentrated primarily on two atom species. This highlights the need for further extensive evaluations of the comparative stability, uncertainty and reproducibility of each system before a redefinition of the second is made. The current state of the art is discussed in §5.

Neutral atom optical lattice clocks are discussed in another paper in this issue [14]. In the following section, I therefore concentrate on the state of research of single ion optical clocks.

4. Single trapped ion optical clocks

Single ions can be confined within a time-varying r.f. quadrupole potential, the basic form of which is the r.f. Paul trap. This comprises a ring and two end-cap electrodes situated either side of the ring on the ring axis. Miniature versions of this trap have approximately 1 mm ring diameter. With an r.f. voltage of a few hundreds of volts applied between the end-cap and ring, an oscillating saddle potential is set up, thereby creating a time-averaged pseudo-potential well. Drive frequencies in the range 10–70 MHz are typical, with resulting pseudo-potential radial and axial ‘secular’ frequencies of approximately a few megahertz. Additional small DC voltages are applied either to one or to more of the electrodes, and to auxiliary electrodes, in order to push the single ion to the zero potential at trap centre. This is necessary to minimize the ‘micro-motion’ at the drive frequency, which is increasingly prevalent if the average position of the ion is shifted from this zero position. Loading a single ion into the trap is achieved by the simultaneous ionization of a very weak atomic beam, generated from a small tubular oven, and laser cooling on a strong dipole transition connected to the ion ground state. Ionization can be achieved by either electron beam bombardment or photo-ionization, and in recent years the latter has become the method of choice. Single ion loading requires very weak atomic beam flux crossed with a weak ionizing intensity in order to minimize the generation of surplus charge and patch potential build-up on the electrodes. Laser cooling of the trapped ion achieves an ion temperature of approximately 1 mK, close to the Doppler cooling limit for the cooling transition. This corresponds to a distribution over motional state quantum numbers of approximately 10–20 in the trap. In this arrangement, an ion can be confined at trap centre to the Lamb–Dicke region [44] of dimensions less than $\lambda/2\pi$, whereby the first-order Doppler shift is removed and the second-order shift is minimized. Additionally, electric field perturbations are minimized at trap centre, and, under ultra-high-vacuum conditions, background collisions are minimized. This provides the scope for searching and probing high-\(Q\) optical clock transitions with long interrogation times of about 1 s. The ion thus approximates to an isolated, unperturbed particle, virtually at rest.

Phil. Trans. R. Soc. A (2011)
Doppler cooling and state prep. transitions

Figure 4. Partial term scheme of the $^{171}$Yb$^+$ ion. (Online version in colour.)

There are a number of variants to the r.f. Paul trap design that are also used for ion clocks. These include the end-cap trap, which comprises two inner end-cap electrodes with two coaxial outer electrodes that replace the ring functionality, and the r.f. linear trap, which has a parallel four-electrode geometry with r.f. and ground separately applied to the two diagonally opposite electrode pairs. The latter provides a radial trapping geometry to ions on the cylindrical axis, but requires two low-voltage DC electrodes on-axis to confine the ions in that direction. This arrangement is used for the quantum logic trap in order to confine the separate-species ion pair. In addition, there have been a number of micro-trap developments, where a linear trap arrangement has been constructed using lithographic techniques on alumina and silicon wafers.

(a) $^{171}$Yb$^+$ optical clock

One particular class of ion clock transition is the $^2S_{1/2} - ^2D_{5/2}$ quadrupole transitions in $^{171}$Yb$^+$, $^{199}$Hg$^+$, $^{88}$Sr$^+$ and $^{40}$Ca$^+$, which have lifetimes ranging from 0.05 to 1.1 s. I concentrate on the $^{171}$Yb$^+$ ion on the basis that this ion clock is representative of a clock transition in the visible region, has contributed significant data to date and also has a second clock transition in the form of the highly forbidden $^2S_{1/2} - ^2F_{7/2}$ octupole transition. The partial term scheme for $^{171}$Yb$^+$ is shown in figure 4. Laser cooling is effected on the $^2S_{1/2} - ^2P_{1/2}$ 369 nm dipole transition. Interruptions to the cooling cycle by occasional decays into the $^2D_{3/2}$ metastable level are recovered by driving out of this level with a 935 nm diode laser.

