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Optical frequency / wavelength references

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Abstract
For more than 100 years, optical atomic/molecular frequency references have played important roles in science and technology, and provide standards enabling precision measurements. Frequency-stable optical sources have been central to experimental tests of Einstein's relativity, and also serve to realize our base unit of length. The technology has evolved from atomic discharge lamps and interferometry, to narrow atomic resonances in laser-cooled atoms that are probed by frequency-stabilized cw lasers that in turn control optical frequency synthesizers (combs) based on ultra-fast mode-locked lasers. Recent technological advances have improved the performance of optical frequency references by almost four orders of magnitude in the last eight years. This has stimulated new enthusiasm for the development of optical atomic clocks, and allows new probes into nature, such as searches for time variation of fundamental constants and precision spectroscopy.

1. Introduction

Ideas for using visible light from atomic transitions for precision instrumentation and metrology go back at least to the 1800s. There are several good reasons to use optical frequencies, and with the scientific and technological advances of the last century—relativity, quantum mechanics, electronics, coherent microwave sources, lasers . . . —we now have optical frequency references (OFRs) with truly exceptional performance. Optical atomic frequency references were developed out of basic scientific explorations in precision spectroscopy of atoms and molecules. They are now used in numerous scientific applications, serve to realize the unit of length, the metre, in the international system (SI), and are practical tools for dimensional metrology. Over the last 100 years the frequency accuracy of OFR has improved by about nine orders of magnitude, from 6 digits achievable with discharge lamps to modern frequency-stabilized lasers referenced to laser-cooled atoms that have a frequency reproducibility of about 15 digits.

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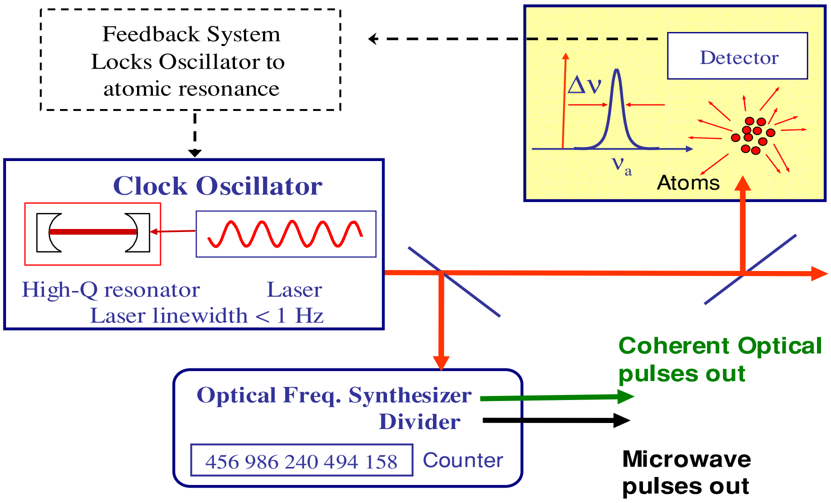

Figure 1. Schematic diagram of a modern optical frequency reference based on a spectrally sharp atomic resonance. The atomic resonance is probed by a narrow linewidth laser that is pre-stabilized to a high-Q Fabry–Perot cavity. On longer times scales the feedback control system steers the laser frequency to the atomic resonance. By adding the totalizing optical frequency counter/divider shown at the bottom of the figure, the optical frequency reference becomes an optical atomic clock.

By 'optical frequency references' or standards we mean a stable optical frequency that is referenced to a quantum transition in an atom, ion or molecule (all considered to be an atom, for simplicity in what follows) as shown in figure 1.

In addition to many practical considerations, the performance of frequency references is characterized by two basic figures of merit: accuracy and instability. These have formal definitions [1, 2]. But, as expected, instability is a measure of how much the frequency changes over time, and accuracy is a measure of how well the standard gives the correct frequency. If we consider the history of time keeping and compare clocks as different as sundials, pendulum clocks and quartz clocks, it is clear that greater accuracy and better instability are possible as the oscillation frequency increases. The improvement in clock performance that is anticipated by using optical frequencies derives mainly from the much higher frequencies, which divide time into smaller intervals. Whereas the oscillator in a caesium atomic clock vibrates at a microwave frequency ($f \sim 10^{10}$ Hz), a clock using a visible laser as its oscillator 'ticks' about 10^5 times faster, potentially allowing time intervals to be measured with a similar increase in precision. The fractional frequency instability that is expected for an atomic clock with N atoms operating in the quantum-projection-noise limit is approximately $\sigma_y \approx \frac{\Delta\nu}{\nu_0 s/n} \approx \frac{\Delta\nu}{\nu_0\sqrt{N\tau}}$, where $\Delta\nu$ is the linewidth of the transition, ν_0 is the oscillation frequency, s/n is the signal-to-noise ratio, and τ is the averaging time [2, 3]. (Frequently, for simplicity of notation, we will use the word 'stability' to describe a frequency that is stable in time, but the actual numerical values will be given in terms of fractional frequency 'instability' as in the equation above.)

In the late 1990s all the pieces were coming together for a major advance in the performance of optical frequency standards. Four main factors contributed to the rapid advance in performance: advanced methods for laser cooling and trapping of atoms to microkelvin temperatures [4], methods for laser frequency stabilization using high finesse Fabry–Perot cavities producing laser linewidths less than 1 Hz at 500 THz [5–7], methods of precision laser spectroscopy of trapped atoms producing line Q as high as $(\nu/\Delta\nu) = 10^{14}$ [8–10], and, finally, a convenient method for synthesizing and counting optical frequencies using

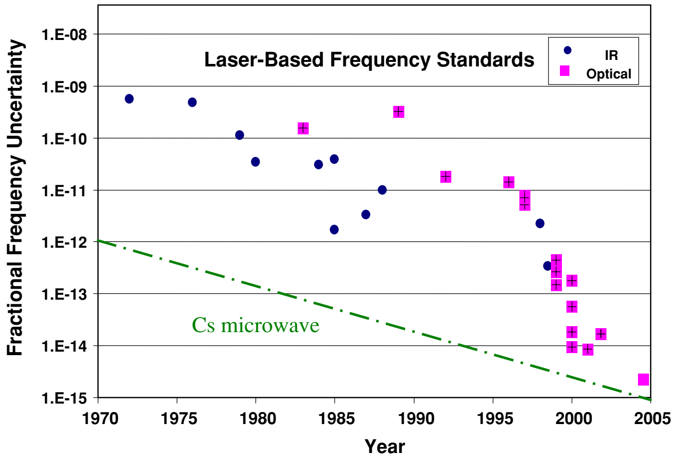

Figure 2. Fractional frequency uncertainty of state-of-the-art optical frequency standards verses date. The fractional frequency uncertainty represents the 'accuracy' of optical standards which can be compared to the dot-dash line representing the approximate performance of state-of-the-art Cs primary frequency standards at corresponding dates.

femtosecond laser optical frequency combs [11–13]. It now seems clear to most experts that the highest performance atomic clocks of the near future will be based on stable lasers probing narrow optical transitions in cold atoms. Already, OFRs have demonstrated better stability than the best microwave frequency standards for measurement times from a few seconds to a few hours [14]. And in several national laboratories, accuracies approaching a few parts in 10^{15} have been shown [15–17]. Optical clocks are now a reality and are beginning to perform at levels that eclipse previous non-optical technologies.

The accuracies to which optical atomic frequencies have been determined relative to primary Cs frequency standards are illustrated in figure 2 [18–23]. For the plot we have arbitrarily chosen the starting date as 1972, the year when the 3.39 μm methane-stabilized HeNe laser was first measured, but, in fact, there were a few measurements of far-infrared and CO_2 laser frequency references prior to that date.

This paper does not comprehensively review optical frequency standards, but selectively uses highlights of experiments to illustrate the state-of-the-art and to provide some perspective on how optical frequency standards have evolved, as well as their close connection to Einstein's theory of relativity over the past 100 years. Many modern high accuracy tests of special relativity (SR) have been founded on precision laser measurements and optical atomic frequency references. Moreover, the international system (SI) units of time, frequency and length are based on the quantum clock transition in Cs (a microwave transition) and the now-defined speed of light c. Other SI base units, the ampere and candela, also rely on frequency as part of their definition. Within the theoretical foundation of SR, the constant velocity of light and existing stable optical frequency references, it is very natural that both length and time could be measured with light. Recent advances in the technologies of optical frequency standards significantly enhance our capabilities to measure physical quantities in space–time and quantum energy differences.

A large number of people have contributed to the ideas discussed here, and serious research efforts are growing in laboratories around the world. Many of the most important concepts for OFR and optical clocks can be traced to leading pioneers in the field such as J L Hall, V P Chebotayev, S N Bagayev, C J Borde, A Clairon, T W Hänsch, G Kramer, J Helmcke and many many others. In the limited space here our discussion is necessarily incomplete, and we apologize from the outset for not giving proper recognition of important contributions of unnamed others. In hopes of rectifying this we refer the reader to some papers and compilations that provide different perspectives and more thorough reviews [7, 24–30].

2. Historical perspective on optical frequency references

The connection between Michelson's experimental results and Einstein's relativity is obvious today. Even so, scientists and historians have long discussed the relative significance of the contributions of Michelson, Lorentz, Fitzgerald, Poincaré and others to our understanding of space–time and the development of Einstein's relativity. In any case, precision optical measurements of length, frequency and the velocity of light have played central roles [31, 32]. We do not have to search far to find the thread that connects Michelson's experiments and Einstein's relativity to our modern optical atomic clocks. Now, 100 years later, we are at the beginning of the era when optical frequency standards are expected to overtake their microwave counterparts as the highest performance atomic frequency standards.

It appears that the idea to use optical references for length measurements goes back at least to Babinet about 1829 [33, 34], and it is well documented that as early as 1887, Michelson proposed using optical atomic emission lines (i.e. atomic energy level differences) and optical interferometry for length metrology [35–38]. Interferometry was used to measure the wavelengths of light from atomic emission sources in terms of the standard metre bar. The resulting wavelength references were subsequently used for other length measurements. This approach represents the beginning of the application of optical frequency references based on quantum transitions as metrological tools. However, real frequency-referenced optical metrology would have to wait 96 years until both units of time and length were based on atomic transitions and the speed of light was taken as a fixed and defined number. Michelson pioneered the use of optical atomic wavelength references and optical interferometry for the calibration of gauge blocks and length standards. Starting about 1892, studies were proceeding to identify the most appropriate optical atomic lines for precision length metrology and to develop appropriate measurement technologies. Promising candidates included well-known strong emission lines of Na, Li, Hg, H and Cd. As is usually the case, with the development of a new promising technology there was an interesting parallel evolution of the scientific psychology.

> The brilliant green (mercury) line gives beautifully clear circles even with a difference of path of half a million waves, so that in all probability this will be used as the ultimate standard of length (Michelson and Morley (1889) [36]).

> The green mercury line is one of the most complex yet examined (Michelson 1892).

Michelson, Morley, Benoît, Fabry and Perot developed extremely ingenious opto-mechanical systems and powerful interferometric methods [38–41]. Precise wavelength measurements by these scientists between 1893 and 1906 were used in 1927 by the CIPM (Comité International des Poids et Mesures) to define the 'Ångstrom' unit in terms of the wavelength of the red Cd transition at 644 nm. This served as an independent 'spectroscopic unit of length', and presumably was done because of its practical utility in spectroscopy. It is noteworthy that the definition of physical length remained connected to the official metre bar in Paris [41]. After considerable experimental work on optical sources and wavelength metrology the ^{86}Kr emission line at 605.7 nm was chosen as the official SI definition of the metre in 1960, finally replacing the metre bar. This optical atomic length standard was implemented interferometrically and endured for 23 years, being replaced in 1983 when the metre was redefined in terms of atomic frequency units based on the SI second (Cs $\sim$ 9.2 GHz clock transition) and the defined speed of light [34, 42].

2.1. The velocity of light and metre from optical frequency references

The speed of light c has intrigued humans since the beginning of time, and it plays a central role in both theoretical and experimental physics and in practical applications. The imaginative experiments of Ole Rømer in 1676 were the first to show that the velocity of light was actually finite. Rømer's experiments used the periodically pulsed source of light provided by eclipses of Jupiter's moons and the reference of length provided by the motion of Earth along its orbit around the sun. It is interesting that Rømer's original experiments on the velocity of light were performed at l'Observatoire de Paris, which still serves today as the location of the primary atomic frequency standards of France [43]. More precise astronomical measurements of c by E Halley and others followed [44, 45]. Subsequent ground-based measurements with rotating mirrors and toothed wheels confirmed the astronomical measurements and improved the precision [45].

