Cesium Primary Frequency References

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Abstract
Primary frequency standards with stated inaccuracies of $\delta f/f \approx 10^{-15}$ or slightly better are in use today in several national timing laboratories. These standards, which are the most accurate in the world today, use laser-cooled cesium atoms to obtain this level of performance. We discuss the operation of these standards as well as possible future improvements that should see the inaccuracy of the realization of the second fall to the $10^{-16}$ Plevel over the next 5 to 10 years.

1. Introduction

The second is one of seven base units of the international system (SI) and is used to define of some of the other base units. The meter is, in fact, defined with respect to the second along with the speed of light, and the volt is maintained by the Josephson effect in terms of a constant times a frequency (and the frequency is, of course, defined in terms of the second). Because frequency can be measured easily with very low uncertainty, many physical quantities are measured with transducers that convert the measurement into a frequency measurement, as with the Josephson effect voltage standard above. The development and operation of high-quality frequency standards is therefore a priority at many national metrology laboratories around the world.

Since 1967, the SI second has been defined as being 9,192,631,770 cycles of the ground-state hyperfine splitting of the cesium atom, where the atom is taken to be unperturbed, at rest, on the reference geoid of the Earth [1,2]. This definition is essentially impossible to realize in a laboratory, as it requires that the atom be in zero magnetic field; further, the environment seen by the atom would have to be at a temperature of absolute zero and the atom would be required to have no residual velocity. Primary frequency standards, such as those operated at national metrological laboratories, attempt to measure the effect of residual magnetic fields: cesium atoms not at absolute zero, etc., on the output frequency of the standard. These measurements are used to “correct” the output frequency to mimic the definition of the second. How, and how well this is done is the subject of this paper.

In the late 1940’s and early 1950’s work began in several national laboratories, notably the National Physical Laboratory (NPL) in England and the National Bureau of Standards (NBS; now the National Institute of Standards and Technology, NIST) in the United States to build frequency references based on atomic transitions [3,4]. Prior to the introduction of atomic clocks, quartz oscillators calibrated to the orbital motion of the Earth about the Sun were the best frequency standards available, with fractional frequency uncertainties of about $2 \times 10^{-8}$ (throughout this paper the quoted frequency uncertainty is given as $\delta f/f_p$, where $\delta f$ is generally the one-sigma uncertainty of the measurement of the accuracy of the clock frequency, $f_p$). This relative frequency uncertainty allows comparison between
different frequency standards (e.g., those based on quartz or cesium). The first generation of atomic standards quickly surpassed the quartz standards of the day, and since then have continued to improve at the rate of about a factor of 10 every 10 years. Fig. 1 illustrates this trend for the primary frequency standards at NBS/NIST, and this curve is representative of the development primary frequency standards over time worldwide.

Primary atomic frequency standards work by frequency-locking an external oscillator to a particular atomic resonance. Generally speaking, the narrower the resonance, the more stable and accurate the clock can be made. The width of the resonance, $\Delta \nu$, is set by the period, $\tau_d$, the atoms spend interacting with the external oscillator. The longer this period, the narrower the measured resonance. The fractional width of the resonance is $\frac{\Delta f_o}{f_o} \propto \frac{1}{\tau_d f_o}$. It can be seen that the optimum performance is obtained by making the interaction period, $\tau_d$, as long as possible and the frequency, $\nu_o$, of the atomic resonance as high as possible. These simple considerations led to cesium being adopted for the international definition of the second; cesium is a relatively heavy atom (133 amu) and as a result its mean thermal velocity is fairly small (~130 m/s at room temperature). This relatively low velocity allows the cesium to stay in the interaction region for longer periods than for example hydrogen, which has a mean thermal velocity of about 1600 m/s at room temperature. Cesium also has one of the highest hyperfine frequencies of any atom, at 9.2 GHz, compared to 6.8 GHz for rubidium and 1.4 GHz for hydrogen.