The 435 nm $^{171}$Yb$^+$ $^2S_{1/2} - ^2D_{3/2}$ quadrupole clock transition is driven via a frequency-doubled extended-cavity diode laser at 871 nm, stabilized to a high-finesse ULE reference cavity. It has a natural linewidth of approximately 3.1 Hz. Experimental cold ion linewidths of 10 Hz have been observed [45], though linewidths of a few tens of hertz are more routinely employed [46,47].
Redefinition of the second

outlined above, the D3/2 multiplet can also be accessed during the cooling cycle. Thus, a suitable D3/2 upper level for the clock transition, which is not connected to those D3/2 levels involved in the cooling process, is required. This is the D3//2 F = 2 level, which can also be driven from the ground state via a magnetic-field-insensitive mF = 0 → mF = 0 transition. The clock transition has been measured to an uncertainty of 0.73 Hz relative to Cs or 1.1 × 10−15 [26]. Comparison of the quadrupole clock in two separate traps has demonstrated frequency differences of less than 0.2 Hz (3 parts in 1016) over a few hours, with frequency instabilities (Allan deviation) of approximately 10−16 at 10,000 s averaging [26].

The second optical clock transition in 171Yb+ is the 467 nm 2S1//2–2F7//2 octupole transition. This is an extremely forbidden ultra-weak high-Q absorption brought about by the existence of a very-long-lived low-lying metastable F7//2 level in the ion spectrum, with an upper level measured lifetime of approximately 6 years [48], and corresponding natural width in the nanohertz range. It is not possible to take full advantage of the associated theoretical Q of approximately 1023, as this would require interrogation times equivalent to the lifetime and a local oscillator far narrower than is currently possible. However, it allows the use of relatively long interrogation times (in excess of 1 s) with associated Fourier-limited linewidths equal to state-of-the-art subhertz probe laser widths. Such long interrogation times lead to frequency correction cycle times in the region of 10–100 s to build adequate quantum jump statistics. This requires good ULE cavity drift compensation algorithms to ensure that the local oscillator probe can be held on-resonance long enough to make the probe frequency servo correction.

The long lifetime of the F7//2 level is also a problem in that the level acts as a sink for the ion, and, in repetitive frequency interrogation of the octupole transition, it is necessary to quickly recover the ion from the F state. This is achieved by additional laser wavelengths at 638 nm or 760 nm. Coherent population trapping and optical pumping in the ground state and D3/2 level during the cooling cycle, with magnetic fields close to zero, result in loss of fluorescence. Polarization switching is used to avoid ground-state population trapping. The octupole transition also has an mF = 0 → mF = 0 component, which is first-order Zeeman-insensitive. Transform-limited cold 171Yb+ octupole linewidths of 7 Hz have been observed (N. Huntemann 2011, private communication). The second-order Zeeman shift has been measured to be −1.72(3) mHz μT−2 [49], small for fields of approximately 1 μT. With magnetic shielding, second-order shifts below 1 mHz should be achievable. The electric quadrupole shift, which results from interaction of the electric quadrupole moment with residual field gradients in the trap, has been measured recently to be −0.035 ea02 (N. Huntemann, private communication). This is nearly an order of magnitude below the calculated value [49] and is the lowest reported value for those ions with quadrupole moments. Currently, the largest shift associated with the octupole transition is the AC Stark shift due to the relatively large 467 nm probe laser intensity needed to drive the transition. The AC Stark shift has a coefficient of 48(10) μHz W−1 m2 [50]. In order to take account of the AC Stark shift, it is measured as a function of laser power and extrapolated to zero power to provide the unshifted value. This is a significant correction for large linewidths, but becomes less dominant as the probe laser linewidth is reduced.
towards the 0.1 Hz level, on account of the reduced intensity needed to provide sufficient spectral intensity to drive the transition [49]. Alternatively, the new technique of hyper-Ramsey interrogation, where the phase and detuning of the second Ramsey pulse are varied relative to the first, could be used to reduce the magnitude of the shift [51].