In the 1890s, the situation became both more complicated and more interesting as high precision optical interferometry measurements by Michelson indicated that light did not follow exactly the same Doppler shift rules that were well known for the velocity of sound.

Measuring the velocity of light requires a known length for the light to traverse, a method for detecting the light transit, and some type of reference clock to measure the duration of the transit. To come up with an absolute number the length and time references must be calibrated in a traceable way to the standard units of measure of that date. Throughout the last century, optical measurements of the velocity of light and optical tests of special relativity evolved from measurements of the difference in the arrival time of light pulses, to interferometric measurements of phase differences of white light traversing different optical paths but maintaining nearly equal time delays, to interferometry using narrow atomic emission lines derived from flames and discharge lamps, to coherent laser interferometry over longer distances and time intervals, to modern phase-coherent measurements of optical frequency and phase that span the electromagnetic spectrum from microwaves to the UV. The latest experiments use length and time standards referenced to stable atomic transitions, and optical frequency references ultimately resulted in the speed of light being defined as a constant in the SI systems of units.

Although, the experimental technologies used by scientists for the early measurements were very different from the tools at our disposal today (and presumably dwarfed by the experiments and technologies of the future), the field of metrology has always relied on state-of-the-art instruments of their time. A historic example is the speed of light measurements made by Michelson and collaborators in the 1920s [46–49]. In that case the optical path length was between Mounts Wilson and San Antonio in California. The 35 km distance between the mountain tops was measured with a claimed uncertainty of $\sim$18 mm (approx. 1 part in 2000 000) by the US Coast and Geodetic Survey using accurate surveying methods that relied upon 50 m long Invar tapes that were calibrated by the National Bureau of Standards (now NIST). (This brings up another curious connection between precision metrology, tests of relativity, and fundamental science. It turns out that Invar was invented by C-E Guillaume, who was the director of the BIPM 1915–1936 and who won the 1920 Nobel Prize in physics for Invar and its understanding [41].) The timing reference for Michelson's speed of light measurements was actually a driven mechanical tuning-fork operating at about 135 Hz and calibrated against a standard pendulum, which was in turn calibrated to the then astronomical definition of the SI second (1/(86 400) of the mean solar day). Some mistakes were made in these precision measurements even after years of developing the measurement technologies (e.g. even with optical pulses generated with cw discharge lamps and rotating mirrors the group velocity dispersion effects were non-negligible for the light pulses travelling through air). As always, when the actual numerical values are important, it is necessary to have multiple experiments, different methods and several groups involved. In any case, it is quite remarkable that experiments using surveying tapes for length and tuning forks for time measured the speed of light to $c = 299\,796 \pm 4$ km s^{-1}, a value consistent with the accepted number today within a precision of $\sim 1.3 \times 10^{-5}$ [45, 49].

The precision in the SI unit of length improved rapidly with advances in atomic physics and laser spectroscopy using frequency stabilized lasers. Lasers were far superior to lamps in spectral and spatial coherence, and when stabilized to atomic transitions, provided much better wavelength (actually frequency) reproducibility than was possible with even the best discharge lamps. A paradigm change and landmark achievement occurred when measurements of the speed of light evolved from detection of the arrival time of an optical envelope, to direct measurements of the frequency and wavelength combined with the basic relationship $c = \lambda\nu$. Highly precise measurements of c were made using microwave resonators by L Essen and others from about 1947 on, but it took another 34 years before this was achieved with highly stable lasers by a team effort at NBS (now NIST) [50]. The main impediment to the practical implementation of length metrology based on laser frequencies was that even though laser frequencies could be very stable and reproducible, the actual frequencies of the atomically stabilized lasers were not known at that time with sufficient precision in terms of the SI unit of time/frequency, which since 1967 has been the Cs 9192 631 770 Hz hyperfine transition. (As an aside, it is interesting that L Essen is also recognized for the first publication of an operating Cs atomic clock at NPL in 1955 [51], while similar Cs clock activities were ongoing at other locations including MIT with Zacharis and at NIST with Lyons and Kusch [52–54].)

Most of the leading national measurement laboratories, NRC, NPL, NIST, PTB, BNM-SYRTE, VNIIFTRI and Institute of Thermal Physics-Novosibirsk, started significant research efforts focused on the goal of measuring the frequency of atomically stabilized lasers. Towards that purpose, optical frequency 'chains' were developed to multiply the frequency of the caesium atom at 9.2 GHz up to optical frequencies in the hundreds of terahertz range. These elaborate systems typically consisted of electronic frequency multipliers from microwave to millimetre waves, and eventually to laser radiation in the far-infrared region, then through several IR lasers eventually reaching the 3.39 μm HeNe laser (88 THz) stabilized to a saturated absorption signal in methane, CH_4. The first frequency measurement of the metrologically important CH_4-stabilized HeNe laser was accomplished by Evenson *et al* in 1972 [55]. That historic and heroic effort, along with others that followed shortly thereafter, provided the impetus for fixing the speed of light. To make that change it was also necessary to know the wavelength of the CH_4-stabilized laser relative to the then standard of length provided by the orange 605.7 nm, $2p_{10}$–$5d_5$ emission line from a ^{86}Kr discharge lamp. This was accomplished by Barger and Hall in 1973 using precise interferometric comparisons of the 3.39 μm HeNe laser [56]. The leading uncertainties were due to the asymmetries in the ^{86}Kr emission lineshape. Very high accuracy measurements of this wavelength ratio, and the ratio of the Kr line to the important iodine-stabilized 633 nm HeNe laser, were also made by Rowley *et al* at NPL and Baird *et al* at NRC [57–59]. It appears that some of the early wavelength comparisons of the 3.39 μm methane-stabilized laser relative to the ^{86}Kr lamp, completed by R Barger and P Giacomo at the BIPM about 1970, actually used one of the original Michelson interferometers from Michelson's time at the BIPM (figure 3) [60, 61]. Combining the high accuracy frequency and wavelength measurements of the 3.39 μm CH_4 reference and the

Figure 3. Photograph of a Michelson inteferometer at BIPM used by Michelson for dimensional metrology with light. It seems that this same instrument was used in about 1970 by Barger and Giacomo to make wavelength comparisons between the ^{86}Kr lamp that served to define the metre and a methane stabilized 3.39 μm HeNe laser. (2003 photo with permission from Tai Hyun Yoon, KRISS, Korea).

633 nm iodine line served as the 'final' measurements of the speed of light. They resulted in a fixed and defined speed of light, $c = 299\,792\,458$ m s^{-1} exactly, as recommended officially by the CGPM (Conférence Générale des Poids and Mesures) in 1975 [50, 62–67].

It took another ten years before actual frequency measurements reached the visible region of the spectrum by using the elaborate optical frequency chain illustrated in figure 4 [68]. Finally, in 1983 optical frequency chains were able to measure the frequency of the very stable and technologically important red HeNe laser (633 nm) stabilized to molecular iodine [69]. That and other optical frequency measurements provided the solid foundation for the 1983 redefinition of the metre in terms of the speed of light and the SI second [34, 42, 70]. Those earliest frequency chains were not phase coherent because the lasers were not all phase locked together, nonetheless they were real frequency measurements as opposed to wavelength measurements. Phase-coherent optical synthesis to the visible from microwave sources was first achieved with a harmonic chain by PTB in 1996 [71].

By definition in the SI, 'the metre is the length of the path travelled by light in vacuum during a time interval of 1/299 792 458 of a second'; now length, time and the velocity of light are consistently connected to the highly reproducible and accurate atomic frequency standard. This approach seems most natural in the framework of Einstein's relativity with the fixed velocity of light c in all reference frames. Excellent historical technical summaries of the development of optical frequency standards and the frequency measurements that led to the defined speed of light and eventual redefinition of the metre can be found in reviews by Petley, Rowley, Jennings and Hall, who were some of the principals involved in these important measurements [20, 25, 34, 63]. With the redefinition of the metre, the international standards bodies (the CGPM/CIPM) prescribe three basic methods for 'realizing' the SI metre. These are approximately as follows: (1) by a time interval measurement and the defined velocity of light $L = c\Delta t$, (2) from the wavelength of light with a known frequency measured relative to the Cs frequency $\lambda = c/\nu$ and (3) by using one of now 13 recommended radiations of frequency stabilized lasers, or lamps (Hg, Kr, Cd), or iodine absorption lines. All are described in

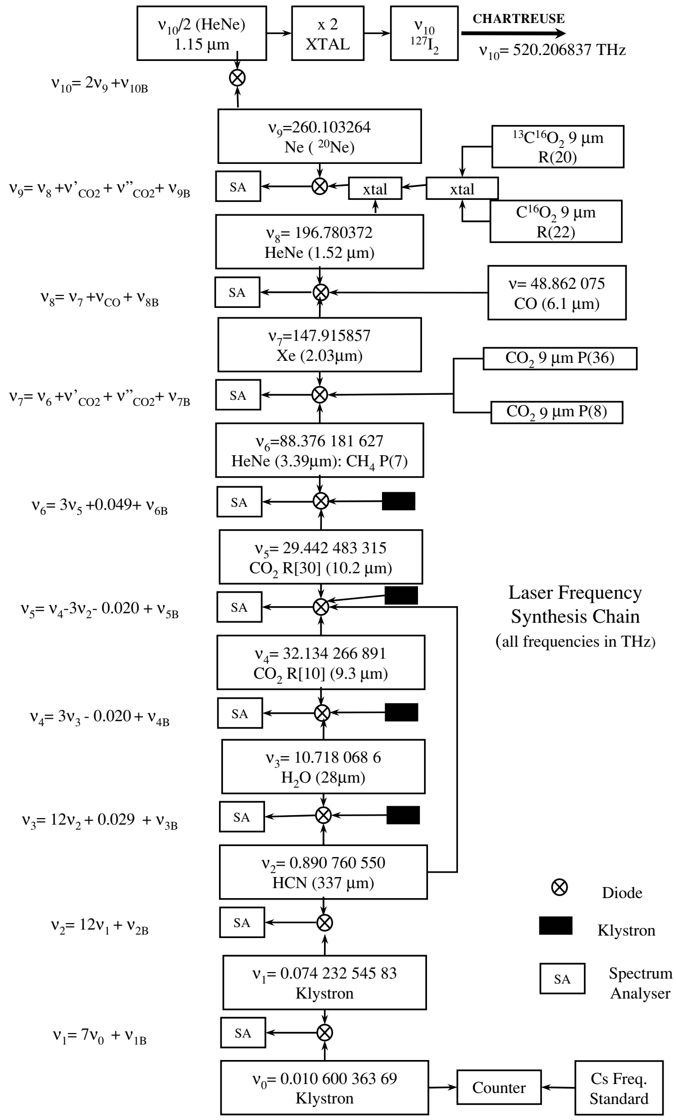

Figure 4. Diagram of the optical frequency chain used by Jennings *et al* for the first measurement of a visible laser frequency relative to the Cs atomic frequency standard [68, 69]. The system was based on a series of harmonic multiplication steps from the ∼9 GHz caesium frequency to the iodine line at 520 THz (described as chartreuse in colour by Jennings, Evenson *et al*). The chain relied upon klystrons in the microwave range and then a number of different types of lasers sources from the far-infrared to the visible, all of which needed to be frequency stabilized. Equations to the left indicate the mixing relationship for each link in the chain, where the ν_{iB} $(i = 1, 2, 3\ldots)$ represent the various beatnote frequencies that are measured with a spectrum analyser or counter.

detail with numerical values and uncertainties in the latest official report on optical frequency references for length metrology by T Quinn of the BIPM [72, 73]

Today, high quality optical frequency references have uncertainties that are orders of magnitude smaller than the uncertainties achieved in actual length metrology. Dimensional uncertainties are not limited by frequency standards but rather by physical effects in materials and optical interferometry (optical phase shifts, index of refraction, diffraction, optical aberrations, material creep, thermal expansion, etc). In fact, even the first few frequency measurements of the 633 nm I_2–HeNe laser [69] (with fractional frequency uncertainties of $\approx 2 \times 10^{-10}$) are still adequate (even 20 years later) for present high accuracy dimensional measurements. Once the frequency of the atomically stabilized laser is known, it can be reproduced and used for a traceable realization of the SI metre. It is not necessary to go back to the caesium definition of frequency unless higher accuracy is required. Present iodine stabilized HeNe lasers have frequency uncertainties of about 5 kHz at 473 THz ($\sim 1 \times 10^{-11}$, fractionally) with resetabilities of individual lasers about an order of magnitude better [72, 74–78].