Most of the basic concepts of modern atomic frequency standards were developed by Rabi and his co-workers at Harvard in the 1930's and 40's [5]. In the early work of Rabi the atomic transition (resonance) was interrogated with one long (microwave) interrogation pulse. This provided the needed long interaction time, but led, for various reasons, to the output frequency of the standard being subject to Doppler shifts, and being critically dependent on the amplitude of the microwaves as well as the uniformity of a small DC magnetic field in the interaction region (for historical reasons, this field is known as the c-field). Ramsey's method of separated oscillatory fields 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

![Fig. 1. The inaccuracy of typical primary frequency standards based on the cesium atom hyperfine transition. The slope of the line represents an improvement of about a factor of 10 in accuracy per decade.](image-url)
short pulses at the beginning and end of the interaction zone. This two pulse process (now universally known as Ramsey interrogation) reduces the sensitivity to microwave power fluctuations and magnetic field inhomogeneity (by factors of 10 to 100 or more) and essentially eliminates the Doppler effect [6].

Cesium is a complicated atom with $F = 3$ and $F = 4$ ground states. Associated with the $F$ quantum number is another quantum number, $m_F$, which is limited to $2F+1$ integer values between $-F$ and $+F$. There are therefore $7+9=16$ possible ground states for the cesium atoms in the atom beam (labeled $|F,m_F\rangle$), and all of these states are essentially equally populated in the atomic beam from the oven. Unfortunately, only the $m_F = 0$ state is useful in a primary frequency standard, because the other states are sensitive in first order to magnetic fields (Zeeman effect), while the transition between the $|4,0\rangle$ and $|3,0\rangle$ states used by the clock is insensitive to magnetic fields (to first order).

2. Cesium Beam Frequency Standards

2.1 Magnetic State-Selected Standards

Fig.2 shows a schematic diagram of a conventional magnetically state-selected cesium beam standard. The design can be directly traced back to Rabi and Ramsey’s seminal work, and essentially all commercial cesium atomic clocks available today (2006) operate using this general design, as did all primary frequency standards until quite recently.

On the left of Fig. 2 is a cesium oven from which a beam of cesium atoms emerges when the oven is heated. This beam of cesium atoms is collimated and directed through a Stern-Gerlach magnet (the a-magnet in Fig. 2), that deflects and focuses those cesium atoms in the beam that are in the desired state through a hole in the magnetic shield, so the atoms are now traveling from left to right in the Figure. Atoms that are not in the desired state are defocused and absorbed by the getter shown in the Figure. The net result of this is that the a-magnet selects only the $|4,0\rangle$ state and “throws away” most (15/16) of the flux of cesium atoms from the oven. The remaining atomic beam next enters the first microwave interaction region (cavity) and continues through the vacuum chamber until it reaches the second interaction region. The atoms then leave the magnetically shielded region at the right edge in the figure and pass through another Stern-Gerlach magnet (b-magnet). Atoms that have changed state, from $|4,0\rangle$ to $|3,0\rangle$, as a result of the microwave interaction, are directed to the hot-wire ionizer, and the resulting ions are collected in the ion-collector. Atoms that have not changed their atomic state are directed to the getter. The current induced in the
hot-wire ionizer and ion collector is proportional to the flux of atoms hitting the hot wire. Maximizing this current, all other things being equal, maximizes the number of atoms making the transition and thus assures that the frequency of the microwaves is on resonance.

There are several challenging aspects of this design. Loss of the majority of the atoms compromises the potential signal to noise (S/N) ratio. The Stern-Gerlach magnets have large magnetic fields associated with them that can cause frequency shifts. Finally the current at the hot wire and ion collector is quite small, further complicating the S/N situation. Nonetheless this design provided the world with all of its primary frequency standards up until about 1990, and several national metrological laboratories continue to operate such clocks with great success. The C S - 1 and C S - 2 clocks at the Physikalisch-Technische Bundesanstalt (PTB) in Germany use this basic design and are currently contributing to international atomic time. C S - 1 has the lowest stated frequency inaccuracy of any clock of this type ever built with a claimed $\delta f/f \approx 5.0 \times 10^{-15}$ [7].

2.2 Optically State-Selected Standards

As early as 1950, Kastler suggested the use of optical pumping to replace the state-selection magnet (a-magnet) in thermal beam standards [8]. It was not until the development of reliable room-temperature laser diodes in the 1980’s that the idea was seriously pursued as practical by the national metrology laboratories. Optical pumping is a method that uses optical transitions of the atom to try to "pump" all of the atoms into a desired state. Simple optical pumping schemes in cesium will pump all atoms into the $|3, 0\rangle$ state by a factor of $16/7$, a little more than 2. This is accomplished by exciting the cesium atoms in the F=4 ground state to a F'=3 excited state with a laser tuned to the F=4$\rightarrow$F'=3 transition. Atoms in the F=3 optically excited state decay back to the ground state in about 30 ns with roughly equal probability of decaying to F'=3 and F=4 (the ' symbol in F' is used to denote an optically excited state). Atoms that decay to F=3 do not interact with the laser and are therefore quiescent, whereas atoms decaying into F=4 are simply re-excited until they decay to F=3: eventually all of the atoms are in the F=3 state. More complicated optical-pumping schemes involving two lasers can pump all of the atoms into the $|3, 0\rangle$ state with a consequent increase in signal by a factor of 16.