One particular advantage of the $^{171}$Yb$^+$ double optical clock is in the search for time variation in the fine-structure constant $\alpha$. The ratio of the quadrupole and octupole clock frequencies has the highest sensitivity factor of 6.8 [52] of currently available clock transitions for this search. Measurements of this ratio at the 1 part in $10^{17}$ level uncertainty, made 1 year apart, would provide sensitivity for a rate of change of $\alpha$ below $2 \times 10^{-18}$ per annum. Additionally, if these measurements are made with the same ion in the same trap, there will be an exact equivalence of the perturbing fields due to the environment, and some common mode rejection of the systematic shifts.

(b) $^{27}$Al$^+$ quantum logic clock

Ions of the IUPAC (International Union of Pure and Applied Chemistry) group 13 of the periodic table include $^{10}$B$^+$, $^{27}$Al$^+$, $^{115}$In$^+$ and Tl$^+$. These all have clock transitions in the deep UV. While this presents some difficulty in generating clock laser light at those wavelengths, there are significant advantages in that the $^1S_0$–$^3P_0$ clock transitions have narrow natural widths (for $^{27}$Al$^+$ it is 8 mHz), do not have a quadrupole shift and have very small blackbody shifts. However, the main problem is that the strongly allowed electric dipole cooling transitions are even further in the VUV. The dipole transition in $^{27}$Al$^+$ is at 167 nm. In order to use these species as ion clocks, one has to overcome this lack of an accessible cooling transition. In 2001, an innovative solution based on quantum information techniques was proposed [53], whereby the clock functionality was separated from the cooling requirement by using two ions of different species within a linear Paul trap. The cooling (or logic) species would be an ion with an accessible strong cooling transition in the visible or near UV, which, once cold, would sympathetically cool the clock ion species by means of the collective motion of the two ions in the trap owing to their Coulombic interaction. While this solved the cooling problem, the additional difficulty was the lack of cooling fluorescence for the clock ion once the clock transition had been driven. Here, entanglement techniques would be used to map back the state of the clock ion onto the logic ion by means of the collective motional quantum state, and then read out from the logic ion’s fluorescence.

This quantum logic technique was first demonstrated, in principle, in 2005 on the $^{27}$Al$^+$ $^1S_0$–$^3P_1$ transition with $^9$Be$^+$ as the logic ion [54], and subsequently on the real $^1S_0$–$^3P_0$ clock transition in 2007 [55]. Recently, a second $^{27}$Al$^+$ quantum logic clock was built with $^{25}$Mg$^+$ as the logic ion, and comparisons between the two clocks have demonstrated a frequency uncertainty of approximately $9 \times 10^{-18}$, with an instability of $2.8 \times 10^{-15} \tau^{-1/2}$ and a frequency difference between the clocks of $-1.8 \times 10^{-17}$ [24]. With an observed ion linewidth of 2.7 Hz, this gives the highest atomic $Q$ observed to date of $4.1 \times 10^{14}$, and is currently the lowest frequency uncertainty for optical clock systems.
Table 2. Current optical clock status for different ion and atom species.

