Atomic Frequency Standards for Ultra-High-Frequency Stability

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In this article, the general features of the $^{199}\text{Hg}^+$ trapped-ion frequency standard are outlined and compared to other atomic frequency standards, especially the hydrogen maser. The points discussed are those which make the trapped $^{199}\text{Hg}^+$ standard attractive—high line $Q$, reduced sensitivity to external magnetic fields, and simplicity of state selection, among others.

I. Introduction

Since the inception of the DSN, the need for stable frequencies to support navigation and certain radio science experiments has been well recognized. In the early days of the DSN, when missions were limited to lunar distances, the precision required for navigational parameters implied that stabilities on the order of one part in $10^{10}$ would be adequate for all applications. For this level of performance, the quartz crystal oscillator was quite adequate.

The more stringent requirements of missions exploring the planets, however, modified this perception. The advent of VLBI further accentuated the need for more stable frequencies, and thus the application of atomic frequency standards became necessary at the DSN.

The first such standards were the rubidium cell devices, with stabilities in the range of $5 \times 10^{-13}$ over averaging intervals of $10^2$ to $10^4$ seconds. Stabilities provided by these standards were inadequate for a number of applications that subsequently emerged, and thus these standards soon gave way to the more stable hydrogen masers. Hydrogen masers then became the primary frequency standards throughout the DSN, providing users with stable reference signals for navigation and radio sciences.

A number of problems with hydrogen masers have been encountered since the early years of their application in the DSN. The major problem associated with masers relates to reliability. Hydrogen masers experience unexpected failures that are usually associated with the large flow of hydrogen gas into the vacuum pumps. Despite ongoing attempts to predict or prevent these failures, at present the only certain way to assure the DSN of the availability of reference signals is through redundancy. This step is, however, expensive, since hydrogen masers currently cost approximately $450K.

Aside from these reliability issues, a number of more stringent requirements for frequency stability have emerged in the past few years which exceed the current capability of hydrogen masers. The most notable of these requirements is the need for stabilities in the range of one part in $10^{17}$—a level nearly two orders of magnitude beyond the stability of masers currently employed in the DSN.
The cost associated with the procurement and maintenance of the masers, together with the need for better frequency stabilities, led to an effort to identify technologies which could provide the DSN with more attractive alternatives. It was soon recognized that confined-ion technology had the potential to provide a viable alternative to hydrogen masers. This led to the establishment of a task, under the Advanced Systems Program of the TDA, to carry out a research-and-development effort aimed at the production of a mercury standard based on ion confinement. This effort recently culminated in the demonstration of a mercury ion frequency standard, described in detail in a separate article [1]. The purpose of the present article is to outline the technical background of atomic frequency standards and to identify the features which render the trapped-ion device attractive for frequency-standard applications.

The first part of this article will describe the general characteristics of atomic frequency standards. The major portion of this discussion will center on a description of the hydrogen maser, with which there is a more general familiarity. The description of trapped-ion devices will be presented in parallel with that of the maser in order to elucidate features which characterize each device. The final segment of the article will focus on specific characteristics of trapped-ion devices.

II. General Characteristics of Atomic Standards

Atomic frequency standards are based on the discrete energy structure of atomic systems (atoms and ions). A pair of discrete energy levels may be connected by a photon, the energy of which is equal to the energy separation of the two levels. This may be represented as \((E_u - E_l) = hf\), where \(E_u\) and \(E_l\) are the energies of the upper and lower levels, respectively; \(h\) is Planck's constant; and \(f\) is the frequency of the photon.

For an excited state, there is a finite probability that the atom may remain in that state before decaying to a state of lower energy by emission of a photon. The manifestation of this probability is the natural energy width associated with every excited state. The frequency of the emitted photons then exhibits a finite width corresponding to the energy width of the upper state.

This quantum mechanical condition provides a recipe for establishing precise sources of frequencies based on atomic structures. One selects an atomic system containing a suitable pair of energy levels, with the upper level having a narrow width. The frequency of the photon connecting the two levels will then define the precise frequency on which the standard could be based.

