REVIEW

Standards of Time and Frequency at the Outset of the 21st Century

S. A. Diddams,* J. C. Bergquist, S. R. Jefferts, C. W. Oates

After 50 years of development, microwave atomic clocks based on cesium have achieved fractional uncertainties below 1 part in 10^{15}, a level unequaled in all of metrology. The past 5 years have seen the accelerated development of optical atomic clocks, which may enable even greater improvements in timekeeping. Time and frequency standards with various levels of performance are ubiquitous in our society, with applications in many technological fields as well as in the continued exploration of the frontiers of basic science. We review state-of-the-art atomic time and frequency standards and discuss some of their uses in science and technology.

As important as "time" might be to those who are navigators, scientists, or even musicians, it is no more than an arbitrary parameter that is used to describe dynamics, or the mechanics of motion. David Mermin was struck by this as he wondered about the role that time and space would play in physics in the next century (*1*):

> *How can people talk about spacetime turning into a foam at the Planck scale when we barely manage to define space and time at the atomic scale? Time, for example, is nothing more than an extremely convenient and compact way to characterize the correlations between objects we can use as clocks, and clocks tend to be macroscopic. To be sure, we can generate frequencies from atoms and correlate them with macroscopic clocks, but the shorter the length scale, the more it looks like you're talking about energies divided by Planck's constant. The connections with clocks become increasingly indirect. There seems to me to be a considerable danger here of imposing on an utterly alien realm a useful bookkeeping device we've merely invented for our own macroscopic convenience.*

The definition of time can be puzzling exactly because of the apparent arbitrariness that Mermin described. It is through the external or internal periodic dynamics of one object that we define time, and armed with that time scale, we can characterize the dynamics of other objects—an oddly circular argument. Another conundrum: How do we determine the period (or its inverse, frequency) of our time standard, or any clock, to be uniform? Clearly, time is relative and several time sources must be compared toward establishing the most stable and accurate definition of the second, our base unit of time in the international system (SI).

In view of this, there is considerable irony in the fact that the second is the most accurately realized unit of measurement, with fractional uncertainty now below 1 part in 10^{15}. Moreover, it could be argued that the technologies based on the arbitrarily defined second have had an impact like few others in our modern society. Everyday systems including the electric power grid, cell phones, the Internet, and the Global Positioning System (GPS) depend critically on time and frequency standards for continued operation. Because of its position of metrological preeminence, the second is also used to define three other SI units (meter, candela, and ampere), and several other important physical quantities are defined or measured in terms of the second. For example, accepting that the speed of light is a constant, the meter is defined as the path length traveled by light in a vacuum during the time interval of 1/299,792,458 of a second. As a result, lasers with a known and fixed frequency have become the standards for length metrology, where they guide precision measurements of physical distances (e.g., in the etching and lithography of semiconductor wafers). Another example is the definition of the volt, which can be obtained via the Josephson effect in terms of the product of a physical constant and the frequency. For these and other reasons, the development and operation of high-quality time and frequency standards is an important endeavor at National Metrology Institutes (NMI) around the world, with consequences that reach into our daily lives.

In many research laboratories, the scientific impact of precision time and frequency measurements has been considerable. Examples include the prediction of gravitational radiation from binary pulsars (*2*) and the most precise direct measurement of the gravitational red shift (*3*). Additionally, the development of time and frequency standards over the past 60 years has gone hand-in-hand with many scientific advances in the fields of atomic, molecular, and optical physics. Modern atomic clocks have their roots in the microwave spectroscopy experiments of Stern, Rabi, and Ramsey. Many of the techniques that are used in modern atomic clocks resulted from the development of the maser, the laser, and the new field of laser spectroscopy that followed. These developments led to laser cooling and trapping of atoms and ions, which now provide clockmakers with isolated and nearly motionless quantum references for what are now the best clocks in existence.

In labeling the time of a physical event, one must count the number of cycles (and perhaps fractions of cycles) of some periodic occurrence relative to an agreed-upon time origin. For some situations, the time origin is of great importance; in other cases, it is only the interval, or time difference, between two events that is of interest. In what follows, we focus on the latter and discuss the issues surrounding the generation and characterization of the source of the periodic events (often called a frequency standard) with which time intervals are generated and measured. As many excellent reviews on the development of atomic frequency standards already exist (*4*–*6*), we concentrate on the most accurate atomic clock—the cesium fountain clock—and then discuss the new optical clocks, which are anticipated to be the atomic timepieces of the future.

Historical Background

The best choice for a timekeeping device is an object whose dynamical period is well characterized, not easily perturbed, and (ideally) constant. A natural, macroscopic candidate that could be used to define the unit of time is some phenomenon of nature, whose period is especially uniform. Figure 1 compares the performance of a few important clocks from recent history. For many centuries, the daily rotation of Earth on its axis seemed to offer a uniform time base, but as time standards and measurement techniques improved, the length of a day was found to fluctuate and generally grow longer (which was attributed in part to tidal friction). Astronomers seeking a more stable unit of time chose the period of the orbital

---

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

*To whom correspondence should be addressed. E-mail: sdiddams@boulder.nist.gov

motion of Earth about the Sun (nominally 1 year) as the basis for the definition of the second. In 1956, the Ephemeris Second (1/31,556,925.9747 of the tropical year 1900) was formally adopted by the General Conference of Weights and Measures as the best measure of time.

Although the orbital motion of Earth in the solar system might be more uniform than the solar day, its period was impractically long for most purposes and was likely to suffer unpredictable changes and aging effects (hence, the definition of Ephemeris Time based on a particular solar year). Already when this definition of the second was adopted, scientists were investigating resonances or transitions in microscopic atomic systems as a more suitable means for defining time intervals and frequency. Many transitions between energy states in well-isolated atomic systems are highly immune to perturbations that would change the atomic resonance frequency ν_o, making these systems ideal candidates for clocks. Quantum mechanics dictates that the energies of a bound system (e.g., an electron bound to an atom) have discrete values. Hence, an atom or molecule can make a transition between two energy levels (E_1 and E_2) by the absorption or emission of energy in the form of electromagnetic radiation having the precise frequency $\nu_o = |E_1 - E_2|/h$, where h is the Planck constant. On the basis of this principle, most atomic frequency standards (atomic clocks) work by steering the frequency of an external oscillator to match a particular value of ν_o.

