Status Report on Primary Frequency Standards
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Abstract

This report surveys the state-of-the-art in atomic frequency standards with exclusive regard to their use as primary frequency reference; i.e., only accuracy is discussed. The report covers operational standards as well as devices which are still in the research or exploratory development phase. It is predicted that accuracies of better than $1 \times 10^{-11}$ will be achieved within a few years, and that, as a consequence of new techniques, accuracy may be treated statistically in the not too distant future. Also, clocks may become available which remain accurate continually.

Key words: Atomic frequency standards; Accuracy; Cesium beam tubes; Clocks; Hydrogen masers; Lasers; Primary frequency standards.
INTRODUCTION

Since the advent of quantum electronic frequency standards in 1948 [1], research has been done on a great variety of different devices, techniques, and methods. The majority of existing standards is based on the cesium atom. Cesium hyperfine resonance standards or clock devices were developed in the early 1950's, and the first routinely used clock was built in 1955 by the National Physical Laboratory of the United Kingdom [2]. The subsequent refinement of these devices in many laboratories in the world and the increasing usage of cesium standards for the generation of time scales led in 1967 to the international agreement at the General Conference on Weights and Measures to adopt the cesium resonance frequency for the definition of the second. As a result of this agreement, the Universal Coordinated Time (UTC) has been based since 1971 [3] on these atomic seconds. Time scales which refer to the rotation of the earth are approximated by inserting or leaving out seconds at certain specified dates during the year, as necessary.

Other devices which have been and are being used as primary frequency references include the hydrogen maser oscillator. This device has reached some technical perfection, and is being used by several laboratories not only as a primary frequency reference but also as a clock for time scale generation. Other promising devices which have been investigated, or are presently under investigation, include beam tubes based on the thallium atom, the barium oxide molecule, and the magnesium atom. Also studied are ion storage devices using mercury or barium, as well as devices in the infrared and visible region of the electromagnetic spectrum where lasers are locked to resonances in molecules such as methane, iodine, sulphur tetrafluoride and others.

The principle laboratories currently engaged in research, development and maintenance of primary frequency standards are the National Physical Laboratory of the United Kingdom, the Physikalisch-Technische Bundesanstalt in West Germany, the Laboratoire de l'Horloge Atomique in France, the National Research Council in Canada, the National Research Laboratory for Metrology, and the Radio Research Laboratory in Japan, the Istituto Elettrotecnico Nazionale in Italy, the Australian National Standards Laboratory, and the National Bureau of Standards in the United States. In addition to the work at these laboratories, a significant amount of work has been done in other government agencies of the various countries and at universities.
Present Status of Primary Frequency Standards

I. Cesium

At the present time, there are four operating primary laboratory type cesium beam frequency standards located at the PTB in Germany [4], NRC in Canada [5], and NBS in the United States [6]. These standards have been evaluated with respect to most parameters affecting their output frequency; i.e., experiments and theoretical studies have been performed which yield knowledge about the biases which cause the output frequency to differ from the unperturbed atomic resonance frequency. The accuracy which is then ascribed to the standards is a statistical combination of the uncertainties associated with biases. These four frequency standards have been built independently; they have considerably different designs, and the methods of evaluation are quite different although the same parameters are being evaluated. Most recently, a comparison has been made between the corrected output frequencies of these three primary standards using the International Atomic Time (TAI) Scale as a common reference: the corrected frequencies of the three standards agree to within a peak-to-peak variation of 1 part in $10^{13}$. Each of the laboratories has preliminarily and independently claimed an evaluated accuracy between 1 and 2 parts in $10^{13}$. The frequency is about 1 part in $10^{12}$ lower than the current frequency of TAI [5,7,8]. Thus, we may conclude at this point in time that TAI runs at a frequency too high with respect to the definition of the second by about 1 part in $10^{12}$.

Similarly designed cesium beam frequency standards are nearing completion and evaluation in Japan and in the United Kingdom. In addition to the traditional way of building a special, usually long, cesium beam machine in order to be able to evaluate the biases, new techniques have been developed. They may permit an evaluation of most existing cesium beam standards, whether they were designed for evaluation or not, including commercial beam tubes [9,10]. Thus, many more laboratories could join the group of laboratories owning primary frequency standards provided they go through certain electronic test procedures with existing cesium beam standards found suitable for the application of these techniques. This may increase the confidence in the value of the unperturbed cesium frequency.
II. Hydrogen Devices

The hydrogen maser has been under study since 1960 [11]. It has been presently developed to be one of the most stable oscillators, with a fractional frequency stability of $2 \times 10^{-15}$ and better [12] which exceeds that of a cesium standard for measurement times of the order of an hour. However, the stability of the hydrogen maser is not better than that of cesium for measurement times much longer than one day. In other words, the present principal utility of hydrogen maser oscillators lies in highly precise measurements of relatively short duration but its performance as a clock is not superior to that of cesium beam tubes. The accuracy of the hydrogen maser is mainly limited due to the uncertainty in the knowledge of the frequency shift caused by the collision of the hydrogen atoms with the walls of the storage bulb used inside of the microwave cavity. The traditional evaluation methods have been shown by several laboratories (NBS, NRC, NPL) [13,14,15] to yield an accuracy of about 1 part in $10^{-7}$. These methods are based on testing a series of storage bulbs of different size in the same hydrogen maser. In an operational sense, this method also suffers the disadvantage of not allowing an easy and operational re-check of the accuracy of a given device because its operation has to be interrupted for a considerable length of time for any reevaluation procedure.