The same sort of scheme can be used to eliminate the b-magnet from the design in Fig. 2 as well. If the atoms emerging from the cavity are illuminated with laser light tuned to the F=4$\rightarrow$F'=5 transition, then the atoms in F=4, that is, atoms that have made the transition from the F=3 state, will scatter photons from the laser beam into a photodiode (or photomultiplier) detector. Maximizing the detected light from the atomic beam locks the microwave frequency to the atomic transition. The system can be optimized so that each atom emits several detected photons, increasing the S/N ratio substantially.

Several national metrology laboratories have built and operated optically pumped thermal beam standards: NIST, the Laboratoire Primaire du Temps et Fréquences (LPTF) in Paris, the Communications Research Laboratory (formerly CRL now NICT) in Japan, and the Korea Research Institute of Standards and Science (KRISS) in S. Korea [9 - 12]. The best of these standards have frequency inaccuracies in the range of $3 \times 10^{-15} \leq \delta f/f \leq 5.0 \times 10^{-15}$. It can be seen that the best optically pumped thermal beams are only slightly more accurate than the best magnetically selected thermal beams. In some sense this is not surprising, as both types of standard use essentially identical cesium beams with atom velocities around 100 m/s. This large velocity fundamentally limits the drift-time of the atom in the interaction region to less than 10 ms. Attempts to lengthen the interaction region (and hence the interaction time) beyond a meter or so have not been notably successful. As the interaction region is extended the slower atoms in the atomic beam begin to "fall out of the bottom" of the beam under the influence of gravity. The beam also spreads in the direction transverse to the flight direction, causing more of the atoms to be lost to various apertures in the system. NBS-6 had an interaction region (Ramsey cavity structure) that was 3.74 m long and was not notably superior to its contemporaries with much shorter interaction regions [13].

The thermal beam standards all finally reach uncertainty levels set by the limited interaction time caused by the large atomic velocity. A longer interaction time seems necessary to decrease the
inaccuracy to the level of better \( \delta f/f \approx 1.0 \times 10^{-15} \) better.

3. Cesium Fountain Frequency Standards

The original concept of a cesium fountain was introduced in the 1950's by Jerrold Zacharias [6]. Zacharias' idea was simply to build a cesium beam clock vertically with one Ramsey interaction zone. Slow atoms from the cesium oven would traverse the microwave interaction zone while traveling upward, reverse their velocity under the influence of gravity, and traverse the same microwave interaction zone while traveling downward. The two interactions with the microwaves reproduced Ramsey's two-pulse interaction scheme, and a ballistic flight traveling only a meter upwards would give interaction times approaching one second instead of the 10 ms typical of thermal beam clocks up to that time. Unfortunately, Zacharias' idea was premature; for obscure technical reasons involving collisions between cesium atoms in a thermal beam (ideas that Zacharias was, eventually, the first to understand!) no signal was ever seen in Zacharias' clock.

Zacharias' idea was resurrected in the late 1980's by Steven Chu and coworkers at Stanford, who made the world's first working cesium fountain using a technique known as laser-cooling which was made possible by advances in laser technology [14]. Researchers at the LPTF (now BNM-SYRTE) in Paris later built the first primary frequency standard based on Zacharias' fountain concept [15]. Many other researchers in metrology laboratories around the world have built (or are building) laser-cooled cesium fountain clocks. Currently fountain clocks at NIST, PTB (Germany), BNM-SYRTE, Istituto Nazionale di Ricerca Metrologica (formerly IEN, now INRM) in Turin, Italy, the National Physical Laboratory (NPL) in England and NMIJ (Japan) contribute to international atomic time using cesium fountain clocks.