<table>
<thead>
<tr>
<th>ion/atom</th>
<th>clock transition</th>
<th>λ (nm)</th>
<th>estimated sys. freq. uncert./ absolute uncert.</th>
<th>freq. instability at 10 s</th>
<th>reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{27}$Al$^+$ ion</td>
<td>$^{1}$S$_0$–$^{3}$P$_0$</td>
<td>267</td>
<td>$9 \times 10^{-18}$</td>
<td>$6 \times 10^{-16}$</td>
<td>[24]</td>
</tr>
<tr>
<td>$^{199}$Hg$^+$ ion</td>
<td>$^{2}$S$<em>{1/2}$–$^{2}$D$</em>{5/2}$</td>
<td>282</td>
<td>$2 \times 10^{-17}$</td>
<td></td>
<td>[25]</td>
</tr>
<tr>
<td>$^{87}$Sr in lattice</td>
<td>$^{1}$S$_0$–$^{3}$P$_0$</td>
<td>698</td>
<td>$1.5 \times 10^{-16}$</td>
<td>$1.5 \times 10^{-15}$</td>
<td>[27]</td>
</tr>
<tr>
<td>$^{171}$Yb in lattice</td>
<td>$^{1}$S$_0$–$^{3}$P$_0$</td>
<td>578</td>
<td>$3.4 \times 10^{-16}$</td>
<td>$1.5 \times 10^{-15}$</td>
<td>[28]</td>
</tr>
<tr>
<td>$^{171}$Yb$^+$ ion</td>
<td>$^{2}$S$<em>{1/2}$–$^{2}$D$</em>{5/2}$</td>
<td>435</td>
<td>$4.4 \times 10^{-16}$</td>
<td>$1.1 \times 10^{-15}$</td>
<td>[26]</td>
</tr>
<tr>
<td>$^{40}$Ca$^+$ ion</td>
<td>$^{2}$S$<em>{1/2}$–$^{2}$D$</em>{5/2}$</td>
<td>729</td>
<td>$2.4 \times 10^{-15}$</td>
<td></td>
<td>[56]</td>
</tr>
<tr>
<td>$^{88}$Sr$^+$ ion</td>
<td>$^{2}$S$<em>{1/2}$–$^{2}$D$</em>{5/2}$</td>
<td>674</td>
<td>$3.8 \times 10^{-15}$</td>
<td></td>
<td>[57]</td>
</tr>
<tr>
<td>$^{40}$Ca atom</td>
<td>$^{1}$S$_0$–$^{3}$P$_1$</td>
<td>657</td>
<td>$7.5 \times 10^{-15}$</td>
<td></td>
<td>[58]</td>
</tr>
<tr>
<td>$^{171}$Yb$^+$</td>
<td>$^{2}$S$<em>{1/2}$–$^{2}$F$</em>{7/2}$</td>
<td>467</td>
<td>$1.8 \times 10^{-14}$</td>
<td></td>
<td>[59]</td>
</tr>
<tr>
<td>$^{115}$In$^+$</td>
<td>$^{1}$S$_0$–$^{3}$P$_0$</td>
<td>237</td>
<td>$1.8 \times 10^{-13}$</td>
<td></td>
<td>[60]</td>
</tr>
<tr>
<td>$^{199,201}$Hg in lattice</td>
<td>$^{1}$S$_0$–$^{3}$P$_0$</td>
<td>266</td>
<td></td>
<td></td>
<td>[61]</td>
</tr>
</tbody>
</table>

5. Optical clock status, stability and frequency shifts

Table 2 shows the current status of uncertainty and stability associated with the various optical clock ion and atom species. It should be noted that the various activities are very much work in progress. Thus, the table will become more extensively populated, and, of course, given results will also improve. The data shown in the table refer to published data in the refereed literature, but it should be recognized that preliminary results are continually being reported at major conferences. One example of this is the $^{171}$Yb$^+$ $^{2}$S$_{1/2}$–$^{2}$F$_{7/2}$ octupole transition, where a significantly improved preliminary value has been presented (N. Huntemann 2011, private communication) but not yet published.

Table 2 is arranged in order of decreasing uncertainty given by published values. The quoted uncertainties for the top five entries are estimated uncertainties, based on a combination of measured frequency shifts and their respective uncertainties and analysed shifts and uncertainties where related local or environmental parameters are known. The systematic uncertainty in these cases is below that of the Cs fountain standard operating at the time of the measurements. These results exemplify the difficulty of comparing high-accuracy standards, where the primary standard uncertainty would provide an unrealistic limit to the performance of the optical standards. The remainder of the entries refer to absolute frequency values that are ultimately referenced to a Cs fountain primary standard that provides the limiting uncertainty. In these cases, there are other species-specific uncertainties contributing that raise the uncertainty above that of the Cs standard.