The first atomic clocks owe their genesis to the explosion of advances in quantum mechanics and microwave electronics before and during the Second World War. Much of the seminal work specific to clock development was done by Rabi. Although he may have suggested using cesium as the reference for an atomic clock as early as 1945, it was the inversion transition in the ammonia molecule at ~23.8 GHz that served as the reference for the first "atomic" clock in 1949 (*7*). In 1955, the first operational cesium atomic clock was built at the National Physical Laboratory, Teddington, UK (*8*). It was immediately noted that observations of the Moon over a period of several years would be required to determine Ephemeris Time with the same precision as was achieved in a matter of minutes by the first cesium clock (*9*). Although the fate of astronomically defined time seemed certain, more than a decade passed before the definition of the SI second was changed to be 9,192,631,770 cycles of the ground-state hyperfine splitting of the unperturbed cesium atom (*10*).

Cesium Clocks

Clocks are often characterized by their stability and accuracy. Stability is a measure of the degree to which the interval between "clock ticks" remains constant. Accuracy is a measure of how well the time between the clock ticks matches the defined second on the cesium hyperfine splitting. An accurate clock is necessarily stable over long intervals, but not all stable clocks are accurate.

On paper, clocks based on atomic processes are ideal, but there are fundamental as well as practical limitations to both their stability and accuracy. Atoms absorb or emit energy over a small range surrounding ν_o, not at ν_o alone. All other parameters being equal, the stability of an atomic clock is proportional to ν_o and inversely proportional to the small spread $\Delta\nu$ (linewidth) of absorption frequencies. This is more typically expressed in terms of the fractional frequency instability,

$$\sigma \propto \frac{\Delta\nu}{\nu_o}\,\frac{1}{S/N} \qquad (1)$$

where S/N is the signal-to-noise ratio, and high stability is equivalent to a smaller value of σ. From this expression we see that atomic clocks will generally benefit from operating at higher frequencies with transitions having narrow linewidths. In addition, Eq. 1 shows that the instability decreases with an increase in the S/N ratio with which the absorption signal is measured.

In the operation of an atomic clock, the atom must be illuminated by the electromagnetic radiation emitted from an external oscillator. Cesium clocks require a microwave oscillator, whereas the optical clocks discussed below require an optical oscillator (laser). The difficult challenge is to then tune the oscillator frequency to exactly match ν_o. There is always some ambiguity in this process because, as mentioned above, the resonance frequency has a nonzero linewidth associated with it. One factor that can limit the minimum observed linewidth of the reference transition is the time that the atom is in the radiation field. In those situations,

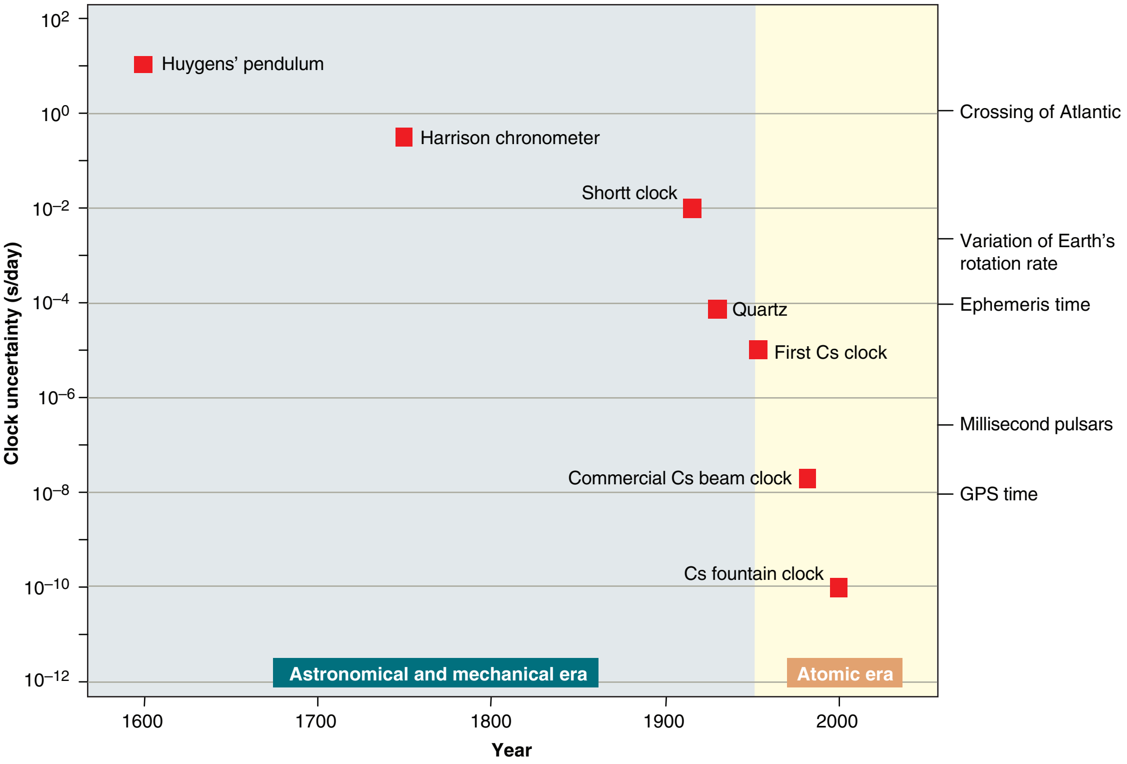

Fig. 1. Some of the major milestones in the improvement of clocks over the past 400 years. A clock with ~1 s uncertainty per day was required for crossing the Atlantic in 1750, and the Harrison chronometer demonstrated advancements that allowed such accurate navigation. The Shortt clock is the most accurate mechanical clock. Ephemeris time, as determined from astronomical observations, was accurate to ~0.1 ms per day. A clock based on a pulsar could have less than 1 μs of uncertainty per day. GPS time represents what typically can be achieved with a single receiver.

the observed linewidth of the resonance decreases as the measuring time increases.

Many other effects can act to degrade the stability and the accuracy of an atomic clock. The motion of the atoms introduces uncertainty by causing apparent shifts in the resonance frequencies (the Doppler effect). Similarly, collisions between atoms are a source of frequency shifts and linewidth broadening. Defects in the electronic measuring equipment as well as stray electromagnetic fields (including the ever-present thermal radiation) perturb the resonance frequency and introduce potential errors. Therefore, a good atomic clock must not only establish a steady periodic signal but must also minimize these potential errors.