The fundamental limitation on atomic frequency standard operation is imposed by its operating frequency, \(v_0\); by the number of atoms or ions available, \(N\); and by the interrogation time, \(T\). Since the radiation-limited lifetime of atomic microwave transitions is essentially infinite (10^4 years is typical), an effective limit is imposed instead by the interrogation time in which the frequency source is required to operate. For measurements shorter than (and equal to) this interrogation time, a secondary frequency source must be provided. This limit can be expressed as

\[
\sigma_f(\tau) = \frac{1}{2v_0} \sqrt{\frac{T}{NT}} \sqrt{\frac{T}{\tau}}
\]

From this it is seen that high stability is achieved by the following factors: high frequency operation allows a small fractional linewidth to be determined within the allowed interrogation time; many particles yield good statistics to determine the frequency even within that linewidth; and well isolated atoms (together with an excellent local oscillator) allow a long interrogation time.

The simplicity of the principle outlined above greatly distorts the degree of difficulty encountered in actual implementation. For one thing, statements made in the paragraphs above pertain to "unperturbed" atoms. This signifies atoms that are free of the influence of all external fields and free of interaction with all external systems. Such an ideal situation is difficult to realize, and thus one would have to be content with providing an environment which minimizes the influence of external fields and interactions. Furthermore, the frequency of the photon should be in a practical range that is easily accessible for measurement and conversion. At the current level of technology, this implies frequencies in the microwave regime, limiting the suitable energy states to the hyperfine levels of atomic systems, or vibrational levels of molecules.

These limitations reduce the number of candidate atomic systems suitable for frequency-standard application to the hyperfine levels of alkali and alkali-like atoms. Alkali atoms, such as rubidium and cesium, have a single electron outside a closed shell, resulting in a simple ground-state hyperfine structure. This characteristic is also a feature of the hydrogen atom and the mercury ion.

Atomic systems are utilized in two modes for frequency-standard applications. In the active mode, the atom acts as an oscillator, emitting radiation with the characteristic sharp center frequency and narrow linewidth that is then used to stabilize the frequency of a slaved oscillator. In the passive mode, the role of the atom is much like that of a filter for the absorbed radiation derived from the slaved oscillator. exam-
amples of active atomic frequency standards include the hydrogen maser and the rubidium maser. Passive standards include the cesium beam clock, the rubidium gas cell standard, and the trapped mercury ion standard. The variance of the stability for active sources characteristically shows a $1/\tau$ dependence on the measuring time, while that for passive sources shows a $1/\sqrt{\tau}$ dependence. The $1/\tau$ behavior results from follower amplifier noise added to the very weak microwave signal available from the atomic radiators. In practical terms, an active source (the hydrogen maser) shows rapidly increasing stability at moderate averaging times ($\tau < 1000$ seconds), while passive sources show improvement that is slower but that extends to longer averaging times ($\tau > 10,000$ seconds).

III. Operational Characteristics

Whether the atomic system is applied in the active or passive mode, it is first necessary to prepare the atom in the appropriate hyperfine state. In thermal equilibrium, the population of energy levels of the atom is governed by the Boltzmann distribution for a given temperature. At room temperature, hyperfine levels of the ground state have nearly equal populations owing to the relatively small differences in their corresponding energy splitting compared to the thermal energies. In order to absorb or emit photons at the hyperfine frequency $f$, a population difference between the levels must be created.

The atoms in the desired hyperfine state may be selected either by rejecting the ones in the undesired state or by converting all atoms to the desired state. In the case of the hydrogen maser and the cesium clock, this preparation is accomplished by passing the beam of atoms through an inhomogeneous magnetic field which focuses the desired species and rejects atoms in the unwanted states by diverting them out of the beam. This action is due to the interaction of the atomic magnetic moments, corresponding to a given hyperfine state, with the magnetic field gradient.

For the rubidium cell standard and the trapped mercury ion device, atoms in the unwanted state are converted to the desired state via a process called optical pumping. In this process, atoms in the undesired hyperfine state absorb an optical photon from a resonant light source and are promoted to an excited energy state. The excited state is selected such that the excitation is short-lived (typically lasting a few nanoseconds) and transitions are allowed to both hyperfine states. Since the excitation takes place out of the undesired state but the decay of the optically excited states is to both hyperfine levels, the population of the undesired state is depleted, and eventually all atoms absorbing the pump light end up in the desired hyperfine state.