2.2. *Optical frequency references and Einstein's relativity*

The original Michelson–Morley experiments were designed to measure the characteristics of the 'luminiferous ether' that was hypothesized to permeate all of space and serve as the medium for transmission of light. The experiments compared the relative phase of optical fields that made two-way transits over paths with different orientations in space but with nearly equal path lengths. Searches for differential phase delays were analysed with respect to the motion of the earth around the sun. Discrepancies between the experimental results and expectations based on an ether forced alternative theoretical interpretations. To distinguish between the theories more precise experiments were required. The goal was to precisely measure the relative velocity of light both along the direction of motion and orthogonal to the motion with an accuracy that could unambiguously resolve effects to second order in velocity, $O(v/c)^2$. The largest conveniently available velocity at that time was the velocity of the Earth in its orbit around the sun, so $(v/c)^2 \approx 10^{-8}$. Relative optical path length measurements to a fractional precision about 100 times better than this were required to make definitive statements about the actual magnitude of the second-order terms [79–81]. These experiments, now regarded as evidence for the constancy of speed of light and a repudiation of the ether, were viewed by many at that time as disappointing failures because they did not detect the ether that certainly must exist. The results now seem central to SR, but some historians claim that Einstein was not influenced very much by Michelson's experiments; and it also seems that Michelson was not an enthusiastic supporter of the relativistic interpretations of the experimental results [82, 83]. Nonetheless, 100 years later the puzzle fits together rather nicely.

Laser-based tests of special relativity (SR) were done as early as 1964 by Jaseja, Javan, Murray and Townes using 'infrared masers' (1 μm HeNe lasers) [84]. Improved versions of these Michelson–Morley and Kennedy–Thorndyke-type experiments by Brillet and Hall (see figure 5), and Hils and Hall respectively, were done as comparisons between an optical path length of a physical spacer and an optical atomic frequency of a quantum-based reference [85, 86]. In those cases, resonant modes of a stable Fabry–Perot cavity were compared to accurate and stable HeNe lasers stabilized to either the 3.39 μm methane reference or the 633 nm iodine transition. The results of comparing physical lengths to atomic frequency standards can be interpreted in the context of SR by adding small expansion parameters to the theory, as is done in the well-known test theories of Robertson, and Mansouri and Sexel (RMS) [87–89]. Experimental data are used to constrain the values of the small parameters in the theory and thus set limits on possible deviations from SR. Detailed discussions of experimental tests of relativity can be found in the excellent books by Born, Will, Zhang and other compilations [31, 32, 90, 91].

Some recent versions of these experiments use cryogenic optical Fabry–Perot cavities or a microwave resonator to test spatial isotropy and continue to reduced uncertainties [92–95].

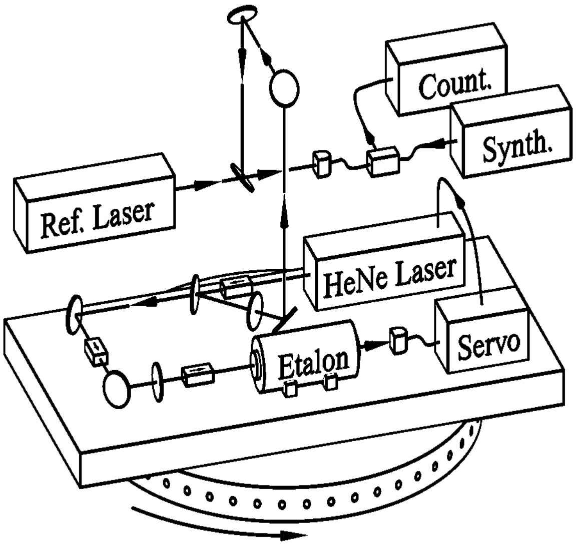

Figure 5. Diagram of the experimental apparatus used by Brillet and Hall in 1979 for an isotropy of space experiment of the Michelson–Morley type [85]. In this case, the optical length of the physical Fabry–Perot etalon was compared to an optical frequency standard (Ref. Laser) that was a 3.39 μm methane stabilized HeNe laser. The etalon, transfer HeNe laser and associated components were mounted on a stable granite block that was continuously rotated about a vertical axis. (Figure adapted with permission from [85].)

To date, and within the RMS framework, no experimentally reproducible discrepancy from SR has been found at the level of $\sim 2 \times 10^{-9}$ for Michelson–Morley, and $\sim 4 \times 10^{-7}$ for Kennedy–Thorndyke-type experiments. These experiments essentially test for variations in the two-way speed of light relative to a preferred frame as a function of orientation and velocity, respectively.

The relativistic time dilation, or second-order Doppler shift $v' = \nu(1 - (v/c)^2)^{1/2}$ causes problematic frequency shifts ($\sim 5 \times 10^{-13}$ fractionally) in atomic clocks that use room temperature atoms with corresponding thermal velocities of ~ 300 m s^{-1}. This problem becomes negligible when using laser-cooled atoms with low velocities (~ 10 cm s^{-1}). Conversely, the time-dilation effect can be enhanced and measured by using precision laser spectroscopy of fast moving atomic beams as done by Snyder, Riis, McGowan and Saathoff and their respective colleagues [96–99]. Measuring the optical frequency difference between fast and slow atoms determines the relativistic time dilation. The experiment with the highest precision used an apparatus similar to that shown in figure 6. Again, special relativity holds within experimental uncertainties, presently at the level of $\sim 2 \times 10^{-7}$ for the magnitude of the time dilation factor.

The gravitational red shift is a central component of Einstein's relativity and is a major frequency shift for high accuracy atomic frequency standards. In the gravitational potential of the Earth U, the fractional frequency shift is $\Delta f/f = -\Delta U/c^2 \sim (1.09 \times 10^{-16}\,\mathrm{m}^{-1})\Delta z$, where Δz is a relative change in height above the Earth. In the global positioning system (GPS) the gravitational red-shift and second-order Doppler shift are significant and must be taken into account for precise timing [100]. At our NIST laboratory in Boulder, the elevation is $\sim$1650 m corresponding to a gravitational red shift of $\sim 1.8 \times 10^{-13}$ (fractionally) relative to similar clocks at sea level (where the SI second is defined). This requires knowing the red shift at the NIST building with a fractional uncertainty of $\sim$0.1% for international comparisons to

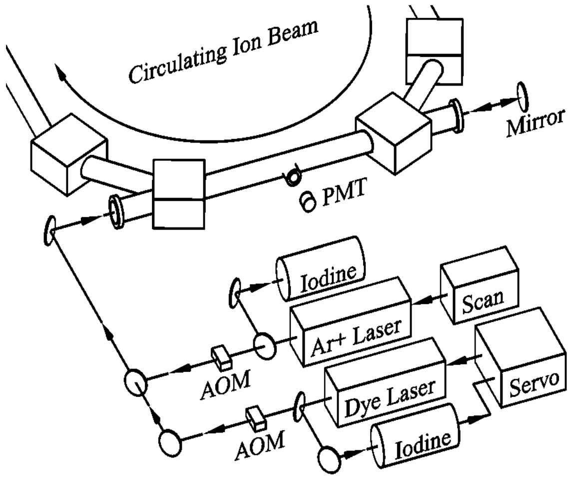

Figure 6. Simplified diagram of the apparatus used by Saathoff *et al* [99] to measure the time dilation effect (second-order Doppler shift, transverse Doppler shift) by using a saturated absorption transition in a beam of fast Li^+ ions (548 nm when at rest). Lasers with very different wavelengths (Ar^+ laser at 514 nm and a dye laser at 585 nm) were used to compensate the very large Doppler shifts that occur parallel and anti-parallel to the ion beam direction. (Figure adapted with permission from [99].)

achieve the level of accuracy of modern primary Cs fountain standards $\sim 5 \times 10^{-16}$ [101, 102]. Even a few metres of elevation makes a significant, measurable difference in the frequency. Optical frequency standards have not yet played an important role in experimental tests of the gravitational red shift, but their exceptional stability should prove useful for future gravitational red-shift measurements.

Another question is whether the fundamental 'constants' (such as the fine structure constant e^2/hc, m_e/m_p, g factors, etc) and hence atomic clocks are really constant in time. This has been a topic of discussion since Dirac suggested that the constants might vary based on arguments relating to the expansion of the universe and simple relationships between constants. Clocks are made from the most stable and measurable physical quantities that we find in nature, and for now, these are based on differences in atomic energy levels. It is thus natural to use stable atomic frequency references to test physical principles and theories and to search for new physics or forces that might depend on time, space, velocity, etc. The theories of such clock tests have been discussed in detail, and a few high precision experiments have been done [31, 103–110]. A popular experimental approach is to measure ratios of atomic frequencies and search for any reproducible systematic changes that occur over time. Reproducible measurements can be made as long as any time variation of the 'constants' occurs on a time scale that is longer than the time interval between frequency measurements, which is limited by the averaging time required to reach the accuracy of the standards. Measurements between optical standards can presently be made at a level of $\sim 2 \times 10^{-15}$ in 10 s (more on this in section 5.1). By comparing atoms with different atomic number Z it is possible to make some qualified statements about possible time variations of ratios of fundamental constants, including the fine structure constant α, the electron to proton mass ratio m_e/m_p, and gyromagnetic ratios. These atomic physics experiments can also be viewed as tests of SR since atoms with large Z have significant relativistic corrections to the atomic wavefunctions. Thus a change in α or relativity could show up as differential changes in the energy levels between light and heavy atoms [111]. Astronomical observations of optical

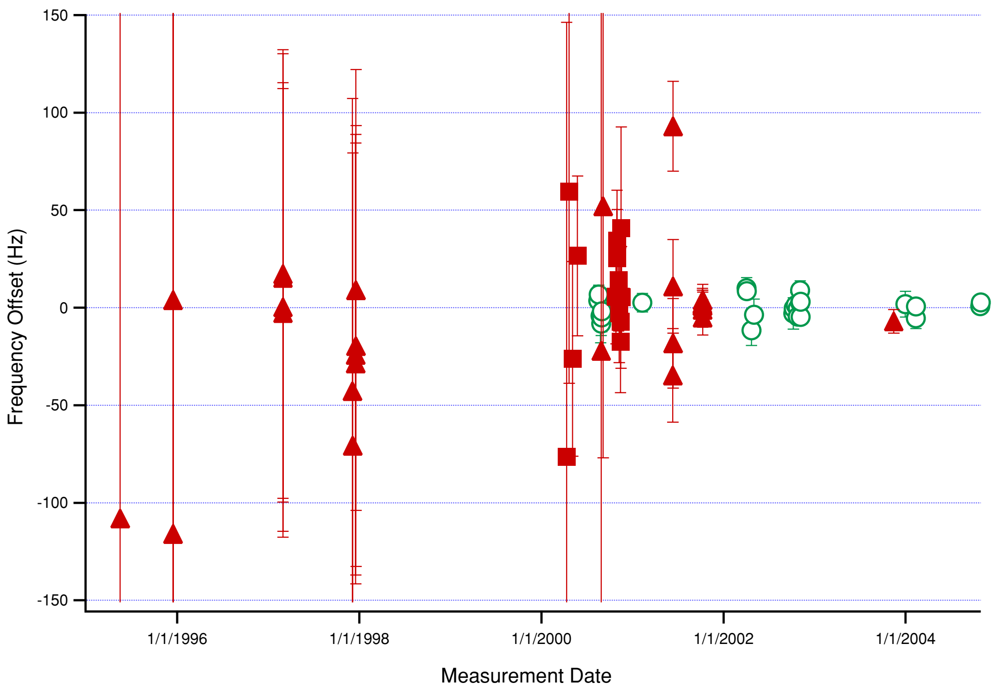

Figure 7. Historical record of published absolute frequency measurements of the Ca and Hg^+ optical standards (456 THz and 1064 THz respectively) relative to the Cs primary frequency standard. The vertical axis is in units of hertz away from the respective mean values. The Hg^+ data (open circles, green) were taken at NIST, while the Ca data come from both PTB (triangles, red) and NIST (squares, red) [118, 119].

atomic transitions have shown hints that the atomic frequencies might have been different in the early universe [112, 113]. However, the astronomical data are, at present, inconclusive [114–116]. At the current epoch, the few high-accuracy laboratory experiments that have been completed show no reproducible, convincing evidence of time-dependent changes to atomic clock frequencies [15, 108, 109, 117]. Some data showing the Ca and Hg^+ optical frequencies measured relative to Cs standard over some years are shown in figure 7. There are various ways to interpret these data in terms of possible variations of fundamental constants, but it is clear from the data that within measurement uncertainties there is no indication of any systematic temporal variation of these atomic frequencies relative to each other.