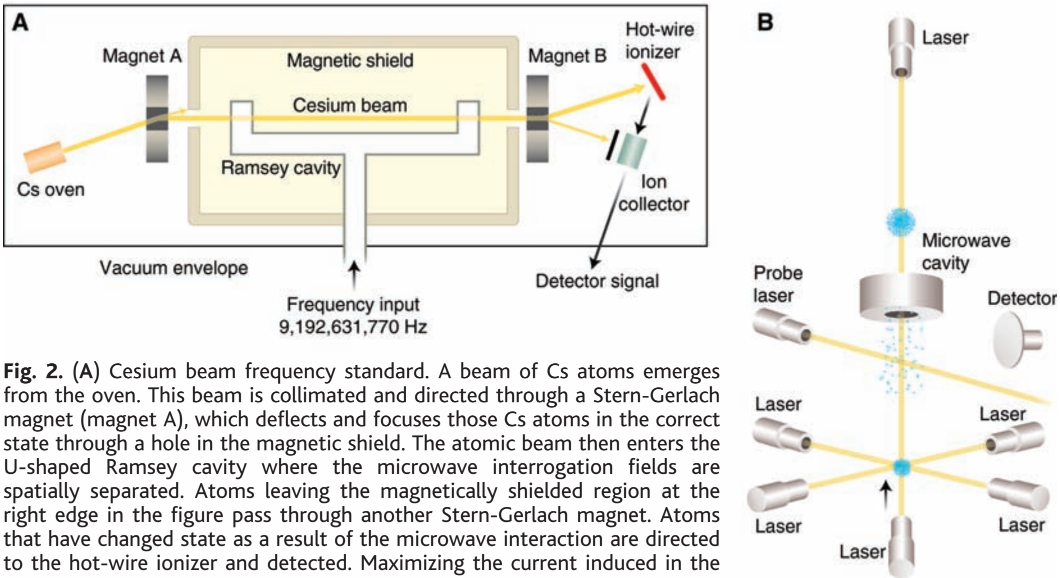

Fig. 2. (**A**) Cesium beam frequency standard. A beam of Cs atoms emerges from the oven. This beam is collimated and directed through a Stern-Gerlach magnet (magnet A), which deflects and focuses those Cs atoms in the correct state through a hole in the magnetic shield. The atomic beam then enters the U-shaped Ramsey cavity where the microwave interrogation fields are spatially separated. Atoms leaving the magnetically shielded region at the right edge in the figure pass through another Stern-Gerlach magnet. Atoms that have changed state as a result of the microwave interaction are directed to the hot-wire ionizer and detected. Maximizing the current induced in the hot-wire ionizer maximizes the number of atoms making the transition and thus assures that the frequency of the microwaves matches the atomic resonance frequency. (**B**) Cs fountain clock. The basic operation of the Cs fountain proceeds in a sequence of steps. First, a sample of ~10^8 cesium atoms is laser-cooled at the intersection of six laser beams to below 1 μK. These atoms are next "launched" upward at ~4 m/s by frequency detuning of the laser beams. The lasers are then turned off and the Cs atoms continue along their ballistic flight path. These atoms now enter the microwave cavity. The passage through the cavity on the way up provides the first pulse of the two-pulse (Ramsey) microwave interrogation sequence. The atoms reach apogee above the microwave cavity and eventually fall through the microwave cavity a second time. Atoms that have made a transition in state due to the interaction with the microwave field are detected optically with a laser.

In the early work of Rabi, the atomic resonance was interrogated with the radiation from one long microwave pulse. This provided the needed long interaction time between the atom and microwave field, but led for various reasons to the output frequency of the standard being subject to Doppler shifts and other sensitivities. Ramsey's method of separated oscillatory fields (*11*) provided a critical improvement that has been adopted by all modern primary frequency standards. In Ramsey's method, the microwave excitation is done in two relatively short pulses at the beginning and end of the interaction zone. This two-pulse process (now known as Ramsey interrogation) reduces these sensitivities by factors of 10 to 100 or more.

A schematic of a conventional magnetically state-selected atomic beam cesium standard is shown in Fig. 2A. The design can be directly traced back to Rabi's and Ramsey's seminal works; essentially all commercial cesium atomic clocks use this general design, as do the cesium clocks in the GPS satellites. Moreover, this design provided the world with all of its primary frequency standards up until about 1990, and even today roughly 300 such clocks at more than 50 NMIs are included in the averaged international time scale known as Coordinated Universal Time (UTC) (*12*). The CS-1 clock of the Physikalisch-Technische Bundesanstalt (PTB) has the lowest stated systematic frequency inaccuracy of any clock of this type ever built, with $\delta\nu/\nu_o \sim 8 \times 10^{-15}$ (*13*). A slight twist in the design of a cesium beam standard is to replace the state-selection magnets (magnets A and B in Fig. 2A) with lasers that optically pump the cesium atoms into specific energy states, thereby making state selection and detection more efficient. This provides some improvement, and the best optically pumped cesium beam standards have frequency inaccuracies on the order of $\delta\nu/\nu_o \approx 3 \times 10^{-15}$. This is only slightly more accurate than the best magnetically selected thermal cesium beams, because both versions are limited fundamentally by the large atom velocities and the resulting short interaction time.

A solution to this problem is to use slowly moving cesium atoms in a fountain geometry, as originally conceived by Zacharias in the 1950s (*14*). The idea was simple—to build a cesium beam clock vertically with one Ramsey interaction zone. Slow atoms in the cesium beam would traverse the microwave interaction zone traveling upward, reverse their velocity under the influence of gravity, and traverse the microwave interaction zone a second time traveling downward, resulting in Ramsey's two-pulse interaction scheme. With a ballistic flight traveling only a meter upward, the interaction time approaches 1 s instead of the 10 ms typical of beam clocks. Unfortunately, the scheme could not be realized; collisions between fast and slow cesium atoms in the beam very efficiently removed all the slow atoms Zacharias was counting on for the signal.

This idea was resurrected in the late 1980s when Chu and co-workers made the world's first working atomic fountain (*15*), which used laser cooling (*16*) to produce atoms at microkelvin temperatures. Researchers at the BNM-SYRTE (Bureau National de Metrologie–Systèmes de Référence Temps Espace) later built the first cesium primary frequency standard based on the fountain concept (*17*). Many other researchers in metrology laboratories around the world have built (or are building) laser-cooled cesium fountain primary frequency standards similar to the schematic design shown in Fig. 2B. The resulting interaction time, ~1 s, allows fountain-based frequency standards to achieve much lower inaccuracy than beam standards. The present systematic inaccuracy of the NIST fountain clock (NIST-F1) is $\delta\nu/\nu_o < 4 \times 10^{-16}$, with other fountain frequency standards having similar or only slightly larger inaccuracy (*18–21*).