The frequency instabilities observed for the $^{27}$Al$^+$ and $^{171}$Yb$^+$ ion clocks and the Sr neutral lattice clock are shown in figure 5. For comparison, the stability plot for a microwave Cs fountain standard against a hydrogen maser is given, and this comparison demonstrates the improved capability of optical clocks to

Phil. Trans. R. Soc. A (2011)
reach lower stabilities in faster times than their microwave counterparts. Also shown is the instability for a ULE cavity-stabilized optical LO stabilized to a 30 cm ULE cavity. The LO shows a characteristic flicker floor in the region of 1–100 s averaging before the drift sets in above 100 s. With recent advances in cavity designs that minimize cavity vibration sensitivities [38–43], the limiting floor to cavity stability is close to that set by thermal noise [41,43]. The thermal noise floor itself can be reduced to the $10^{-16}$ level fairly easily by optimizing cavity component material and dimension parameters (e.g. 30 cm ULE cavity with fused silica mirrors), but it becomes difficult to reduce it much below this without significant extra complexity in cavity design. These developments might include cooling a cavity of different material, e.g. silicon, to cryogenic temperatures and extending the cavity length to 1 m or more [62].

This LO stability floor has a significant implication for improvements in optical clock performance. The short-term performance is determined by the cavity-stabilized LO until the point where it can be steered by the cold atomic reference out to long averaging times, following a white-noise $\tau^{-1/2}$ dependence. For single ion clocks in figure 5, the cycle time to make a frequency correction can be seen to be 6–30 s, owing to the time needed to build statistics by sequential interrogations of a single ion. For the neutral atom case, frequency corrections can be made after a single 1 s interrogation on account of the improved signal-to-noise ratio due to parallel interrogations of many atoms. The actual cavity performance does not compromise this in that its floor stability is equal to or lower than the stability at the atomic reference cycle time. However, if one projects forward and assumes a 1 s interrogation time for both ion and atoms, and $N = 10^4$ atoms probed in

Figure 5. Frequency instability (Allan deviation) of various atomic clocks and a local oscillator as a function of averaging time: Al$^+$ data [24]; Yb$^+$ data [26]; Sr data [63]; narrow-linewidth laser data [64]. (Online version in colour.)
the lattice, the corresponding quantum-limited stabilities from equation (1.1) at equal averaging times are about one order and three orders of magnitude lower than currently observed. Thus to reach quantum-limited operation will require better LOs with lower thermal noise, especially to take advantage of the $\sqrt{N}$ improvement in signal-to-noise ratio in the atom lattice case.

If we contrast the ion and atom clock performances due to the various clock frequency shifts contributing, it is possible to highlight particular ion and atom species for which the majority of shifts are small. For the single ion case, the choice of a species with zero (e.g. $^{27}\text{Al}^+$ or $^{115}\text{In}^+$) or small (e.g. $^{171}\text{Yb}^+$ octupole) quadrupole shift has attraction. In parallel, the room-temperature blackbody shifts for $^{27}\text{Al}^+$, $^{115}\text{In}^+$ and $^{171}\text{Yb}^+$ octupole clock transitions are $-0.04$, $-0.14$ and $-1.6 \times 10^{-16}$, respectively [65,66]. For the atom clock species, the blackbody shifts are $16 \times$ and $34 \times$ higher for $^{171}\text{Yb}$ and $^{87}\text{Sr}$ than for the $^{171}\text{Yb}^+$ octupole transition, respectively. Of the atoms, only $^{199}\text{Hg}$ has a low blackbody shift ($-1.6 \times 10^{-16}$) [65,66] on account of its UV frequency. Generically, the odd-isotope atom systems have linear Zeeman shifts some three orders of magnitude lower than the corresponding ions, and the ion clocks will generally require magnetic shielding. Some ion clocks are first-order Zeeman-insensitive, though the ion clocks and atom clocks where this is not the case can have effective first-order Zeeman insensitivity by alternately stabilizing the components of a Zeeman pair. While this species comparison does not address all systematic frequency shifts, it serves to confirm the leading frequency stability and uncertainty performance of the $^{27}\text{Al}^+$ ion clock that has been observed at this time.