Laser-cooling of atoms, the key to making a fountain clock work successfully, is only briefly explained here—there are numerous good references on the subject [16]. A schematic of a cesium-fountain clock is shown in Fig. 3. Atoms from a background gas of room-temperature cesium vapor are cooled at the intersection of the six laser beams to temperatures around 1 µK. Laser-cooling can be explained as essentially a refrigerator; a "sink" at very low entropy (the laser beam) interacts with a sample with much higher entropy (the atomic sample). Entropy is transferred from the atoms to the light field, via optical interactions between the atom and the light field. The entropy of the light field is raised (the atom scatters many photons out of the laser beam with random direction and phase), while the entropy of the atomic sample is lowered substantially. In the case of the scheme used in cesium fountains, known as optical molasses, the room temperature cesium atoms can be cooled to a temperature of a few microkelvins (or below) in a few hundred milliseconds. At these temperatures the thermal...
velocity of the cesium atom is around 10 mm/s as opposed to the 100 m/s velocity at room temperature. This has the result that a spherical sample of cesium atoms gathered in the optical molasses with a diameter of about 1 cm roughly doubles its diameter in 1 second. As an alternative to the thermodynamic view presented above, laser-cooling can be viewed as mechanical effect of the light on the atom. If the laser is tuned slightly lower in frequency than the optical resonance, the atom will, as a result of the Doppler effect, preferentially absorb photons from the laser beam it is moving towards. Each photon the atom absorbs carries momentum $\vec{p} = \frac{h \omega}{c} \hat{k}$ which is directed in opposition to the atomic motion. The atom reemits this photon in a random direction and re-emits slightly more energy than it absorbed (the atom re-emits at the resonance frequency). This cycle of absorbing a photon a slightly lower energy than the reemitted photon in a random direction and is directed in opposition to the atomic motion. The optical resonance, the atom will, as a result of the Doppler effect, preferentially absorb photons from the laser beam it is moving towards. Each photon the atom absorbs carries momentum $\vec{p} = \frac{h \omega}{c} \hat{k}$ which is directed in opposition to the atomic motion. The atom reemits this photon in a random direction and because the laser is tuned below resonance, the atom re-emits slightly more energy than it absorbed (the atom re-emits at the resonance frequency). This cycle of absorbing a photon a slightly lower energy than the reemitted photon is repeated many ($\approx 10^7$) times/second and provides the basic laser cooling cycle.

The basic operation of the cesium fountain proceeds in a sequence of steps: first a sample of ~10^4 cesium atoms is laser-cooled at the intersection of the six laser beams shown in Fig. 3. These atoms are next "launched" upwards at approximately 4m/s by detuning the frequency of the up and down laser beams to make a moving optical molasses. The laser light is then extinguished by shutters so that no laser light interacts with the cesium atoms along their ballistic flight path. The cloud of launched cesium atoms, about 1 cm in diameter, is typically in the F=4 ground state, but all mF=0 levels are populated. The "ball" of cesium atoms is next state-selected with a short microwave pulse that drives the $|4,0\rangle$ atoms into $|3,0\rangle$ and leaves the other F=4 atoms unperturbed. The remaining F=4 atoms are removed from the cloud with a short optical blast. At this point the remaining cesium atoms, all in the $|3,0\rangle$ state, enter the microwave cavity shown in Fig. 3 with a velocity of around 3 m/s. 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 about 1 m above the microwave cavity and begin to fall. The second passage through the cavity, in the opposite direction this time, occurs about 1 second after the first. The atoms are detected optically with a laser tuned to the F=4→F'=5 optical transition, similar to the detection process in an optically pumped and detected thermal beam.

The fountain arrangement results in a line-width, $\Delta \nu$, of order 1 Hz (comparable to the ~100 Hz line-width of thermal beam standards). Most of the frequency uncertainties that limit the inaccuracy of the thermal beam standards scale with the inverse of the interaction time, allowing fountain-based frequency standards to achieve much lower inaccuracy than the older thermal beams. For example, the present systematic inaccuracy of NIST-F1 is $\delta f/f = 4.0 \times 10^{-16}$ with other fountain frequency standards having similar or only somewhat larger inaccuracies [17 - 22].