At this level of performance, one finds that the limitations arise from a variety of interesting physical interactions that are fundamental in nature. For example, given a certain number of cesium atoms, the instability of the clock output is limited by quantum-mechanical measurement statistics, whereas the accuracy of the clock is affected primarily by uncertainty in the ambient blackbody radiation, knowledge of the local gravitational potential, and collisions between the cesium atoms. In the case of NIST-F1, the cesium atoms are bathed in the blackbody radiation from the surrounding room at 300 K. This results in a correctable shift of

$\delta\nu/\nu_o = 2 \times 10^{-14}$, but the uncertainty in the correction is $\delta(\delta\nu/\nu_o) \approx 2.5 \times 10^{-16}$, corresponding to an uncertainty of 1 K in the temperature of the thermal radiation. At a similar level, the gravitational redshift moves the clock frequency by approximately $\delta\nu/\nu_o = 10^{-16}$ per meter change in the elevation of the cesium with respect to sea level. Furthermore, in NIST-F1, collisions between cesium atoms cause a frequency shift with an uncertainty of $\delta\nu/\nu_o \approx 1.5 \times 10^{-16}$. The next generation of cesium fountain clocks are being designed to overcome some of these limitations, with the intention of reducing the total uncertainty in accuracy to the level of $\delta\nu/\nu_o \approx 10^{-16}$. After that, the best cesium clocks might have to operate in orbit around Earth, where the weightless environment would allow interaction times of 5 to 10 s with slow cesium atoms, resulting in projected fractional uncertainties as low as 5×10^{-17} (*22*, *23*).

Optical Frequency Standards and Clocks

Although present-day cesium microwave frequency standards perform at an already remarkable level, a new approach to timekeeping based on optical atomic transitions promises still greater improvements. By using optical ($\nu_0 \sim 10^{15}$ Hz) rather than microwave ($\nu_0 \sim 10^{10}$ Hz) frequencies, such a clock operates with a much smaller unit of time (comparable to using a second rather than a day as the basic unit). One can see from Eq. 1 that basing a standard on a transition in the optical rather than the microwave region of the spectrum could in principle lead to an enormous reduction in instability. Optical standards should be considerably more accurate as well, as several key frequency shifts are fractionally much smaller in the optical domain. Moreover, the investigation of these shifts will be greatly accelerated by the much smaller instability of the optical standards.

These potential advantages were recognized in the early days of frequency standards, but a coherent light source was needed to serve as the local oscillator. Soon after the first demonstrations of lasers in the early 1960s, the search began for relevant transitions on which to stabilize the laser frequencies. Some of the earliest stabilized lasers were based on transitions in the well-known helium-neon laser, whose frequency was locked to absorption lines in methane (in the infrared) or iodine (at 633 nm) molecules (*24*, *25*). With the advent of tunable lasers, frequency standards based on molecules, neutral atoms, and ions now exist throughout the visible spectrum. These standards extend to the near-infrared wavelengths as well, including important references for the densely spaced fiber communications frequency channels in the 1.3- and 1.5-μm region (*26*).

However, optical standards did not truly begin to realize the potential gains expressed in Eq. 1 until the past decade, when several critical technologies reached maturity. First, advances in laser cooling techniques of atoms and ions made it possible to cool a variety of atoms and ions (including those with narrow clock transitions) to millikelvin temperatures and below (*16*). The use of laser-cooled atomic samples enables the extended interaction times (along with reduced Doppler shifts) required to observe a narrow transition linewidth (Δν in Eq. 1). Second, to resolve narrow linewidths, probe lasers need to be spectrally pure. Recent improvements in laser stabilization based on environmentally isolated optical reference cavities have enabled laser linewidths at the subhertz level to be achieved (*27*, *28*). Finally, and perhaps most critically, a simple means for counting optical frequencies and linking them to other frequencies in the microwave and optical domains became available with the development of the mode-locked femtosecond (10^{-15} s) laser frequency comb (*29*, *30*). As a result of these improvements, there is renewed enthusiasm in this field, with many groups now racing to develop new standards and clocks based on various transitions.

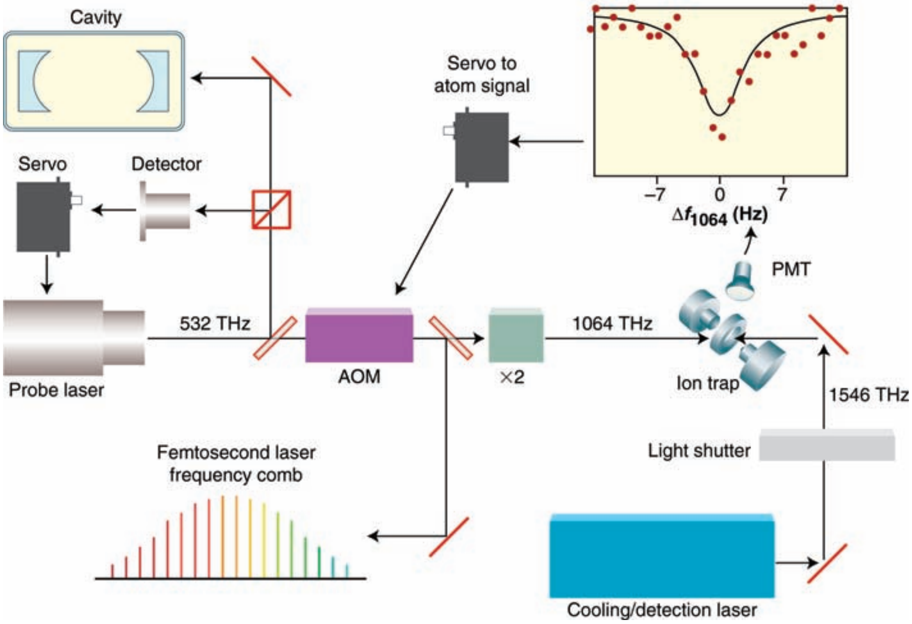

Fig. 3. Main components of an optical atomic clock. The probe laser, whose frequency is prestabilized on an optical cavity, is used to excite transitions in the laser-cooled trapped ion. A servo system uses the signal from the ion to keep the probe laser frequency centered on resonance. Light is sent to the femtosecond-laser frequency comb, which enables counting of the clock cycles.