The higher precision and accuracy becoming available with optical atomic frequency standards (see figure 2) will allow tests of Einstein's relativity and other foundations of physics with much higher accuracy. In essence, the optical atomic frequency standards and optical frequency combs offer more precise rulers for time, frequency and length than have been available, and will allow deeper probing into the structure of space/time. Our enthusiasm for the future of stable optical frequency references is not new.

> Optical and infrared masers make possible and attractive new experiments and refinements of old ones, where great precision in measurement of length is needed. (C H Townes (1964) regarding laser tests of relativity [84].)

With the improved experimental tools of today, we can certainly travel further down this path that was started long ago.

3. Present status of optical frequency standards

Experimental atomic physics is the foundation that supports optical frequency standards, and the current technology is based on precision, nonlinear laser spectroscopy of simple atoms that have been laser cooled and trapped. The two basic types of optical atomic frequency standards are systems based on single trapped ions and those that use clouds of cold neutrals. They share many of the same technologies but have some characteristic differences.

As we start this discussion it is useful to recall generally what we mean by specific characteristics, such as frequency stability (does the frequency change over a specified time interval τ), frequency reproducibility (self-reproducibility is a measure of whether a single reference gives the same frequency each time it is used, moved, turned off or on etc, and cross-reproducibility measures how well similar frequency standards give the same frequency), and accuracy (which has two meanings: how well does the standard produce an exact frequency in terms of the SI 'second', or sometimes it is used more loosely to mean how well does the standard represents the natural frequency of that specific atomic transition). Some experiments require only frequencies that are stable, for example, when looking for effects that can be modulated, as in the rotating interferometers in Michelson–Morley experiments. Other experiments, such as searches for time variation of fundamental constants, require accurate measurements that can be repeated. Good stability is a necessary condition but it is insufficient to guarantee high accuracy.

3.1. Single ion optical frequency standards

The essential ingredient of an optical frequency standard is the atomic transition to which the frequency of the optical oscillator is steered. Ideally, these references are anchors in frequency space that are totally immune to any perturbations. But the motion of atoms produces shifts that can never be fully eliminated because the atoms never come fully to rest; and, likewise, external perturbations, which can cause shifts to the internal level structure are never fully eradicated. The pursuit of the 'best' optical frequency and time standard has coalesced in most laboratories to an effort to identify the 'best' anchor—where 'best' refers to the optimum compromise between stability and accuracy on one hand and practical issues, such as reliable and simple lasers, on the other. Clearly, the best stability and highest accuracy will be provided by slowly moving laser-cooled atoms and trapped ions. In such systems, fractional frequency instabilities at or below 10^{-14} at 1 s and reproducibilities better than one part in 10^{15} have already been demonstrated [14–16, 108, 119, 120]. It may be possible to benignly hold laser-cooled atoms in optical wells generated by the intersection of light beams far detuned from any internal transition [10]. If the atoms are sufficiently cold, their motion will be confined to an individual well and hence to vibrational excursions less than an optical wavelength (the so-called Lamb–Dicke limit). In this limit, a domain achieved routinely with laser-cooled and trapped ions and, more recently, with laser-cooled and lattice-trapped neutral atoms, first-order Doppler shifts are eliminated and second-order shifts are reduced to below 10^{-18}. A million atoms confined to the Lamb–Dicke limit offer enticing predictions for stability and accuracy of an optical clock, but much work remains.

Narrow, weakly allowed optical transitions of single, laser-cooled, trapped ions are nearly ideal references for optical frequency and time standards. High resolution is possible because perturbations can be made small and interrogation times long. A key advantage of single ions is that they can be laser cooled to the lowest vibrational state of motion and benignly confined to the Lamb–Dicke limit. Therefore, because the laser-cooled ion is restricted to a small volume in space, most systematic shifts are either small, such as Doppler shifts to all orders,

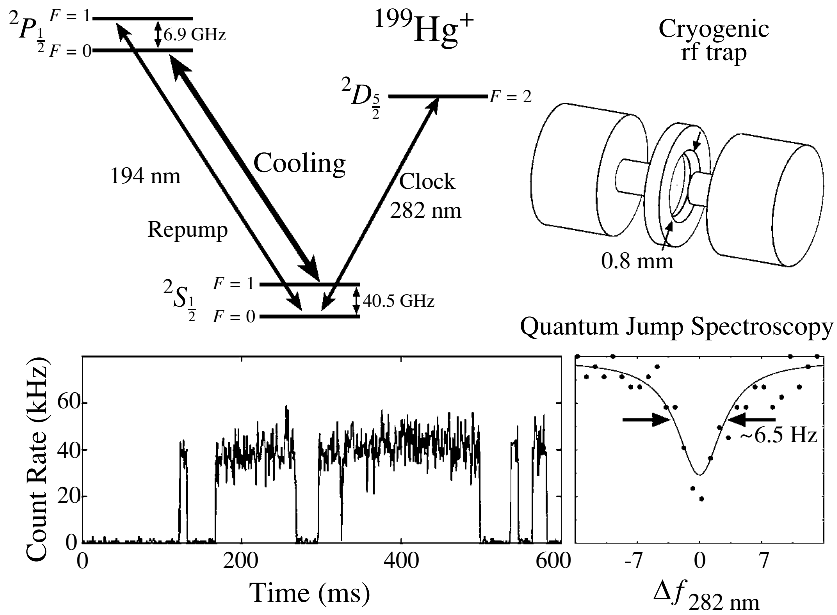

Figure 8. Some of the key components of the Hg^{+} single-ion optical clock. Upper left is a simplified optical energy level diagram of $^{199}Hg^{+}$ and to its right is a diagram of the miniature, cryogenic ion-trap showing the two endcap electrodes and the centre, ring electrode. Lower left shows quantum jumps used for state detection of the clock transition [122]. If the ion remains in the ground state following a clock excitation pulse at 282 nm, the ion will readily fluoresce on the strongly allowed transition at 194 nm. Conversely, if the clock transition is made, then no fluorescence at 194 nm is seen. Lower right is a Fourier-limited spectrum of the clock transition (probe time, 120 ms) [9].

or can be well controlled. Most single-ion optical clocks are expected to ultimately reach systematic fractional frequency uncertainties approaching 10^{-18}. Furthermore, the fractional frequency uncertainty reported for several single-ion standards, all based on weakly allowed, electric-quadrupole transitions, are less than 1×10^{-14} [15, 17, 108, 121]. In figure 8, we show some of the essential features and components of the optical time and frequency standard based on a single Hg^{+} ion.

One of the largest systematic uncertainties that had previously limited the accuracy of the Hg^{+} standard as well as each of the other ion-based optical standards so far demonstrated is the electric-quadrupole shift that arises from the interaction of the excited state quadrupole moment with any static electric-field gradient. Recently, the quadrupole moments pertinent to the optical clock transitions in Sr^{+} [17], Yb^{+} [16] and Hg^{+} [123] have been measured and compared to the predicted values obtained from a multiconfiguration Dirac–Hartree–Fock calculation. Any quadrupole shift of the clock frequency in $^{199}Hg^{+}$ is now conservatively estimated to be fractionally less than 1×10^{-15} [123]. More importantly, we expect that the quadrupole shift will no longer limit the accuracy of the mercury ion optical clock when it is operated with alternating orthogonal components of a small applied magnetic field of constant amplitude [124], which is stepped each measurement cycle. (We note that an alternate scheme exists and has been used to cancel the quadrupole shift of the clock frequency in Sr^{+} [17].)

In a complementary effort at NIST, we are working towards the development of a two-ion, 'quantum logic' optical clock [125]. In this system, two ions of different species are held in a linear RF trap. One ion (the 'clock' ion) provides a suitable optical transition for an atomic clock, whereas the other ion (the 'control' ion) provides a strongly allowed transition for laser cooling the two-ion ensemble as well as the rapid state detection of the clock ion. In this scheme, the clock ion is sympathetically cooled to the motional ground state of the trap via Coulomb coupling with the laser-cooled control ion, and then its clock resonance is probed by radiation whose frequency is alternatively stepped to either side of the resonance. The resulting superposition of state amplitudes for each step is quickly (<0.1 ms) mapped onto the control ion through the quantized motional states of the coupled two-ion system using elementary quantum logic operations. The state projected in the subsequent probe of the control ion is a faithful measure of the original superposition of states in the clock ion. In the version being pursued at NIST, we are using an Al^+ clock ion and a Be^+ control ion. The weakly allowed $^1S_0$–$^3P_0$ transition in $^{27}Al^+$ at 267 nm is a particularly attractive reference for an accurate and stable optical clock owing to its narrow linewidth and the absence of a static electric-quadrupole shift.

During the last year, we have trapped a single Be^+ ion together with a single Al^+ ion and were able to achieve ground state cooling of the Be^+/Al^+ ion crystal by applying Doppler- and Raman sideband cooling on the Be^+ ion. After optimizing the cooling process and the coherent state manipulation of the beryllium ion, we demonstrated a proof-in-principle of the basic clock protocol by first exciting the more allowed $^1S_0$–$^3P_1$ transition in Al^+ [126]. The natural resonance of this transition has a width of about 500 Hz, and its frequency was known from spectroscopic data to within an uncertainty of 2 GHz. A high-finesse optical cavity that is held in an evacuated, temperature-regulated chamber provided us with a frequency reference with a day-to-day frequency uncertainty of better than 5 kHz. We optimized the basic measurement protocol and recently observed the $^1S_0$–$^3P_1$ transition with all its hyperfine components. A measurement of their difference frequencies agrees well with theoretical predictions of the hyperfine splitting constant for the $^3P_1$ state [127]. We also made a preliminary absolute frequency measurement of the $^1S_0$–$^3P_1$ clock transition. In the next few months, we will apply the quantum logic measurement protocol to search for the narrow, $^1S_0$–$^3P_0$ optical clock transition in Al^+ ($\tau \approx 35$ s). Then, as a first application, we will perform a high-precision search for time variation of the fine-structure constant by comparing the frequency of the Al^+ clock to both the frequency of the Hg^+ standard and that of the Ca standard at various intervals.

3.2. Neutral atom optical frequency references

The past decade or so has seen an explosion of interest in laser-cooled neutral standards. These standards differ from trapped-ion standards in two significant ways. First, large numbers ($>10^6$) of neutral atoms can be trapped, thus enabling outstanding stability through a large signal-to-noise ratio. Simple estimates show that instabilities of 10^{-17} at 1 s or less are possible with neutral atom standards, although considerable improvement in present day local oscillators would be required to support such performance [3, 8, 128]. Second, due to their lack of charge, neutral atoms require modified trapping and measurement strategies. These strategies usually entail turning off the trap during probe periods to avoid unwanted frequency shifts, but thereby require the control of systematic shifts due to Doppler effects resulting from thermal motion and gravity.

The main neutral candidates come from atoms with narrow clock transitions and broad, strong cooling transitions. These include the alkaline earth atoms Ca, Sr and Mg, and other promising candidates such as Yb, Ag, and H. The Ca optical frequency standard (figure 9) serves as a convenient example because it is one of the most advanced of the neutral atom frequency standards, and it is being explored at a few different laboratories [119]. The clock

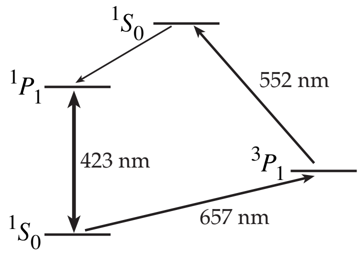

Figure 9. Relevant energy levels of ^{40}Ca for laser cooling and the optical clock. First stage laser cooling uses the 423 nm transition, while the second stage uses the 657 nm clock transition in conjunction with the 552 nm quenching transition. The clock signal is derived from the transition at 657 nm, which has a natural linewidth of 400 Hz.