Optical pumping of rubidium and cesium is made possible by dye lasers and semiconductor lasers. Use of lasers for optical pumping of mercury is not practical because the required UV radiation has a wavelength of 194 nm, which is outside the range of present-day lasers. Frequency mixing techniques are possible, however, and the time and frequency research group within the National Bureau of Standards has demonstrated laser pumping of mercury ions [2].

After state selection, the energy structure of the atomic system is interrogated for the precise frequency. In the case of the hydrogen maser, the beam of atoms in the upper hyperfine state enters a high-Q cavity which is tuned to resonate at the hydrogen atom hyperfine energy difference (1.4 GHz). The oscillation in this cavity coherently stimulates the emission of more 1.4-GHz radiation from the atoms into the cavity, overcoming its losses and sustaining the maser oscillation.

The atomic linewidth is limited by the interrogation time, which for the maser corresponds to the time the atoms are contained in the high-Q cavity. The atoms are contained for about 1 second inside a Teflon-coated quartz bulb, giving 1 Hz atomic linewidth. The bulb occupies a region of the cavity where the microwave field has nearly constant phase and is smaller than the wavelength of the emitted radiation, thus eliminating the first order Doppler effect through a process called Dicke narrowing [3].

During the 1 second storage time, the hydrogen atoms undergo many collisions with the walls of the quartz bulb; they also undergo spin exchange collisions with each other as well as with other atomic and molecular species that may be present in the vacuum system. These collisions limit the performance of the maser in a number of ways. The hydrogen atom energy levels are shifted because of the large local electromagnetic fields encountered during collisions. This generates a frequency offset from the ideal unperturbed hydrogen atom that will vary from bulb to bulb and with the density of hydrogen atoms in the bulb.

Collisions with the walls of the quartz bulb also constitute a loss mechanism for the atoms in the desired hyperfine state; recombination and other chemical reactions reduce the number of excited atoms. One consequence of this loss is a reduction in the power level output of the maser and an associated reduction in the signal-to-noise ratio. Ions contained in an electromagnetic trap are free of perturbations associated with wall and spin exchange collisions. Ions generated by the impact of electrons on a background gas are trapped in a small region of space, typically smaller than a cubic centimeter. The confinement is produced through the application of appropriate electromagnetic fields to a three-electrode trap structure with the appropriate geometry. The
fields may be static electric and static magnetic (as employed in Penning traps), or they may be rf electric fields (employed in the Paul’s or rf trap). Depending on the geometry of the trap and the value of the field parameters, a particle with a given ratio of charge/mass can execute closed orbits and be trapped. Like the quartz bulb in the hydrogen maser, the ion trap confines the ions to a region smaller than the wavelength of the 40.5 GHz radiation (~7.5 mm), thus eliminating the first order Doppler effect.

The problem of wall collisions and their associated frequency shift is thus eliminated for trapped ions. Furthermore, the small density of the ions (on the order of 10^6 per cubic centimeter) resulting from the space charge effect virtually eliminates de-phasing collisions with other confined ions. Finally, the confinement time of the ions is governed by the density of the background gas, which may increase the kinetic energy of the ions through collisions and knock them out of the trap. The confinement time, however, can be made as long as many hours, depending on the vacuum conditions. This means that narrower lines corresponding to longer interrogation times may be obtained with trapped ions.

Since one of the parameters governing the stability of the atomic frequency standard is the line Q (where Q = Δf/f0, with Δf the linewidth and f0 the center frequency), a transition with a larger center frequency yields a higher attainable stability for the same Δf and the same signal-to-noise ratio. Thus the line Q of the hydrogen maser is typically a few times 10^9, corresponding to the linewidth of a hertz and a transition frequency of 1.4 GHz. The hyperfine transition of mercury ions (isotope 199), by comparison, is about 40.5 GHz. A linewidth of one hertz produces a Q which is nearly a factor of 30 larger than the corresponding Q of hydrogen. This feature, together with the simple hyperfine structure of the ground state and the possibility of optical pumping with a discharge lamp, renders mercury ions particularly suitable for trapped-ion standards.