When developing a new standard, the choice of the atomic reference transition itself is clearly one of the most critical aspects. For state-of-the-art performance, one desires a narrow transition that is extremely insensitive to external perturbations such as frequency shifts caused by magnetic and electric fields. Such transitions (often called clock transitions) exist in both ions and neutral atoms, and the choice currently comes down to working with a few trapped ions or a large number of neutral atoms.

A trapped ion can be laser-cooled to the zero point of its motion, thereby suppressing Doppler effects that can shift the resonance frequency (*31*). Moreover, an ion can be probed while trapped, hence long interaction times can be achieved. Although these factors give trapped-ion standards excellent prospects for high accuracy, their S/N ratio is limited, because in most cases ion-ion interactions become a limiting factor if more than a few ions are in the trap. Nonetheless, outstanding performance has been demonstrated with optical transitions in a variety of single-ion systems, including Hg^+, Yb^+, Sr^+, and In^+ (*32–34*).

Large numbers of neutral atoms can be probed simultaneously, which enables an extremely high S/N ratio and potentially very high stability. Unfortunately, methods of confining neutral atoms, although efficient for collecting atomic samples, can lead to shifts for the reference transition. Thus, the atoms typically need to be released during the probe cycle, which leads to troublesome Doppler-related systematic shifts and limited interaction times. Still, excellent performance has been demonstrated with several laser-cooled neutral-atom standards (*35*). Moreover, a possible solution to the confinement problem has been proposed, which

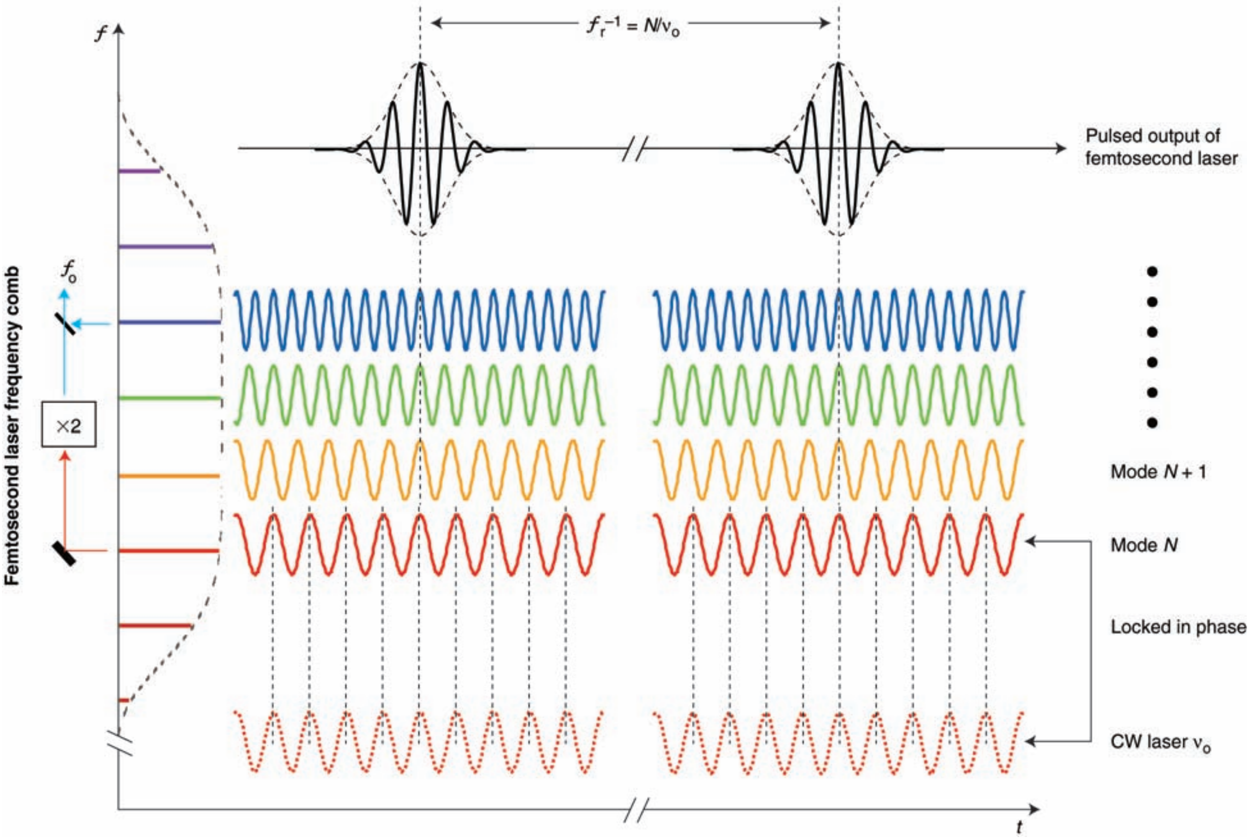

Fig. 4. Illustration of the relationship between the atomically stabilized CW laser with frequency ν_o and the mode-locked femtosecond laser pulses. When the femtosecond laser comb is phase-locked to ν_o, the frequency of the pulse repetition rate is simply a rational fraction of ν_o. For clarity, only a few of the femtosecond laser comb elements are shown here. Actual devices might have ~500,000 modes separated by 1 GHz.

involves loading neutral atoms into a specially designed laser lattice (*36*). The wavelength of the lattice laser beams can be chosen so as to minimize shifts of the clock transition. In this way, one could have the long interaction times and small Doppler shifts associated with ions, along with the good S/N ratio achievable with large numbers of neutral atoms. Laser-cooled neutral-atom systems currently under development for optical clocks include Ca, Sr, Yb, Mg, and H (*32–34*).