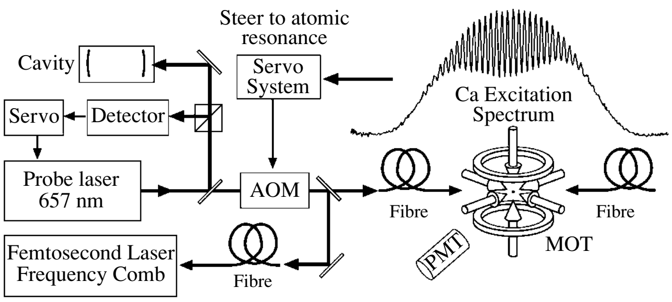

Figure 10. Schematic of the calcium atomic clock apparatus. Using two stages of laser cooling, about 10^6 atoms are trapped and cooled to a temperature of 10 μK. With the trapping laser light blocked, the freely expanding atoms are probed by a laser that is pre-stabilized on a narrow resonance of an optical cavity. A servo system keeps the probe laser frequency fixed on the atomic resonance via a signal derived from the excitation spectrum. Light is sent through a fibre to a mode-locked femtosecond-laser frequency comb where the optical oscillations can be counted and/or compared to other frequency standards.

transition at 657 nm has a natural linewidth of 400 Hz and can be easily accessed with inexpensive diode lasers. The laser cooling transition at 423 nm (35 MHz linewidth) is used to collect millions of atoms in a magneto-optic trap (MOT) with a residual temperature of 2 mK. Advanced versions of the standard now use a second stage of laser cooling based on the clock transition to further reduce the atomic temperature and thereby the Doppler shifts [129, 130]. Because the laser cooling light induces large Stark shifts on the clock transition, this light is turned off and the atoms are released for ballistic expansion during probing of the clock transition. This approach necessarily leads to a measurement cycle that contains alternate periods of trapping and probing.

For the calcium atomic clock at NIST, a typical cycle commences with a 15 ms trapping period during which several million atoms from a beam are loaded into a 423 nm MOT (the clock apparatus is depicted in figure 10). This is followed by 7 ms of second-stage cooling that transfers 25% of the atoms into a shallower MOT producing a residual temperature of 10 μK. With the laser cooling beams extinguished, the clock transition is then excited with a Bordé–Ramsey sequence [131] consisting of two pairs of 3 μs counter-propagating pulses separated in time by $\sim$330 μs. The probe beams are filtered with optical fibres to ensure spatial modes of high quality. In order to achieve high resolution on the clock transition, it is necessary to pre-stabilize the frequency of the probe laser on a narrow resonance of a high finesse cavity that is environmentally isolated. This enables the probe laser to achieve a laser linewidth of a few hertz for times of several seconds. Depending on its detuning from the atomic resonance, the excitation pulse sequence from the stabilized laser excites some fraction of the atoms from the $^1S_0$ ground state to the $^3P_1$ state. This transfer is measured with a shelving detection scheme [122, 132]. Shelving detection uses resonance fluorescence induced on a strong transition (here the 423 nm cooling transition) to measure the absence of population in the ground state rather than detecting a weak decay fluorescence signal at 657 nm from the population in the excited state. With neutral atoms, we can normalize the signal to the number of atoms in the trap by applying two such detection pulses [119, 133]. To generate the excitation spectrum, we slowly sweep the frequency of the stabilized laser relative to its reference cavity fringe with an acousto-optic modulator, while the measurement cycle is continuously repeated. Figure 10 shows a Fourier-transform-limited spectrum taken at relatively low resolution (fringe period = 23.1 kHz), as determined by excitation pulse duration and temporal separation.

For clock operation, the resolution is increased to $\sim$770 Hz and a modulation servo technique is used to keep the probe laser frequency locked to the central fringe of the spectrum. A fraction of the probe light is sent through an optical fibre to a mode-locked femtosecond-laser comb measurement system, which is used to compare the performance of the standard with other atomic standards. Such comparisons have demonstrated fractional frequency instability as low as 5×10^{-15} at 1 s for the NIST Ca standard. This is an exceptionally good stability, but is limited by the small ratio of actual probe time ($\sim$1 ms) to atom preparation/detection time ($\sim$25 ms), which leads to an effective increase in the frequency noise spectrum of the probe laser (through the Dick effect) [128, 134–136]. Future standards (neutral and ion-based) will require that this ratio is increased considerably in order to achieve optimum performance levels.

We are now at the stage where such standards work very well and have excellent stability, but we face the arduous task of verifying that all systematic errors are as small as projected. Many of these errors result from fundamental physics effects (such as residual Doppler shifts, blackbody radiation-induced frequency shifts, gravitational shifts), while others involve nonfundamental technical issues (such as optical table vibrations, frequency chirps in the acousto-optic modulators and defects in the spatial modes of the probe laser beams). Presently, a few papers have begun to address the fundamental issues in detail at the 10^{-16} level, while, overall, fractional frequency uncertainties are now moving just below the 10^{-14} level, limited mainly by technical rather than fundamental issues. Encouragingly, two different labs have measured the absolute frequency of the Ca transition at the $\sim 10^{-14}$ level with excellent agreement [119].

As mentioned previously, there are transitions in other neutral atoms that have attracted considerable interest for use as optical frequency standards [10, 109, 137–145], however, at this stage it is not clear which can give the best performance or most the useful atomic clock.

Atomic hydrogen is particularly interesting because its theoretical simplicity allows direct connection between experimental results and fundamental physics [109, 140, 146, 147]. Hydrogen also has an attractive 1S–2S clock transition with a narrow linewidth and, with some effort, can be cooled to fairly low velocities. It is nearly unique in that the energy level differences in hydrogen can be calculated to approximately the same level of accuracy as they can be measured. Comparisons between theory and experiment for hydrogen have given a wealth of information on atomic structure, Rydberg constant, the Lamb shift, size of the proton, etc. The technological challenges with hydrogen are a significant impediment to its development as an optical clock, but the fundamental physics that can be extracted makes the pursuit very worthwhile. Also, for theoretical reasons there is growing interest in precise optical frequency measurements in helium for connection to fundamental physics [148].

Perhaps the most promising approach for future neutral atom standards is to use odd-numbered isotopes of two-electron atoms, for which it is possible to excite highly forbidden transitions (natural linewidths of 1 to 10 mHz) in atoms confined in an optical lattice [10]. The wavelength of the lattice laser is chosen to yield equal Stark shifts for both the ground and excited states, leaving the clock frequency unchanged. With the atoms confined to the Lamb–Dicke regime (i.e. a fraction of a wavelength), residual Doppler effects will be negligible and long interaction times will be possible. If higher-order Stark shifts can be adequately suppressed, such a system could attain the accuracy level promised by single trapped-ion systems but with the improved stability that comes with large numbers of atoms. Presently, at least seven groups are pursuing lattice-based standards in Ca, Sr and Yb, with several experiments poised to reach maturity in the near future [10, 138, 142, 144, 145, 149]. If it is possible to cool and confine large numbers of neutral atoms in optical lattices to the Lamb–Dicke limit while at the same time maintaining negligible perturbations to the clock transitions then these systems could indeed be the ultimate optical frequency standards. Such a system could have all the advantages of trapped ions—tight confinement, no Doppler, no collisions, long interaction times—as well as the advantages of neutrals—large numbers, large signal-to-noise ratios and convenient laser wavelengths. This might be the best of both worlds; however, there are many scientific and technical issues that must be addressed, including, higher order polarization sensitivities, potential Stark shifts from two-photon transitions, non-trivial optical pumping, efficient internal and external state preparation and achievable cycle times [10, 137, 142, 149, 150].

Further in the future, we might even imagine loading an optical lattice with coherent atoms organized to achieve one atom in the ground state of each lattice site, forming a Mott insulator transition (see the chapter by E Bloch in these proceedings). Coherently prepared atoms in such a lattice could also be exploited by applying entanglement ideas proposed for ion clocks [125] but implemented with neutral atoms, as is being studied for quantum information processing. Under these conditions, the instability could scale as $1/N$ rather than $1/N^{1/2}$.

4. Other applications of optical atomic frequency references

Perhaps the most promising commercial applications of OFR will be in communication and navigation systems, or for special uses such as coherent optical radar. Beyond their present use in scientific applications (such as references for precision spectroscopy) and length metrology, OFRs are just beginning to see some new applications.

One modern application with widespread use of optical frequency references is in optical communication systems utilizing wavelength division multiplexing (WDM). Relatively recently, the telecom industry moved to WDM optical communication systems to achieve high data rates over optical fibres. In fact, for the 'wavelength' multiplexing, the industry wisely chose a frequency-based optical grid. The International Telecommunication Union (ITU) frequency grid for dense wavelength division multiplexing is fixed on an optical frequency of 193.1 THz ($\sim$1552 nm) and supports a variety of channel spacings ranging from 100 GHz down to 12.5 GHz [151]. The accuracy and stability required for the ITU-grid are not at the same levels of performance previously discussed (even 1 GHz at 193 THz is fractionally 5×10^{-6}); but nonetheless, these systems do demand atomic and molecular references for calibration purposes [152, 153]. Some research programmes continue to explore coherent optical communication systems that might benefit from optical frequency references with higher precision, or perhaps optical frequency combs.

Many of the real-world applications of atomic frequency references do not require high absolute frequency accuracy, but rather require robust stable frequency sources for synchronization of events or the precise measurement of time intervals. In these cases having high stability might be more important than absolute accuracy. The excellent stability of optical standards bring new capabilities for ultra-precise timing, subfemtosecond timing jitter and low-phase-noise microwave signals as described below.

5. Optical frequency metrology

The harmonic optical frequency chains developed in the 1970s and 1980s proved that it was possible to count optical frequencies, and these systems played important roles in laboratories responsible for primary standards. However, the chains were too large, complicated and unreliable to see widespread use. That all changed in 1999 when Udem *et al* demonstrated [154, 155] that Hänsch's idea [156] of using ultrafast mode-locked lasers for optical frequency metrology was indeed valid to a high degree of accuracy—just as expected from the Fourier transform. They showed that the frequencies of the discreet modes produced by a mode-locked Ti:sapphire laser and spanning ∼20 THz were indeed spaced precisely by the pulse repetition rate f_{rep} [154]. That paradigm-changing demonstration revolutionized the field. The frequency of any mode of the optical frequency comb produced by a mode-locked laser could be described by the simple comb equation $f_N = N f_{\mathrm{rep}} + f_0$, where f_0 is an offset frequency common to all modes.

Another critical achievement of the optical comb technology was the broadening of the optical spectrum to cover a spectral bandwidth of an octave. This was first accomplished using microstructure optical fibres whose broadening capabilities were discovered about in 1999. With an optical octave spanned, the femtosecond laser optical frequency combs (FLFC) could be stabilized using the 'self-referencing' method [13, 157], as first demonstrated experimentally by Jones *et al* [158]. The technology of optical frequency combs and their applications have developed rapidly over the past five years. Some recent reviews give good summaries of the field [11, 159–162]. In contrast to the harmonic optical frequency chains, the FLFC are fairly easy to operate, robust and reliable. The ultrashort pulses from stabilized self-referenced FLFC provide exquisite timing markers (sub-fs timing jitter is possible) and the spectrally broad optical frequency comb provides an almost ideal ruler for optical frequency synthesis and measurement. When self-referenced and locked directly to an optical frequency reference the comb transfers the accuracy and good stability to other spectral regions (from the RF to UV) and phase coherently divides the optical frequency to the microwave range where it can be interfaced with electronic signals (see figure 11) [163]. When used as an optical frequency divider these system now provide microwave signals with unprecedented low phase noise. Atomically stabilized FLFCs, when used as optical frequency dividers, now provide some unique capabilities, such as relative timing precision of femtoseconds, with unprecedented stability producing microwave signals with phase-noise 20 to 40 dB lower than the best existing microwave sources [164–166].