6. Remote comparison of optical clocks

In preparation for any redefinition, it is paramount to develop collective knowledge of the different optical clock systems through the intercomparison of their frequencies. Dissimilar clocks co-located in the same institute can now readily be intercompared by means of femtosecond combs. The accuracy of comparison can be approximately $10^{-19}$ or lower [29], which is well below the anticipated uncertainty levels of the clocks themselves for the medium-term future. One of the major considerations in such co-located comparisons is the need to take account of the height difference between the atoms or ion in the separate clocks, on account of the gravitational red-shift change. This shift is approximately $10^{-16}$ of the clock frequency for 1 m height difference on the Earth’s surface, so care is needed to ensure that this difference is known at the centimetre level of uncertainty.

The position for comparing optical clocks to the $10^{-18}$ level at locations remote from each other (e.g. between standards laboratories) without loss of clock accuracy is more problematic. There are a number of existing and anticipated remote comparison techniques, which I shall mention briefly. The most established method is by means of two-way satellite microwave frequency transfer or GPS (Global Positioning System) carrier phase. This is limited to approximately $10^{-15}$ per day at best, and is not capable of extension to $10^{-18}$ accuracy. The Atomic Clock Ensemble in Space (ACES), due for installation on the International Space Station in 2013–2014, is predicted to improve this situation, but several days of averaging would be needed to reach $10^{-17}$ uncertainty over intercontinental
On the optical front, over the last few years, there have been some very basic demonstrations of ground–satellite time transfer by laser link (T2L2) and inter-satellite optical links. These methods for optical frequency transfer are in their infancy, although the European Space Agency (ESA) is currently funding studies to take these ideas forward. A further alternative under consideration is the possibility for transportable optical clocks. Here, there will inevitably be a trade-off between portable clock accuracy and clock size and robustness, and it seems fairly clear that the performance of laboratory optical clock systems, where instability and uncertainty are minimized to the state-of-the-art level, will be more difficult to reproduce in the field.

The technique that has demonstrated capability over hundreds of kilometres to meet the necessary remote optical frequency comparison requirements is that of frequency transfer by optical fibre. Over the last half decade, a series of proving experiments have been undertaken between various standards laboratories and research institutes, such that a high confidence level for high-accuracy transfer has been established. There have been three different transfer arrangements demonstrated. The first of these involves r.f./microwave modulation of 1.5 μm-communications laser carrier light prior to fibre transfer to the remote location, and is appropriate for microwave transfer. The second method is based on transmission of the carrier frequency itself. In this case, in order to compare the optical standards, a frequency comb is needed to relate the 1.5 μm carrier frequency to the source optical clock prior to transmission, and a second comb at the remote end to relate the transmitted frequency to the target clock. The third method involves transmitting a portion of the comb itself through the fibre, with the result that both microwave frequencies (comb repetition rate related) and optical frequencies are received at the remote end, possibly without the need for a comb system there.

Figure 6 shows the state of the art for frequency transfer by fibre. The microwave-modulated transfer [68] and microwave transfer by comb [69,70] have achieved good performance, with the modulated carrier technique reaching $10^{-18}$ at 1 day. However, the optical carrier transfer technique can be seen as the most powerful method, demonstrating transfer stabilities below $10^{-18}$ over 250 km with just 100 s of averaging [71]. Other laboratories such as PTB, SYRTE and NPL have achieved or are working on similar proving experiments. PTB and the MPI (Max Planck Institute, Garching, Germany) have achieved a direct frequency link-up using a dedicated dark fibre link across 900 km with intermediate optical amplifiers located in eight intermediate link stations. NPL has conducted comb transfer experiments, and is studying carrier transfer across 86 km of the UK Janet ‘Aurora’ dark fibre network [69,70]. SYRTE and the LPL (Laboratoire de Physiques des Lasers, Villetaneuse, France) are involved in dark channel experiments near Paris using approximately 100 km distances comprising both dark fibre and fibre carrying Internet traffic [72]. Although dark channel transfer is more complicated, requiring drop-out and add-in techniques to optically amplify the signal in a bi-directional manner, the results so far show only marginal compromising of transfer stability.