The single-ion Hg^+ system, a typical state-of-the-art optical frequency standard, is shown in Fig. 3. A measurement cycle commences with laser cooling of the ion, followed by a probe period during which the ion is excited by a pulse from the probe laser, whose frequency is locked to a narrow resonance line of a high-finesse Fabry-Perot cavity with an electronic feedback loop. An optical cavity can serve as near-ideal short-term reference, providing a comb of resonance lines that can be narrow and resolved with high S/N ratio. As the frequency of a given cavity line depends on the spacing of the mirrors, it is essential to isolate the cavity spacer that determines the mirror separation from environmental perturbations. Thus, the cavity spacer is usually made from a special material and placed in an isolation chamber. Using this approach, lasers with linewidths of 1 Hz or less can be maintained for durations of tens of seconds (*28*). A frequency-shifting device such as an acousto-optic modulator (AOM) then adjusts the laser frequency so that it is near the atomic resonance. Near-resonant probe pulses induce excitation in the atomic sample, which is detected by collecting atomic fluorescence on a photomultiplier tube. So-called "shelving detection" schemes based on a strong transition (usually the laser cooling transition) are usually used to enhance the detected signals, thereby enabling atom shot noise–limited performance (*37*, *38*).

Because we are working with a quantum system, a single measurement on a single atom cannot tell us the frequency of the probe ("clock") laser, but only whether the atom was excited by the probe pulse. For a single ion, it is necessary to average the excitation over many probe cycles to determine the excitation probability, which in turn can be related to the frequency of the laser. Alternatively, one can use a single probe cycle to excite a large number of atoms, effectively performing the averaging simultaneously (as one typically does with neutral atom clock samples). As an example, we show in Fig. 3 an averaged excitation spectrum obtained by scanning the probe laser frequency over the Hg^+ clock resonance near 282 nm (this signal has a line $Q = \nu_o/\Delta\nu = 10^{14}$, the highest demonstrated in the microwave or optical region of the spectrum) (*39*). With appropriate modulation techniques, a spectroscopic signal suitable for locking the laser to the center of this resonance can be generated. This second stage of locking uses feedback to the AOM frequency to keep the laser frequency fixed on the atomic resonance, thereby suppressing residual cavity drifts. With such a lock, the probe laser frequency can now exhibit excellent long-term performance and be used as an optical frequency standard.

Several such standards have been constructed using either single ions or about a million neutral atoms, many of which now achieve inaccuracies of 10^{-14} or better, including Hg^+, Yb^+, Sr^+, Ca, and H. It is anticipated that in the next year a few optical standards will be evaluated at the 10^{-15} level and below. It has been predicted that the systematic effects in single-ion optical frequency standards could be controlled at a level that would permit uncertainties approaching 10^{-18} (*38*). Reaching such a level will necessarily require short-term instability

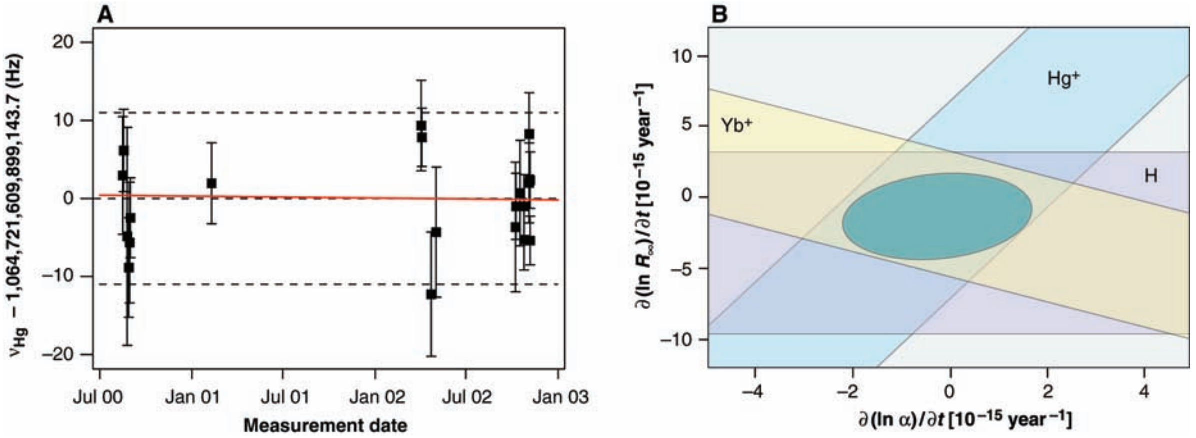

Fig. 5. (**A**) Measurement of the Hg^+ optical clock transition with respect to the Cs ground state hyperfine splitting that defines the SI second. The plot shows the deviation of each measurement from the weighted average value with its statistical error bar and a linear fit to these data (red line). The total systematic uncertainty is represented by the dashed line at ±11 Hz. Using this measurement in combination with other recent comparisons of H and Yb^+ to the Cs ground state hyperfine splitting allows one to constrain possible variations of α and R_∞ as shown in (**B**). The central ellipse represents the region consistent with the combination of all three experiments [see (*56*) for details].

at or below 10^{-15} for 1 s of signal averaging if inordinately long averaging times are to be avoided.

Femtosecond Laser Frequency Divider

To generate an interval of time with an optical standard (and thereby compare it to existing microwave atomic clocks), one requires a reliable and straightforward means of counting the extremely rapid optical oscillations that occur with a period of ~1 fs—a time scale much too fast for any conventional electronic counters. In the past, solutions to this counting problem involved complicated approaches based on harmonic frequency chains (*40*) or interval bisection (*41*). However, the scene changed in 1999 when key experiments by the Hänsch group demonstrated that the frequency comb associated with a femtosecond mode-locked laser could be used for counting optical frequencies (*29*). Indeed, in just a matter of years the new femtosecond comb technology has fully replaced other laboratory technologies that existed for decades.

The basic function of the femtosecond laser in an optical clock is to provide a phase-coherent link between the uncountable optical reference frequency and the more accessible microwave domain. This operation can be understood by considering the underlying frequency comb structure associated with the femtosecond laser (Fig. 4). A femtosecond laser produces short pulses from a superposition of phase-locked cavity modes. In the frequency domain, these modes form a comb of evenly spaced oscillators. The spacing of the modes is given by the repetition rate f_r at which pulses are emitted from the mode-locked laser. Moreover, a difference in group and phase delays inside the femtosecond laser cavity leads to a frequency offset, f_o, of the comb elements from being exact harmonics of f_r. The relationship between these two microwave frequencies (f_o, f_r) and the *n*th element of the optical frequency comb is given by $\nu_n = nf_r + f_o$, where *n* is an integer.