Several recent experiments have tested the fidelity of the FLFCs, including: uniformity of the comb spacing, second harmonic ratios, reproducibility as a frequency translator and divider, etc [154, 167–169]. Even with the high nonlinearities, the strong optical frequency chirp, and distortion of the optical pulses in the lasers and nonlinear fibres, the time-averaged frequencies of the optical comb lines obey the simple optical comb equation. The experiments conclude that, at least for averaging times longer than about 100 ms, the modes of the FLFC are well described by the simple comb equation, and when averaged for several hours can give a precision of 19 to 20 digits [170–172]. Thus, the FLFC are accurate frequency synthesizers

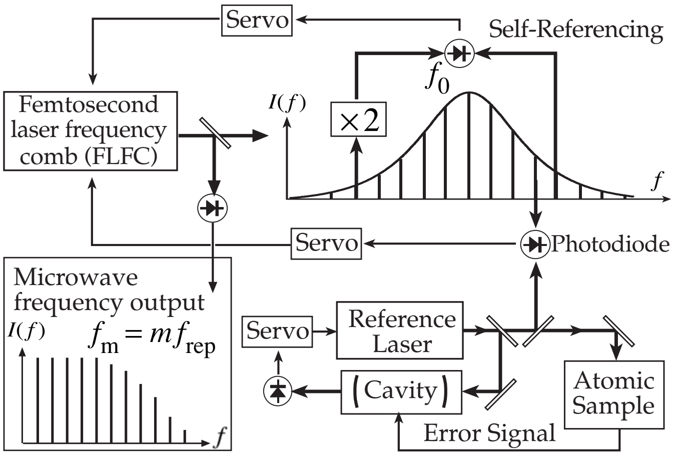

Figure 11. Diagram of a FLFC locked to a stable optical frequency reference based on an optical atomic reference and a stable Fabry–Perot cavity. The mode-locked laser is self-referenced (by the 1f–2f method) and provides two outputs, a precisely controlled optical frequency comb $f_i = (N' \pm i) f_{\rm rep} + f_0$ (where i is an integer and N' is the mode number closest to the frequency of the optical reference), and an electrical pulse train at $f_{\rm rep}$ that produces a microwave comb $f_{\rm m} = m f_{\rm rep}$.

that span from RF to UV, providing frequency uncertainties as small as the primary frequency standards can support.

Most high performance FLFC are based on mode-locked Ti:sapphire lasers, but other stable optical frequency combs that can span an octave or more and be self-referenced have now been demonstrated with Cr:LiSAF [173] (100 MHz, 100 mW, centred at 850 nm) Cr:Forsterite [174, 175] (400 MHz, 400 mW centred at 1.3 μm) and fibre lasers [176–179] (50 MHz, 100 mW, centred at 1.5 μm). It is intriguing that even though the average frequencies seem to be 'perfect', some recent experiments indicate that the noise properties of some types of optical frequency combs (particularly those in the near IR 1 to 2 μm range) can be more complicated and interesting than might be expected from the simple comb equation [175, 177].

5.1. Optical stability advantage

The accuracy of the primary atomic frequency standards has improved considerably, which means that the evaluation of systematic uncertainties now requires long averaging times as dictated by short-term instabilities. For state-of-the-art microwave atomic standards using quartz-based local oscillators (LOs), the short-term instability is limited to about 1×10^{-13}, and, if the atomic instability improves with time as $1 \times 10^{-13}\,\tau^{-1/2}$ (τ in seconds) it requires $\sim$3 h or more of signal averaging to make one measurement at the 1×10^{-15} level. Even measuring between hydrogen masers and a Cs fountain standard within the same building can take a day or two of averaging to make an accurate frequency measurement at the 1×10^{-15} level. Verifying systematic errors using these references then requires a sequence of long averages to evaluate all possible uncertainties [180–182]. The situation is much worse when trying to get the number out of the laboratory and to another location across the world, since averaging GPS signals might require 20 days to reach 1×10^{-15}. As accuracy improves, the process becomes increasingly difficult and quadratically more time consuming, thus short-term stability must be improved. Some alternatives exist for the microwave standards, for example, a UWA-SYRTE collaboration takes advantage of the better short-term stability of

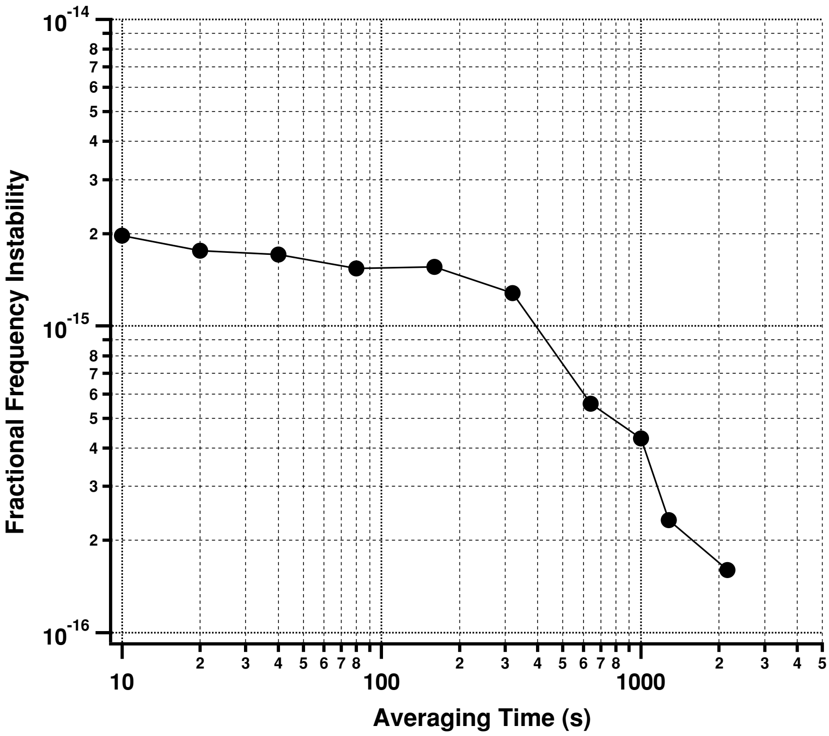

Figure 12. Relative frequency instability between the green Hg^+ (532 THz, 563 nm) and the red Ca (456 THz, 657 nm) optical frequency standards as measured with a FLFC locked to the 532 THz laser in an optical clock configuration similar to that shown in figure 11. The stable Hg^+ and Ca laser light was delivered to the FLFC via optical fibres (200 m and 30 m respectively) and both fibre links were Doppler cancelled [187, 188]. The data include the combined instability of the entire system (Hg^+, Ca, FLFC and two fibre links) and were measured at the 456 THz Ca frequency. The 10 s instability is consistent with expected performance of the atomic standards, but the flatness between 10 and 300 s is not well understood and is likely due to environmental effects that average down on longer times.

cryogenic sapphire whispering-gallery-mode oscillators to improve the short-term stability of Cs and Rb atomic fountains [183], and other schemes for utilizing more atoms are being developed [184–186]. For practicality, it is desirable that the instability at 1 s should be within a factor of 100 of the ultimate fractional frequency uncertainty, although this conclusion can be application dependent.

The high stability of optical frequency references provides a tremendous advantage and time saving when evaluating frequency offsets and uncertainties. Rapid measurements facilitate understanding and detection of unexpected frequency shifts (which usually dominate performance in actual systems), and speed enhances the ability to make corrections. An example helps to illustrate this point. Figure 12 shows the instability achieved by use of two independent optical frequency standards in the NIST building connected via optical fibres and compared with a FLFC similar to that shown in figure 11. The good short-term stability of the Hg^+ and Ca optical standards allows us to make relative frequency measurements with fractional uncertainties of $\approx 2 \times 10^{-15}$ in 10 s and averaging down to $\sim 1 \times 10^{-16}$ in 2000 s. The stability of this whole system could be improved, but is already more than good enough to make measurements between optical standards at the 1×10^{-16} level. The challenge now is for the optical frequency standards to set absolute frequency uncertainties at comparable levels.

6. Summary

In the past 100 years, optical frequency references based on narrow quantum transitions have played critical roles in the discovery of new physics and in testing fundamental theories, and have provided new tools and technologies for precision measurements that now serve as a foundation for our base units. As stability and accuracy improve, the effects of Einstein's relativity become increasingly important—a fractional frequency uncertainty of 10^{-18} would require knowledge of the geoid potential surface to <1 cm. Alternatively, perhaps OFR will be used for measuring gravitational potentials and making more stringent experimental tests of Einstein's ideas.

With recent advances, OFRs are demonstrating performance that makes them the compelling choices for the atomic clocks of the not-too-distant future. Some optical frequency references are demonstrating unprecedented frequency stability, and a few are beginning to show frequency reproducibility that is comparable to that of the best caesium atomic fountain standards. Several groups (including PTB, NRC, NPL, BNM-SYRTE, Max-Planck Garching, University of Tokyo, JILA, NIST ...) are working hard to develop systems with high accuracy and stability, and numerous interesting atomic systems are being considered. However, at this point, the optimal choices are not yet clear. It is conceivable that one atomic system will be optimal for the best short-term stability and another for ultimate accuracy (much as is the case today, where hydrogen masers typically provide good short-term stability and caesium clocks give ultimate accuracy).

When technologies have major advances, there is naturally an initial period of enthusiasm that the new discoveries will solve all our past problems. Perhaps we are just coming out of that phase now with optical atomic clocks; the systems are working and delivering spectacular data, but much hard work remains to reach the 10^{-18} accuracy levels that have been promised for more than 20 years now.

In any case, the high stability of optical frequency standards afford unsurpassed new capabilities, including femtosecond timing resolution when stabilized to atomic transitions, Doppler-cancelled optical fibre transmission, ultra-low-phase-noise microwaves and spectral and temporal coherence across the visible with coherence times of several seconds. We can even imagine future advanced systems using arrays of coherent atomic samples, as in spin-squeezing or Mott-insulator BEC experiments, that might allow Heisenberg-limited performance. In that case 10^6 coherent atoms, with a 500 THz transition, and a 1 Hz linewidth, could give an instability of $\sigma_y \approx \frac{\Delta\nu}{\nu_0\sqrt{N\tau}} \approx 10^{-21}\,\tau^{-1/2}$. It is probably unreasonable to extrapolate so far beyond the present performance and experience, but why not strive for limits that the physics says should be possible? One reason is that the present experimental technologies are certainly not up to the task. Laser oscillators are not nearly good enough, and technological constraints and even thermal noise introduce severe limitations [189]. But neither are there obvious fundamental reasons that these projections are unachievable. It seems remarkable that in principle just a few atoms and one second of averaging could provide sufficient information to achieve such high frequency resolution. If for some reasons these limits are not achievable, then there must be interesting science along the way. Only time will tell whether all the projections and promises can be realized; but if history is any indicator, Michelson's 'A Plea for Light' in 1888 [190] has proven well founded, and 117 years later we continue to add more voices to the chorus.

Acknowledgments

We thank our many colleagues at NIST and other laboratories around the world for important contributions to this paper and to the field of optical frequency standards. This is truly a world wide enterprise. We are particularly indebted to J L Hall for helpful comments, to T H Yoon from KRISS for the photo of the Michelson interferometer, and L Robertsson for tracking down historical details at BIPM.