In conclusion, the fibre transfer technique shows the greatest promise for high-accuracy frequency transfer, and it is anticipated that a number of European standards laboratories will be able to compare clocks by fibre within a few years. However, there remain logistical difficulties, both in accessing the dark fibre or
dark channel fibre links on a point-to-point basis, which requires negotiation with the commercial fibre providers, and in intercontinental transfer across oceans, where phase coherent all-optical repeater links will be needed. In the medium to longer term, one might envisage a remote comparison network that makes use of both fibre transfer and satellite optical transfer, underpinned by the concept of an optical master clock in space. A geo-stationary satellite carrying an on-board optical clock could provide the reference for both lower-orbit satellites and ground clocks, some of which would be linked by fibre. In this arrangement, the gravitational red-shift influence posed by the geoid would be reduced on the master clock owing to its lower gravitational potential, with less effect from geoid variations.

7. Conclusions

I have discussed generic options for an optical redefinition of the second, and outlined the state of the art for that which is, arguably, the most likely option of a definition based on a cold atom or ion ‘optical’ clock transition. In this framework, ‘optical’ covers a number of transitions from 700 to 250 nm, dependent on the particular species. I have also pointed out some of the advantages of certain species. However, the underlying question for this paper concerns the likely time scale for a possible redefinition. As a prelude to this, one needs to consider the requirements to be met or put in place for a redefinition. First, clearly, microwave fountain clocks have reached a level of maturity that facilitates their continuous operation over significant periods of time (e.g. months) such that they can contribute effectively to TAI. Improved optical clock reliability is
necessary before these latter systems can contribute in the same way. Secondly, the systematic frequency shifts for the majority of the different candidate species need to be fully evaluated. At this juncture, the rate of optical clock progress in this respect is rapid, as can be seen from figure 1. As these evaluations are made at improved uncertainty levels (e.g. $10^{-17}–10^{-18}$), the rate of progress will slow, owing to the level of uncertainty in shift coefficients and the progressive difficulty in characterizing and controlling perturbing fields. It does not seem sensible to make decisions concerning a redefinition until this rate slows. Thirdly, with full system evaluations and improved reliability, there need to be regular optical clock contributions to TAI. This is already anticipated through the setting up of optical secondary representations. At the moment, it is open for these to contribute to TAI purely for comparison purposes against the primary microwave standards. It is expected that, as confidence in their stability and reproducibility grows, the stage will be reached where, as secondary representations, they form part of the TAI steering process. Finally, comparison of remote optical clocks is essential.

It is still an open question as to the ‘best’ choice of standard for a redefinition, in respect both of atoms or ions, and of the particular species, and this will remain so until the different candidates are more fully evaluated. A major input to this process is the direct comparison of remote clocks, and this relies on the frequency metrology community establishing the infrastructure to carry out high-accuracy free-space and fibre-based optical frequency transfer and comparison. All these issues point to the possibility of an optical redefinition of the second at the earliest at the 2019 General Conference on Weights and Measures (Conférences Générales des Poids et Mesures, CGPM), and very likely not until a date after that. Current technology and industrial timing requirements are satisfactorily met with time and frequency standards underpinned by the microwave Cs fountain primary standard. However, the adoption of state-of-the-art technology often identifies new applications and extended opportunities. A good example of this is the evolution of GPS since its introduction in the 1980s. Further, it is the area of fundamental or high science requirements that will undoubtedly drive the improvement in and benefit from optical clock reliability and accuracy. This is well illustrated in the case of current optical space clock technology development activities within ESA programmes. High-accuracy frequency ratios between optical frequency standards will play a major role in this respect. In the longer term, future introduction of master clocks in space, with reduced sensitivity to the Earth’s gravitational potential, is likely to push fundamental science and Earth observation experiments to new sensitivities.

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Redefinition of the second


Redefinition of the second

4129


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