In general, the group and phase delays in the femtosecond laser fluctuate and are not calculable at a level that permits a high-precision determination of f_o. Therefore, f_o must be directly measured, and an elegant means of accomplishing this involves using nonlinear frequency generation to compare different regions of the frequency comb (*42*). For example, if the laser spectrum covers more than one octave, then the comb elements at the low-frequency end of the spectrum can be doubled in a nonlinear crystal and subsequently heterodyned against the high-frequency components of the comb to yield f_o. An important advance in this respect is the generation of octave-spanning spectra with low-power Ti:sapphire lasers in microstructured fibers (*43*, *44*), or by direct generation from the laser itself (*45*–*47*). Once measured, f_o can then be locked at a fixed frequency with the use of servo-control techniques. If, for example, it is set to zero, then each comb element is an exact harmonic of f_r. Phase-locking one element ($n = N$) of the frequency comb to the low-noise continuous-wave (CW) laser that has itself been steered to the atomic resonance leads to the desired result for optical to microwave conversion: $f_r = \nu_o/N$ (Fig. 4). With such a scheme, the complete connection from the optical to microwave domain has now been demonstrated—starting with either a single ion or a few million neutral atoms and ending with the 1 GHz optical pulse train from a femtosecond laser (*48*). The excess noise of this division process has been verified at a level sufficiently low to support the best current optical frequency standards (*49*, *50*).

Outlook

Today's cesium atomic clocks have timing uncertainty at the level of ~35 ps per day, and optical atomic clocks might someday have uncertainties near 100 fs per day. When most people would get by just fine with a clock that is accurate to a few seconds per day, it is worth asking why scientists, or society for that matter, might want still better clocks. In the 1950s, when microwave atomic clocks based on cesium were first developed, the situation was in some ways similar to where we find ourselves today. Those first atomic clocks were quickly recognized as a great improvement over the existing clocks, yet at the same time they were mainly a tool of scientific interest. At that point, few people would imagine that just 40 years into the future a constellation of satellites containing cesium (and rubidium) atomic clocks would circumnavigate the globe and provide accurate time and position to all people below. The GPS and its constituent atomic clocks are now an integral part of our lives. Similarly, today's voice and data communications systems that are synchronized with atomic clocks would likely have been viewed as science fiction in 1955. In the next 50 years, there is every reason to expect that improved microwave clocks and optical clocks will find numerous applications in communications and navigation—the two areas that have throughout history advanced in parallel with improving time standards. The very stable optical clock ticks may be especially useful for tracking and communication between satellites and spacecraft in the much greater expanses beyond our planet. For example, an optical clock with femtosecond instability would provide potential ranging uncertainty at the micrometer level over millions of kilometers.

For the immediate future, it is already clear that the most advanced clocks will provide interesting new scientific avenues to study our universe, pushing the limits on tests of the most fundamental physical laws to new levels. This includes tests of general relativity and searches for violations of the isotropy of space or a preferred reference frame. Fundamental symmetries between matter and antimatter could be investigated through the comparison of optical clocks, as has been proposed for the 1*s*-2*s* transitions in both hydrogen and anti-hydrogen (*51*). To date, optical and microwave frequency standards have already been used in some of the most accurate determinations of the fine structure constant α and the Rydberg constant R_∞ [e.g., (*52*, *53*)], and laboratory comparisons of clocks based on different atomic transitions are now providing some of the most stringent constraints of the possible variation of fundamental constants (*54*, *55*). An example of this kind of experiment is given in Fig. 5A, which shows the measurement of the Hg$^+$ optical clock transition at $\sim$1064 THz (282 nm) in terms of the cesium hyperfine splitting as realized by NIST-F1 (*54*). Over a period of $\sim$3 years, there is no measurable divergence in the ratio of the output frequencies of these two clocks, constraining a fractional variation of $g_{\mathrm{Cs}}(m_{\mathrm{e}}/m_{\mathrm{p}})\alpha^6$ to be less than 7×10^{-15} per year, where $m_{\mathrm{e}}/m_{\mathrm{p}}$ is the electron-to-proton mass ratio and g_{Cs} is the ^{133}Cs nuclear g factor. Assuming any variation comes only from the α^6 factor, the data constrain any possible linear fractional variation of α to be less than 1.2×10^{-15} per year. The combination of these data with other recent clock comparison experiments has resulted in similar constraints being placed on other fundamental constants, as summarized in Fig. 5B (*56*, *57*).

It seems clear that future atomic clocks will continue to subdivide the second into still smaller units of time. But in contrast to Mermin's original concern, it is more likely that the femtosecond, attosecond (10^{-18} s), or zeptosecond (10^{-21} s) will be considered utterly alien impositions on our macroscopic world, while nonetheless proving to be useful bookkeeping units in the continued quest to better understand the inner workings of the microscopic world.