The use of the hydrogen atom in devices other than the hydrogen maser oscillator has been proposed. Two experimental devices have been built and the results obtained are encouraging. One device (at NBS) is a hydrogen storage bulb system in which a crystal oscillator is locked to the hydrogen resonance via atomic hydrogen detection or via receiving the microwave hydrogen dispersion [20,21]. The other device (at NASA) is a free hydrogen beam device using a Ramsey type microwave cavity and atomic hydrogen detection in order to slave a crystal oscillator to the hydrogen resonance [22].
III. Other Devices

Atomic beam tubes based on the thallium atom have been developed and have been tested [16,17]. The experimental results indicated that accuracies may be obtained which are comparable with those of cesium devices. Their operational and functional aspects are very much like those of cesium beam tubes. From this work it may be concluded that the use of an atom as similar to cesium as thallium does not lead to devices which are fundamentally superior to the cesium beam tube. In fact, certain aspects in the practical operation of such a beam tube are inferior as compared to cesium beam tubes. Therefore, to our knowledge, no work is presently performed on thallium devices.

Other atomic beam devices such as barium oxide [18] or magnesium devices [19] have been considered and have been or are presently being tried. Indications are that these devices have some potential; e.g., reduced sensitivity to magnetic fields, possibility of very high beam intensities, exceptional line-Q due to higher transition frequencies, etc. However, further research is necessary to evaluate the practicality of such advantages.

Ion storage devices in which charged atoms are stored within containing electromagnetic fields have been proposed for a long time and research has been done to a considerable extent [23,24] using mainly the He ion. Recently, actual frequency standard devices based on ion storage have been experimentally built and are currently under test [25] using either mercury or barium ions. Experimental results on their capability are not available at this time.

The whole class of stabilized lasers, where molecular resonances are used to slave a laser oscillator to a fixed frequency, have been tested over the past 5 years with considerable experimental success in stability and promise in accuracy [26,27,28]. Present devices have not yet demonstrated better than 1 part in $10^{11}$ in accuracy. However, no real physical limit down to $10^{-14}$ appears to be known. The most promising candidate is methane (several transitions in methane could be utilized). In a practical sense, the drawback of these optical devices, of course, is the difficulty to obtain precise second ticks. Further refinement of frequency multiplication schemes from the microwave region into the infrared and visible radiation region are therefore necessary both in terms of precision and in technical perfection.
Future Outlook of Primary Frequency Standards

For the next several years it can be assumed with reasonable certainty that cesium beam tubes will remain the basis of the legal definition of the second, as well as the practical sources for the generation of time scales and the unit of time interval. Further improvement in cesium devices is likely which may push the accuracy to better than 10^{-13} within the next few years. These improvements will be based on new designs allowing quicker and easier evaluation of the most significant frequency biases as well as improvements in the stability of these devices which allow faster evaluation and measurement procedures. It will be difficult in the near future for any other device to exceed the cesium atomic beam machine in terms of accuracy. Thus, for practical devices we can be reasonably certain that for the next several years cesium beam devices will remain the workhorse of primary frequency and time standards laboratories. We can also be sure that for the foreseeable future the cesium beam frequency standard will remain the basis of the legal definition of the second.

If one considers accuracy as the primary objective, then beam machines appear to be the best choice since they approximate the ideal of the free, unperturbed atom to the highest degree. In contrast, storage devices such as the hydrogen devices, (maser oscillators or hydrogen storage beam tubes), suffer from the unavoidable interaction of the atom with the storage container. The future of the hydrogen maser as an accurate device hinges crucially on better evaluation procedures. Presently, very little work is going on in this direction. The most notable effort is the big-box storage bulb maser with a variable storage bulb surface-to-volume ratio [29,30]. It is the intention of at least three other laboratories to initiate investigations with hydrogen devices using new techniques [31,32] for measuring the wall shift. In this connection, hydrogen beam or hydrogen storage beam devices appear to be attractive since they offer the use of the known techniques with greater ease (and hopefully precision), than the maser oscillator. New results in this direction may be expected within the next several years, and accuracies rivaling that of the present day cesium beam standards are likely to be achieved in the future.
Ion storage devices are in the initial phase of being investigated in terms of their capability as frequency standards. Again, first experimental results may be expected within the next two years. In ion storage devices, however, it is difficult to know precisely the velocity of the stored ions; the magnitude of the velocity is crucial to the evaluation of the second-order Doppler effect. Also, an increase in the number of stored particles in the ion storage trap is necessary in order to make it competitive with hydrogen or cesium devices.