References

[1] Barnes J A *et al* 1970 *IEEE Trans. Instrum. Meas.* **20** 105–20
[2] Sullivan D B, Allan D W, Howe D A and Walls F L (ed) 1990 *Characterization of Clocks and Oscillators* (NIST Tech. Note 1337)
[3] Hollberg L *et al* 2001 *IEEE J. Quantum Electron.* **37** 1502–13
[4] Metcalf H J and van der Straten P 1999 *Laser-Cooling and Trapping* (New York: Springer)
[5] Drever R W P, Hall J L, Kowalski F V, Hough J, Ford G M, Munley A J and Ward H 1983 *Appl. Phys.* B **31** 97–105
[6] Young B C, Cruz F C, Itano W M and Bergquist J C 1999 *Phys. Rev. Lett.* **82** 3799–802
[7] Hall J L and Ye J (ed) 2001 Laser frequency stabilization: standards, measurement and applications *Proc. SPIE* **4269**
[8] Wilpers G, Binnewies T, Degenhardt C, Sterr U, Helmcke J and Riehle F 2002 *Phys. Rev. Lett.* **89** 230801
[9] Rafac R J, Young B C, Beall J A, Itano W M, Wineland D J and Bergquist J C 2000 *Phys. Rev. Lett.* **85** 2462–5
[10] Katori H, Takamoto M, Pal'chikov V G and Ovsiannikov V D 2003 *Phys. Rev. Lett.* **91** 173005
[11] Holzwarth R, Zimmerman M, Udem T and Hansch T W 2001 *IEEE J. Quantum Electron.* **37** 1493–501
[12] Diddams S A, Jones D J, Ye J, Cundiff T, Hall J L, Ranka J K, Windeler R S, Holzwarth R, Udem T and Hänsch T W 2000 *Phys. Rev. Lett.* **84** 5102–5
[13] Telle H R, Steinmeyer G, Dunlop A E, Stenger J, Sutter D H and Keller U 1999 *Appl. Phys. B* **69** 327
[14] Bergquist J C, Diddams S A, Oates C W, Curtis E A, Hollberg L, Drullinger R E, Itano W M, Wineland D J and Udem T 2002 *Proc. XV Int. Conf. on Laser Spectroscopy* ed V V S Chu, A J Kerman and C Chen (Singapore: World Scientific) pp 106–14
[15] Bize S *et al* 2003 *Phys. Rev. Lett.* **90** 1508021
[16] Tamm C, Schneider T and Peik E 2004 Laser spectroscopy *Proc. XVI Int. Conf. Laser Spectroscopy 2003* ed P Hannaford and H Bachor (Singapore: World Scientific) pp 40–8
[17] Margolis H S, Barwood G, Huang G, Klein H A, Lea S N, Szymaniec K and Gill P 2004 *Science* **306** 1355
[18] Hocker L O, Javan A, Rao D R, Frenkel L and Sullivan T 1967 *Appl. Phys. Lett.* **10** 147–9
[19] Javan A 1968 *Ann. New York Acad. Sci.* **168** 715–20
[20] Jennings D A, Evenson K M and Knight D J E 1986 *Proc. IEEE* **74** 168–79
[21] Klement'ev V M, Matyugin Y A and Chebotaev V P 1978 *Sov. J. Quantum Electron.* **8** 953–8
[22] Chebotayev V P 1979 *Radio Sci.* **14** 573–84
[23] Baird K M, Evenson K M, Hanes G R, Jennings D A and Petersen F R 1979 *Opt. Lett.* **4** 263–4
[24] Gill P (ed) 2002 Frequency standards and metrology *Proc. 6th Symp.* (Singapore: World Scientific)
[25] Hall J L 2000 *IEEE J. Sel. Top. Quantum Electron.* **6** 1136–44
[26] Madej A A and Bernard J E 2001 Frequency measurements and control: advanced techniques and future trends *Topics in Applied Physics* vol 79 ed A N Luiten (Heidelberg: Springer) pp 153–94
[27] Fisk P T H 1997 *Rep. Prog. Phys.* **60** 761–817
[28] Hall J L and Ye J 2003 *IEEE Trans. Instrum. Meas.* **52** 227–31
[29] Baklanov E V and Pokasov P 2003 *Quantum Electron.* **33** 383–400
[30] Ye J, Schnatz H and Hollberg L 2003 *IEEE J. Sel. Top. Quantum Electron.* **9** 1041–58
[31] Will C M 1981 *Theory and Experiment in Gravitational Physics* (Cambridge: Cambridge University Press)
[32] Zhang Y Z 1997 *Special Relativity and its Experimental Foundations* vol 4 (Singapore: World Scientific)
[33] Essen L and Gordon-Smith A C 1948 *Proc. R. Soc. Lond.* A **194** 348–61
[34] Petley B W 1983 *Nature* **303** 373–6
[35] Michelson A A and Morley E W 1887 *Am. J. Sci.* **34** 333
[36] Michelson A A 1889 *Am. J. Sci.* **38** 225
[37] Michelson A A 1893 *Astron. Astrophys.* **12** 556–60

[38] Bennett J M, McAllister D T and Cabe G M 1973 *Appl. Opt.* **12** 2253–79
[39] Michelson A A 1911 *Science* **34** 893–902
[40] Benoit R, Fabry C and Perot A 1907 *Comptes Rendus* **144** 1082–6
[41] BIPM-web-site Bureau International des Poids et Mesures http://www.bipm.org/
[42] Giacomo P 1984 *Metrologia* **20** 25–30
[43] BNM-SYRTE, l'Obsrvatoire de Paris, http://www.obspm.fr/, BNM-SYRTE Bureau National de Metrologie-Systèmes de Référence Temps Espace, http://opdaf1.obspm.fr/
[44] Tuinstra F 2004 *Phys. Today* **57** 16
[45] Froome K D and Essen L 1969 *The Velocity of Light and Radio Waves* (London: Academic)
[46] Michelson A A 1924 *Astrophys. J.* **60** 256–61
[47] Michelson A A 1927 *Astrophys. J.* **65** 1–22
[48] Bowie W 1927 *Astrophys. J.* **65** 14–22
[49] Shankland R S 1967 *Am. J. Phys.* **35** 1095–6
[50] Evenson K M, Wells J S, Danielso Bl, Day G W and Petersen F R 1972 *Phys. Rev. Lett.* **29** 1346–9
[51] Essen L and Parry J V L 1955 *Nature* **176** 280–2
[52] Weiss R 1966 Contribution to Festschrift for J R Zacharias, http://emvogil-3.mit.edu/~weiss/documents/zachfountain.pdf
[53] Lyons H 1952 *Ann. New York Acad. Sci.* **55** 831–71
[54] Jones T 2000 *Splitting the Second, the Story of Atomic Time* (Bristol: Institute of Physics Publishing)
[55] Evenson K M, Wells J S, Mullen L O and Day G W 1972 *Appl. Phys. Lett.* **20** 133–4
[56] Barger R L and Hall J L 1973 *Appl. Phys. Lett.* **22** 196–9
[57] Rowley W R C and Wallard A J 1973 *J. Phys. E: Sci. Instrum.* **6** 647–52
[58] Wood P T, Shotton K C and Rowley W R C 1978 *Appl. Opt.* **17** 1048–54
[59] Baird K M, Smith D S and Berger W E 1973 *Opt. Commun.* **7** 107–9
[60] Hall J L 2004 Private communication
Giacomo P 2003 Private communication
[61] BIPM-Director's-Report, Interferometric measurements, copies kindly provided by L Robertsson of BIPM (1969–1970)
[62] Blaney T G, Bradley C C, Edwards G J, Jolliffe B W, Knight D J E, Rowley W R C, Shotton K C and Woods P T 1974 *Nature* **251** 46–7
[63] Rowley W R C, Jolliffe B W, Shotton K C, Wallard A J and Woods P T 1976 *Opt. Quantum Electron.* **8** 1–14
[64] Baird K M, Smith D S and Whitford B G 1979 *Opt. Commun.* **31** 367–8
[65] Bay Z, Luther G G and White J A 1972 *Phys. Rev. Lett.* **29** 189–92
[66] Rowley W R C, Shotton K C and Woods P T 1980 *Opt. Commun.* **34** 429–30
[67] Hanes G R, Baird K M and DeRemigis J 1973 *Appl. Opt.* **12** 1600–5
[68] Pollock C R, Jennings D A, Petersen F R, Wells J S, Drullinger R E, Beaty E C and Evenson K M 1983 *Opt. Lett.* **8** 133–5
[69] Jennings D A, Pollock C R, Petersen F R, Drullinger R E, Evenson K M, Wells J S, Hall J L and Layer H P 1983 *Opt. Lett.* **8** 136–8
[70] Giacomo P 1984 *Am. J. Phys.* **52** 607–13
[71] Schnatz H, Lipphardt B, Helmcke J, Riehle F and Zinner G 1996 *Phys. Rev. Lett.* **76** 18–21
[72] Quinn T J 2003 *Metrologia* **40** 103–33
[73] BIPM-mep http://www.bipm.org/en/home/mep.html
[74] Madej A A, Bernard J E, Robertsson L, Ma L S, Zucco M and Windeler R S 2004 *Metrologia* **41** 152–60
[75] Bernard J E, Madej A A, Siemsen K J and Marmet L 2001 *Opt. Commun.* **187** 211–8
[76] Picard S *et al* 2003 *IEEE Trans. Instrum. Meas.* **52** 236–9
[77] Ma L S, Picard S, Zucco M, Chartier J M, Robertsson L and Windeler R S 2004 *Metrologia* **41** 65–8
[78] Ma L S, Robertsson L, Picard S, Chartier J-M, Karlsson H, Prieto E, Ranka J K and Windeler R S 2003 *IEEE Trans. Instrum. Meas.* **32** 232–5
[79] Michelson A A and Morley E W 1887 *Am. J. Sci.* **34**
[80] 1928 *Proc. Conf. on the Michelson–Morley Experiment (Mt Wilson Observatory, Pasadena, CA, 4–5 February 1928) Astrophys. J.* **68** 341–402
[81] Shankland R S 1964 *Am. J. Phys.* **32** 16–35
[82] Livingston D M 1973 *The Master of Light: A Biography of Albert A Michelson* (New York: Charles Scribner)
[83] Shankland R S 1973 *Appl. Opt.* **12** 2280–7
[84] Jaseja T S, Javan A, Murray J and Townes C H 1964 *Phys. Rev.* **133** A1221–5
[85] Brillet A and Hall J L 1979 *Phys. Rev. Lett.* **42** 549–52
[86] Hils D and Hall J L 1990 *Phys. Rev. Lett.* **64** 1697–700

[87] Robertson H P 1949 *Rev. Mod. Phys.* **21** 378–82
[88] Mansouri R and Sexl R U 1977 *Gen. Rel. Grav.* **8** 497–513
[89] Mansouri R and Sexl R U 1977 *Gen. Rel. Grav.* **8** 515–24
[90] Born M 1962 *Einstein's Theory of Relativity* (New York: Dover)
[91] Roberts T 2000 http://math.ucr.edu/home/baez/physics/Relativity/SR/experiments.html
[92] Braxmaier C, Muller H, Pradl O, Mlynek J, Peters A and Schiller S 2002 *Phys. Rev. Lett.* **88** 010401
[93] Muller H, Herrmann S, Braxmaier C, Schiller S and Peters A 2003 *Phys. Rev. Lett.* **91** 020401
[94] Wolf P, Bize S, Clairon A, Luiten A N, Santarelli C and Tobar M E 2003 *Phys. Rev. Lett.* **90** 060402
[95] Wolf P, Bize S, Clairon A, Santarelli C, Luiten A N and Tobar M E 2003 Tests of Lorentz invariance using a microwave resonator: an update *Proc. EFTF & FCS Joint Meeting (Tampa, FL 2003)* (Piscataway, NJ: IEEE) pp 205–10
[96] Snyder J J and Hall J L 1975 Laser spectroscopy *Proc. 2nd Int. Conf. (Megeve, 23–27 June 1975)* vol 43 ed J C P-P S Haroche, T W Hänsch and S E Harris (Berlin: Springer) pp 6–17
[97] Riis E, Andersen L-U A, Bjerre N, Poulsen O, Lee S A and Hall J L 1988 *Phys. Rev. Lett.* **60** 81–4
[98] McGowen R W, Giltner D M, Sternberg S J and Lee S A 1993 *Phys. Rev. Lett.* **70** 251–4
[99] Saathoff G, Karpuk S, Eisenbarth U, Huber G, Krohn S, Munoz Horta R, Reinhardt S, Schwalm D, Wolf A and Gwinner G 2003 *Phys. Rev. Lett.* **91** 190403
[100] Ashby N 2003 *Liv. Rev. Rel.* **6** 1–45 www.livingreviews.org/Articles/Volume6/2003-1ashby
[101] Heavner T P, Jefferts S R, Donley E A, Shirley J H and Parker T E 2004 *Proc. Joint Mtg. IEEE Intl. Freq. Cont. Symp. UFFC Conf.*
[102] Jefferts S R, Heavner T P and Donley E A 2004 *Japan. J. Appl. Phys.* **43** 2803–7
[103] Dyson F J 1972 *Aspects of Quantum Theory* ed A S and E P Wigner (Cambridge: Cambridge University Press) pp 213–36
[104] Prestage J D, Tjoelker R L and Maleki L 1995 *Phys. Rev. Lett.* **74** 3511–4
[105] Domnin Y S, Kopylov L N, Malimon A N, Tenishev V P and Shumyatskii P S 1996 *Quantum Electron.* **26** 1081–4
[106] Karshenboim S G 2000 *Can. J. Phys.* **78** 639–78
[107] Karshenboim S G 2002 *Laser Physics at the Limits* vol 34 ed D M H Figger and C Zimmerman (Berlin: Springer) pp 165–76
[108] Peik E, Lipphardt B, Schnatz H, Schneider T, Tamm C and Karshenboim S G 2004 *Phys. Rev. Lett.* **93** 170801
[109] Fischer M *et al* 2004 *Phys. Rev. Lett.* **92** 230802
[110] Bluhm R, Kostelecky V A, Lane C D and Russell N 2002 *Phys. Rev. Lett.* **88** 090801
[111] Dzuba V A, Flambaum V V and Webb J K 1999 *Phys. Rev.* A **59** 230–7
[112] Flambaum V 2004 Laser spectroscopy *Proc. XVI Int. Conf. on Laser Spectroscopy* ed P Hannaford and H Bachor (Singapore: World Scientific) pp 49–57
[113] Webb J K, Murphy M T, Flambaum V, Dzuba V A, Barrow J D, Churchill C W, Prochaska J X and Wolfe A M 2001 *Phys. Rev. Lett.* **87** 091301
[114] Kozlov M G, Korol V A, Berengut J C, Dzuba V A and Flambaum V V 2004 *Phys. Rev.* A **70** 062108
[115] Quast R, Reimers D and Levshakov S A 2004 *Astron. Astrophys.* **415** L7–11
[116] Chand H, Srianand R, Petitjean P and Aracil B 2004 *Astron. Astrophys.* **417** 853–71
[117] Marion H *et al* 2003 *Phys. Rev. Lett.* **90** 150801
[118] Diddams S A, Bergquist J C, Jefferts S R and Oates C W 2004 *Science* **306** 1318–24
[119] Sterr U, Degenhardt C, Stoehr H, Lisdat C, Schnatz H, Helmcke J, Riehle F, Wilpers G, Oates C W and Hollberg L 2004 *C. R. Phys.* **5** 845–55
[120] Oates C W, Curtis E A and Hollberg L 2000 *Opt. Lett.* **25** 1603–5
[121] Madej A A, Bernard J E, Dube P, Marmet L and Windeler R S 2004 *Phys. Rev.* A **70** 012507
[122] Dehmelt H 1982 *IEEE Trans. Instrum. Meas.* **31** 83–7
[123] Oskay W H, Itano W M and Bergquist J C 2005 *Phys. Rev. Lett.* at press
[124] Itano W M 2000 *J. Res. Natl Bur. Stand.* **105** 829–37
[125] Wineland D J, Bergquist J C, Bollinger J J, Drullinger R E and Itano W M 2002 *Quantum Computers and Atomic Clocks (St Andrews, Fife, Scotland)* (Singapore: World Scientific) pp 361–7
[126] Schmidt P O, Rosenband T, Langer C, Itano W M, Bergquist J C and Wineland D J 2005 submitted
[127] Itano W M 2004 Private communication
[128] Quessada A, Kovacich R P, Courtillot I, Clairon A, Santarelli C and Lemonde P 2003 *J. Opt. B: Quantum Semi-Class. Opt.* **5** S150–4
[129] Curtis E A, Oates C W and Hollberg L 2001 *Phys. Rev.* A **64** 031403
[130] Binnewies T, Wilpers G, Sterr U, Riehle F, Helmcke J, Mehlstaubler T E, Rasel E M and Ertmer W 2001 *Phys. Rev. Lett.* **87** 123002