References and Notes

1. N. D. Mermin, *Phys. Today* **54**, 11 (2001).
2. J. H. Taylor, in *Nobel Lectures, Physics 1991–1995*, G. Ekspong, Ed. (World Scientific, Singapore, 1997), pp. 73–91.
3. R. F. C. Vessot *et al.*, *Phys. Rev. Lett.* **45**, 2081 (1980).
4. R. E. Beehler, *Proc. IEEE* **55**, 792 (1967).
5. N. F. Ramsey, *J. Res. Nat. Bur. Stand.* **88**, 301 (1983).
6. C. Audion, B. Guinot, *The Measurement of Time: Time, Frequency and the Atomic Clock* (Cambridge Univ. Press, Cambridge, 2001).
7. H. Lyons, *NBS Tech. News Bull.* **33**, 17 (1949).
8. L. Essen, J. V. L. Parry, *Nature* **176**, 280 (1955).
9. E. C. Bullard, *Nature* **176**, 282 (1955).
10. Resolution 1, 13^{e} Conférence Générale des Poids et Mesures, *Metrologia* **4**, 41 (1968).
11. N. F. Ramsey, *Phys. Today* **33**, 25 (1980).
12. T. J. Quinn, *Proc. IEEE* **79**, 894 (1991).
13. A. Bauch, B. Fischer, T. Heindorff, R. Schröder, *Metrologia* **35**, 829 (1998).
14. N. F. Ramsey, *Molecular Beams* (Clarendon, Oxford, 1956).
15. M. Kasevich, E. Riis, S. Chu, R. DeVoe, *Phys. Rev. Lett.* **63**, 612 (1989).
16. H. J. Metcalf, P. van der Straten, *Laser-Cooling and Trapping* (Springer, New York, 1999).
17. A. Clairon *et al.*, in *Proceedings of the 5th Symposium on Frequency Standards and Metrology,* J .C. Berguist, Ed. (World Scientific, London, 1996), pp. 49–59.
18. T. P. Heavner, S. R. Jefferts, E. A. Donley, J. H. Shirley, T. E. Parker, *IEEE Trans. Instrum. Meas.*, in press.
19. S. Weyers, U. Hübner, R. Schröder, Chr. Tamm, A. Bauch, *Metrologia* **38**, 343 (2001).
20. S. Bize *et al.*, in (*32*), pp. 53–63.
21. F. Levi, L. Lorini, D. Calonico, A. Godone, *IEEE Trans. Ultrason. Ferroelect. Freq. Control* **51**, 1216 (2004).
22. P. Laurent *et al.*, in (*32*), pp. 241–252.
23. T. P. Heavner *et al.*, in (*32*), pp. 253–260.
24. J. L. Hall, C. J. Borde, K. Uehara, *Phys. Rev. Lett.* **37**, 1339 (1976).
25. J.-M. Chartier, A. Chartier, *Proc. SPIE* **4269**, 123 (2001).
26. S. L. Gilbert, W. C. Swann, T. Dennis, *Proc. SPIE* **4269**, 184 (2001).
27. J. L. Hall, *Science* **202**, 147 (1978).
28. B. C. Young, F. C. Cruz, W. M. Itano, J. C. Bergquist, *Phys. Rev. Lett.* **82**, 3799 (1999).
29. Th. Udem *et al.*, *Phys. Rev. Lett.* **82**, 3568 (1999).
30. S. A. Diddams *et al.*, *Phys. Rev. Lett.* **84**, 5102 (2000).
31. F. Diedrich, J. C. Bergquist, W. M. Itano, D. J. Wineland, *Phys. Rev. Lett.* **62**, 403 (1989).
32. P. Gill, Ed., *Proceedings of the 6th Symposium on Frequency Standards and Metrology* (World Scientific, Singapore, 2002).
33. *Proceedings of the Conference on Precision Electromagnetic Measurements* (IEEE, Piscataway, NJ, 2004).
34. J. Ye, H. Schnatz, L. W. Hollberg, *IEEE J. Select. Top. Quantum Electron.* **9**, 1041 (2003).
35. U. Sterr *et al.*, *C. R. Phys.*, in press.
36. H. Katori, M. Takamoto, V. G. Pal'chikov, V. D. Ovsiannikov, *Phys. Rev. Lett.* **91**, 173005 (2003).
37. D. J. Wineland, J. C. Bergquist, W. M. Itano, R. E. Drullinger, *Opt. Lett.* **5**, 245 (1980).
38. H. G. Dehmelt, *IEEE Trans. Instrum. Meas.* **31**, 83 (1982).
39. R. Rafac *et al.*, *Phys. Rev. Lett.* **85**, 2462 (2000).
40. D. A. Jennings, K. M. Evenson, D. J. E. Knight, *Proc. IEEE* **74**, 168 (1986).
41. H. R. Telle, D. Meschede, T. W. Hänsch, *Opt. Lett.* **15**, 532 (1990).
42. H. R. Telle *et al.*, *Appl. Phys. B* **69**, 327 (1999).
43. J. K. Ranka, R. S. Windeler, A. J. Stentz, *Opt. Lett.* **25**, 25 (2000).
44. D. J. Jones *et al.*, *Science* **288**, 635 (2000).
45. U. Morgner *et al.*, *Phys. Rev. Lett.* **86**, 5462 (2001).
46. A. Bartels, H. Kurz, *Opt. Lett.* **27**, 1839 (2002).
47. T. Fortier, D. J. Jones, S. T. Cundiff, *Opt. Lett.* **28**, 2198 (2003).
48. S. A. Diddams *et al.*, *Science* **293**, 825 (2001).
49. L.-S. Ma *et al.*, *Science* **303**, 1843 (2004).
50. A. Bartels *et al.*, in *OSA Trends in Optics and Photonics Series (TOPS) Vol. 96, Conference on Lasers and Electro-Optics (CLEO)* (Optical Society of America, Washington, DC, 2004), paper CPDC10.
51. J. Walz *et al.*, *Appl. Phys. B* **77**, 713 (2003).
52. F. Biraben *et al.*, in *The Hydrogen Atom: Precision Physics of Simple Atomic Systems*, S. V. Karshenboim, F. S. Pavone, F. Bassani, M. Inguscio, T. W. Hänsch, Eds. (Springer-Verlag, Berlin, 2001), pp. 17–41.
53. Th. Udem *et al.*, in *The Hydrogen Atom: Precision Physics of Simple Atomic Systems*, S. V. Karshenboim, F. S. Pavone, F. Bassani, M. Inguscio, T. W. Hänsch, Eds. (Springer-Verlag, Berlin, 2001), pp. 125–144.
54. S. Bize *et al.*, *Phys. Rev. Lett.* **90**, 150802 (2003).
55. H. Marion *et al.*, *Phys. Rev. Lett.* **90**, 150801 (2003).
56. E. Peik *et al.*, *Phys. Rev. Lett.* **93**, 170801 (2004).
57. M. Fischer *et al.*, *Phys. Rev. Lett.* **92**, 230802 (2004).

58. We thank L. Hollberg, D. Wineland, and M. Lombardi for their assistance and thoughtful comments, and E. Peik and his colleagues at the PTB for providing Fig. 5B.

REVIEW

The Route to Atomic and Quantum Standards

Jeff Flowers

Over the past half-century, there has been a shift away from standards based on particular artifacts toward those based on physical effects, the most stable being based on quantum properties of systems. This change was proposed at the end of the 19th century but is still not complete at the start of the 21st. We discuss how this vision has been implemented through recent advances in science and metrology and how these may soon lead to an SI system finally free from artifact standards, with a consistency based on fundamental constants.

Quantities, Units, and Standards

To investigate any physical phenomena, we must make measurements, communicate them to others, and record them in a way that will be understandable in the future. To do so, a system of quantities and units is required. Measurement is a comparison process in which the value of a quantity is expressed as the product of a value and a unit; that is,

$$\text{Quantity} = \{\text{numerical value}\} \times [\text{unit}] \quad (1)$$

where the unit is an agreed-upon value of a quantity of the same type. The concept of a quantity such as length is independent of the associated unit; the length is the same whether it is measured in feet or meters. A standard is a

National Physical Laboratory, Teddington, Middlesex TW11 0LW, UK. E-mail: jeff.flowers@npl.co.uk