Laser devices are, of course, promising, and are being investigated in an increasing number of laboratories in the world. As far as primary frequency standard applications are concerned, the value of these laser-based devices is crucially coupled to the multiplication from the microwave into the infrared and visible radiation regions. Without a practical* and precise multiplier or synthesizer, these devices are not attractive as primary frequency standards or clocks in the classical sense.

Again, new results on the accuracy and on the future potential of multiplication techniques may be expected within the next few years allowing a more final evaluation of the potential of laser-based devices. They are, of course, in their own right of great interest as secondary frequency standards and wavelength references for the optical region.

In connection with new developments in the multiplication from the microwave region upwards, not only the saturated absorption techniques become even more attractive but also beam devices become of interest again: As was mentioned above, they approximate in the best way the ideal of the free and unperturbed atom; thus, beam devices in the far infrared or at even higher frequencies may be of considerable interest. Spectrally pure and convenient excitation sources may be found which match with convenient transitions such as in magnesium or calcium.

*At present, such systems are based on the fragile, fairly unreliable catwhisker type diode and several lasers (H$_2$O, CO$_2$, HCN) which serve as intermediate oscillators in the chain [33].
Summary

It may be predicted that within a few years accuracies of better than one part in $10^{13}$ will be realized with cesium machines. Cesium will remain the workhorse of the primary frequency and time standards for many years to come. Beam devices, including cesium but including also other atoms have the highest probability of reaching even higher accuracies. Storage devices such as those based on hydrogen or storage cells as used in laser stabilization have the potential of equalling or possibly surpassing cesium and are therefore of great interest.

Several developments may be expected to influence the field for years to come. (1) An increasing availability of standards (cesium as well as others) with evaluated accuracies. (2) Several laboratories will compare via TAI their accurate frequencies. (3) The possibility -- due to new techniques -- to automatically control a standard to remain accurate, and (4) the trend towards improved accuracies which are comparable with long term stability values. Thus, operational clocks and corresponding time scales may well be stable and accurate continuously without resorting to occasional (elaborate) calibration and evaluation procedures.
**TABLE:**

**PRESENT STATUS AND FUTURE OUTLOOK FOR ACCURACIES**

<table>
<thead>
<tr>
<th>Class of Devices</th>
<th>Accuracy</th>
<th>Confidence for &quot;Projected&quot;</th>
</tr>
</thead>
<tbody>
<tr>
<td>Microwave Beams (Cs, H, Tl, BaO)</td>
<td>(Cs) $1.5 \times 10^{-13}$ [5,7,8] $\leq 1 \times 10^{-13}$</td>
<td>90%</td>
</tr>
<tr>
<td>Submillimeter Beams (Mg, Ca)</td>
<td>- - -</td>
<td>$\leq 1 \times 10^{-14}$</td>
</tr>
<tr>
<td>Maser Oscillators (H)</td>
<td>(H) $1 \times 10^{-12}$ [13,14,15] $\leq 1 \times 10^{-13}$</td>
<td>80%</td>
</tr>
<tr>
<td>Storage Beams (H)</td>
<td>- - -</td>
<td>$\leq 1 \times 10^{-13}$</td>
</tr>
<tr>
<td>Ion Storage (Hg, Ba, He)</td>
<td>(He) $1 \times 10^{-9}$ [23]</td>
<td>$\leq 1 \times 10^{-12}$</td>
</tr>
<tr>
<td>Stabilized Lasers (CH$_4$, I$_2$, CO$_2$, SF$_6$, etc.)</td>
<td>(CH$_4$) $1 \times 10^{-11}$ [27]</td>
<td>$\leq 1 \times 10^{-13}$</td>
</tr>
</tbody>
</table>

This table lists seven types of frequency standards and their corresponding accuracies at the time of this writing as presently achieved with at least one of the listed molecules. The figures for the "projected" accuracy are based on published or public statements by the workers closely associated with these devices. The last column gives the author's confidence estimate (condition: unlimited resources and time to reach objective) for an actual realization of the "projected" accuracies using one of the possible atoms.
REFERENCES


12. Stabilities of $2 \times 10^{-15}$ for around $10^3$ s sample time were reported independently by R. F. C. Vessot, Smithsonian Astrophysical Observatory, Cambridge, MA; H. E. Peters, NASA-GSFC, Greenbelt, MD; and C. Audoin, Laboratoire de l'Horloge Atomique, Orsay, France.


25. C. Audoin, Laboratoire de l'Horloge Atomique, Orsay France; private communication.


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