[131] Bordé C J, Salomon C, Avrillier S, Van Lerberghe A, Breant C, Bassi D and Scoles G 1984 *Phys. Rev.* A **30** 1836–48

[132] Wineland D J, Bergquist J C, Itano W M and Drullinger R E 1980 *Opt. Lett.* **5** 245–7

[133] Oates C W, Bondu F, Fox R W and Hollberg L 1999 *Eur. Phys. J.* D **7** 449–60

[134] Maleki L 1998 *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **45** 876

[135] Presti L L, Rovera D and Marchi A D 1998 *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **45** 899–905

[136] Santarelli G, Audoin C, Makdissi A A, Laurent P, Dick G J and Clairon A 1998 *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **45** 887–94

[137] Takamoto M and Katori H 2003 *Phys. Rev. Lett.* **91** 223001

[138] Courtillot I, Quessada A, Kovacich R P, Brusch A, Kolker D, Zondy J J, Rovera D G and Lemonde P 2003 *Phys. Rev.* A **68** 030501

[139] Ruschewitz F, Peng J L, Hinderthur H, Schaffrath N, Sengstock K and Ertmer W 1998 *Phys. Rev. Lett.* **80** 3173–6

[140] Cesar C L, Fried D G, Killian T C, Polcyn A D, Sandberg J C, Yu I A, Greytak T J, Kleppner D and Doyle J M 1996 *Phys. Rev. Lett.* **77** 255–8

[141] Niering M *et al* 2000 *Phys. Rev. Lett.* **84** 5496–9

[142] Porsev S G, Derevianko A and Fortson E N 2004 *Phys. Rev.* A **69** 021403

[143] Badr T, Plimmer M D, Juncar P, Himbert M E, Silver J D and Rovera D G 2004 *Eur. Phys. J.* D **31** 3–10

[144] Xu X-Y, Loftus T, Dunn J W, Greene C H, Hall J L, Gallagher A and Ye C Y 2003 *Phys. Rev. Lett.* **91** 193002

[145] Yoon T H, Park C Y and Park S J 2004 *Phys. Rev.* A **70** 061803

[146] Karshenboim S G, Pavone F S, Bassani G F, Inguscio M and Hänsch T W (ed) 2001 *The Hydrogen Atom: Precision Physics of Simple Atom Systems* (Berlin: Springer)

[147] Kolachevsky N, Fischer M, Karshenboim S G and Hänsch T W 2004 *Phys. Rev. Lett.* **92** 033003

[148] Cancio Pastor P, Guisfredi G, De Natale P, Hagel G, de Mauro C and Inguscio M 2004 *Phys. Rev. Lett.* **92** 023001

[149] Hong T, Cramer C, Nagourney W and Fortson E N 2005 *Phys. Rev. Lett.* **94** 050801

[150] Yasuda M and Katori H 2004 *Phys. Rev. Lett.* **92** 153004

[151] International Telecommunications Union 2002 Frequency grid for optical communication channels (ITU Recommendation G.694.1 (06/02))

[152] Gilbert S L, Swann W and Dennis T 2001 *Proc. Soc. Photo-Opt. Instrum. Eng.* **4269** 184–91

[153] Dennis T, Curtis E A, Oates C W, Hollberg L and Gilbert S L 2002 *J. Lightwave Technol.* **20** 776–82

[154] Udem T, Reichert J, Holzwarth R and Hänsch T W 1999 *Opt. Lett.* **24** 881–3

[155] Udem T, Reichert J, Holzwarth R and Hänsch T W 1999 *Phys. Rev. Lett.* **82** 3568–71

[156] Eckstein J N, Ferguson A I and Hänsch T W 1978 *Phys. Rev. Lett.* **40** 847–50

[157] Reichert J, Holzwarth R, Udem T and Hänsch T W 1999 *Opt. Commun.* **172** 59–68

[158] Jones D J, Diddams S A, Ranka J K, Stentz A, Windeler R S, Hall J L and Cundiff S T 2000 *Science* **288** 635–9

[159] Diddams S A, Ye J and Hollberg L 2005 *Femtosecond Optical Frequency Comb: Principle, Operation and Applications* ed E S Cundiff and J Ye (New York: Springer) pp 225–62

[160] Udem T, Holzwarth R and Hänsch T W 2002 *Nature* **416** 233–7

[161] Cundiff S T and Ye J 2003 *Rev. Mod. Phys.* **75** 325–42

[162] Hall J L, Ye J, Diddams S A, Ma L S, Cundiff S T and Jones D J 2001 *IEEE J. Quantum Electron.* **37** 1482–92

[163] Diddams S A *et al* 2001 *Science* **293** 825–8

[164] Bartels A, Diddams S A, Ramond T M and Hollberg L 2003 *Opt. Lett.* **28** 663–5

[165] Bartels A, Diddams S A, Oates C W, Wilpers G, Bergquist J C, Oskay W H and Hollberg L 2004 *Opt. Lett.* **30** 667–9

[166] Hollberg L, Diddams S A, Bartels A, McFerran J, Ivanov E N, Wilpers G, Oates C W, Oskay W H and Bergquist J C 2004 *Generation of Microwaves with Ultra-Low Phase-Noise from an Optical Clock (Ogunquit, Maine)* (Piscataway, NJ: IEEE)

[167] Holzwarth R, Udem T, Hänsch T W, Knight J C, Wadsworth W J and Russell P S J 2000 *Phys. Rev. Lett.* **85** 2264–7

[168] Diddams S A, Hollberg L, Ma L S and Robertsson L 2002 *Opt. Lett.* **27** 58–60

[169] Ma L S, Robertsson L, Picard S, Zucco M, Bi Z, Wu S and Windeler R S 2004 *Opt. Lett.* **29** 641–3

[170] Ma L S, Bi Z, Bartels A, Robertsson L, Zucco M, Windeler R S, Wilpers G, Oates C W, Hollberg L and Diddams S A 2004 *Science* **303** 1843–5

[171] Zimmerman M, Gohle C, Holzwarth R, Udem T and Hänsch T W 2004 *Opt. Lett.* **29** 310–2

[172] Stenger J, Schnatz H, Tamm C and Telle H R 2002 *Phys. Rev. Lett.* **88** 073601

[173] Holzwarth R, Zimmerman M, Udem T, Hänsch T W, Russbuldt P, Gabel K, Poprawe R, Knight J C, Wadsworth W J and Russell P S 2001 *Opt. Lett.* **26** 1376–8
[174] Thomann I, Bartels A, Corwin K L, Newbury N R, Hollberg L, Diddams S A, Nicholson J W and Yan M F 2003 *Opt. Lett.* **28** 1368–70
[175] Kim K, Washburn B R, Wilpers G, Oates C W, Hollberg L, Newbury N R, Diddams S A, Nicholson J W and Yan M F 2005 *Opt. Lett.* **30** 932–4
[176] Tauser F, Leitenstorfer A and Zinth W 2003 *Opt. Exp.* **11** 594–600
[177] Washburn B R, Diddams S A, Newbury N R, Nicholson J W, Yan M F and Jorgensen C G 2004 *Opt. Lett.* **29** 250–2
[178] Hong F L, Minoshima K, Onae A, Inaba H, Takada H, Hirai A and Matsumoto H 2003 *Opt. Lett.* **28** 1516–8
[179] Hundertmark H, Wandt D, Fallnich C, Haverkamp N and Telle H R 2004 *Opt. Exp.* **12** 770–5
[180] Jefferts S R 2002 *Metrologia* **39** 321–36
[181] Weyers S, Hubner U, Schroder R, Tamm C and Bauch A 2001 *Metrologia* **38** 343–52
[182] Levi F, Lorini L, Calonico D and Godone A 2004 *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **51** 1216–24
[183] Santarelli C, Laurent P, Lemonde P, Clairon A, Mann A G, Chang S, Luiten A N and Salomon C 1999 *Phys. Rev. Lett.* **82** 4619–22
[184] Fertig C and Gibble K 2000 *Phys. Rev. Lett.* **85** 1622–5
[185] Levi F, Godone A, Lorini L, Jefferts S R, Heavner T P and Calosso C 2002 *The Multiple Velocity Fountain: A New Scheme for the Cold Collision Frequency Shift Reduction (St Andrews, Fife, Scotland)* (Singapore: World Scientific) pp 466–8
[186] Fertig C, Legere R, Rees J I, Gibble K, Kokkelmans S, Verhaar B J, Prestage J D, Klipstein W, Seidel D and Thompson R 2002 *Cold Collision Frequency Shifts and Laser-Cooled* ^{87}*Rb Clocks (St Andrews, Fife, Scotland)* (Singapore: World Scientific) pp 213–21
[187] Bergquist J C, Itano W M and Wineland D J 1994 *Proc. Int. School of Physics 'Enrico Fermi', Frontiers in Laser Spectroscopy* ed T W Hänsch and M Inguscio (Amsterdam: North-Holland) pp 359–76
[188] Ma L-S, Jungner P, Ye J and Hall J L 1994 *Opt. Lett.* **19** 1777–9
[189] Numata K, Kemery A and Camp J 2004 *Phys. Rev. Lett.* **93** 250602
[190] Michelson A A 1888 *Proc. Am. Assoc. Adv. Sci.* **37** 67–78