In NIST-F1, the frequency inaccuracy is limited by two distinct effects, a density shift and a blackbody shift [17,21]. The blackbody shift is simply the result of the cesium atoms interacting with the thermal radiation emitted by the walls of the 300 K vacuum enclosure. The magnitude of this shift, $\delta f/f = 2.1 \times 10^{-14}$ is quite large: however, in the present generation of cesium fountains its uncertainty can be made as small as $\delta f/f = 2.6 \times 10^{-16}$. This represents a limit that will require either a great deal of theoretical calculation (to better understand the shift) along with difficult experimental determination of parameters for the improved theory, or a cryogenic vacuum system to reduce the magnitude of the effect. We are presently pursuing the second possibility at NIST. The density shift is caused by collisions between the cesium atoms in the launched sample and is quite large, as much as $\delta f/f = 4.0 \times 10^{-16}$ in NIST-F1, with an uncertainty in the correction of $\delta f/f = 4.0 \times 10^{-16}$. There are many proposals for lowering the uncertainties with this shift, but it remains a serious problem in the present generation of cesium fountains.

4. Second Generation Cesium Fountains

As mentioned previously, both the blackbody and cesium collision shifts are major sources of uncertainty in present cesium fountains. In the case of NIST-F1, these two effects have a magnitude of $\delta f/f = 2.6 \times 10^{-16}$ for the blackbody frequency bias and $\delta f/f = 1.6 \times 10^{-16}$ for the collision shift. We are presently assembling NIST-F2 which is designed to minimize the frequency uncertainty associated with these two frequency biases.

The blackbody frequency shift has been the
source of some controversy of late with different
groups calculating different frequency biases for the
shift [23-27]. Even if one of these two competing
results is shown to be correct, the present state of the
art precludes an uncertainty in this bias of better than
about $\delta f/f = 1.0 \times 10^{-16}$. In NIST-F2 we have chosen to
avoid this difficulty by building a cryogenic
($T = 77$ K) vacuum structure that includes the
microwave cavities and flight tube above them. This
way the frequency bias associated with the blackbody
shift becomes $\delta f/f = 7.6 \times 10^{-17}$, and the uncertainty of
this bias is easily 10 times less, removing the
blackbody frequency shift as a major source of
uncertainty.

The collisional frequency shift in NIST-F2 will
be significantly reduced using a clever idea
developed at INRM in Torino, Italy and
experimentally demonstrated at NIST [28-29].

Multiple balls of laser-cooled cesium atoms (as many
as 10 in NIST-F2) are launched in quick succession
(as shown in Fig. 4) with the first launched ball of
cesium atoms having the highest apogee while the
second ball has an apogee just below the first, etc. The
trajectories of all the balls intersect in the detection
region. In this way the average cesium density is
reduced by about a factor of 10, but the signal to noise
ratio is preserved as the same number of atoms is
finally detected. We anticipate that we will most likely
launch a greater total number of atoms in NIST-F2 in
order to achieve a short term stability of $2-3 \times 10^{-13} \tau^{-1/2}$
at low densities while achieving an uncertainty of
$\delta f/f = 3.0 \times 10^{-17}$ for the collision shift.

We therefore expect to achieve a total systematic
fractional frequency uncertainty in NIST-F2 of
$\delta f/f < 1.0 \times 10^{-16}$ dominated by effects due to the
microwave frequency of the clock. Smaller frequency

Fig. 4. The multiple ball toss scheme. The horizontal axis in the diagram is the time axis, while the vertical
axis is the height above the launch region. As described in the text, the first launched ball goes the
highest, while the next ball has an apogee slightly less, and so forth. Finally after a free flight for the
first ball of about 1.2 s all of the atoms are in the detection region, where they are detected as a single
ball.
uncertainties will probably only be achieved by the use of the quickly developing optical frequency standards.

5. Conclusions

The history of atomic clocks from the late 1940's to the present era shows a steady improvement in the fractional inaccuracy of the clocks from the \( \delta f/f \approx 1 \times 10^{-10} \) level in 1950 to the \( \delta f/f \leq 1 \times 10^{-15} \) level in 2000. The present state of the art in atomic clocks is defined by the accuracy of the cesium fountains with fractional frequency uncertainties of \( \delta f/f \leq 1 \times 10^{-15} \) today and improvements likely to the \( \delta f/f \approx 2 \times 10^{-16} \) level before 2010.

Atomic clocks, currently based on atomic microwave transitions will, likely, be replaced eventually by atomic clocks based on optical transitions. Atomic clocks using optical transitions with the resulting much higher frequency (~10^{15} Hz) have the potential to reach inaccuracies of \( \delta f/f \leq 1 \times 10^{-17} \) and beyond. These optically based clocks are being investigated in many laboratories worldwide [30, 31], but are some years away from replacing cesium microwave clocks as the world’s time and frequency standards.

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References

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