Two other characteristics of mercury supplement the features mentioned above. The large mass of the mercury ion implies a small frequency shift due to the second order Doppler effect. This motional effect is due to relativistic time dilation, whereby the frequency of a moving ion is slightly shifted with respect to an observer in the laboratory frame of reference. For particles with a given temperature and thermal energy, those with the largest mass have the smallest speed and thus the smallest second order Doppler shift. This effect represents the ultimate limitation on the long term frequency stability of the trapped mercury ion standard. The size of the effect at room temperature is on the order of one part in 10^{13} for room temperature Hg^+ ions. However, because the trapping force in an rf trap is generated by the motion of charged particles in an inhomogeneous oscillating electric field, 10^6 Hg^+ ions held under typical trapping conditions will have a second order Doppler shift of about one part in 10^{12} [4]. Trap designs which do not increase the second order Doppler shift in this way but that have 10^6 or more ions will be discussed in a future article. Obviously, cooling the particles will lead to the reduction of the second order Doppler effect. Here again, trapped ions lend themselves to drag cooling produced through collisions with a light, inert gas such as helium or by laser cooling. The latter process amounts to the loss of kinetic energy through momentum transfer associated with the directional absorption of photons, followed by the emission of photons in all directions. Such an approach requires a laser at 194 nm, and the NBS group [2] has demonstrated laser-cooled mercury.

Finally, the ease of introduction of mercury in the vacuum system is an important consideration. Isotopic mercury is easily liberated from oxide material with negligible vapor in the needed quantities. Too much background vapor of mercury reduces the trap time through collisions, while too little requires longer periods for loading the trap with ions. Operating pressures of 10^-9 torr are readily achieved in the trap. This reduces the gas load to the vacuum system, allowing long-term and reliable operation with small sorption or ion pumps.

By contrast, the production of hydrogen atoms from naturally occurring hydrogen in the molecular form requires the use of an rf-powered dissociator. The dissociator has an efficiency of atom production typically in the five percent range. The aging of the dissociator also represented a source of failure for the early masers. The low efficiency of atom production and the required high flux of the continuously flowing hydrogen produce a significant load for the maser vacuum pumps, which are required to maintain a background pressure in the 10^-6 torr range. The reliable operation of these pumps, also of the chemical sorption or ion variety, represents the major challenge associated with hydrogen masers.

IV. Sensitivity to Environmental Perturbations

Atomic frequency standards are generally extremely sensitive to environmental influences. Ambient electromagnetic fields perturb the structure of the energy levels and the corresponding frequencies of transition. Thus, the presence of magnetic fields disturbs the atomic level via the Zeeman effect. The Stark effect reflects the interaction of electric fields with energy states. Strong electromagnetic radiation interacts with the atomic levels and results in the “light shift,” or the dynamic Stark effect.
The degree of perturbation produced by the ambient fields naturally depends on the intensity of the fields. However, the interaction is also dependent on the particular level of the particular atomic system. Thus, for example, the dependence of the frequency shift of a trapped mercury ion on ambient magnetic field variations is some 30 times smaller than the corresponding shift in the maser.

Another important source of frequency drift for the maser is the so-called cavity pulling effect. This effect is based on the fact that the maser frequency is related to the hydrogen-atom frequency and the cavity frequency. Any temperature drifts that can cause dimensional changes in the cavity will change the cavity resonance frequency, which in turn will alter the maser frequency. This effect may be reduced through control of the temperature of the cavity, which is chosen from materials already low in their coefficients of thermal expansion.

Nevertheless, the sensitivity of the maser frequency is high enough to detect shifts corresponding to dimensional changes of the cavity at the 0.25 angstrom level. The problem is stabilized to some extent through various schemes for “autotuning” of the cavity [5]. Other environmental effects, such as changes in barometric pressure and even in humidity, have been observed to influence sensitive components to produce frequency drifts [6]. The same mechanism for frequency drift due to dimensional changes does not exist for the trapped mercury ion standard, since there is no resonant cavity. An important ramification of this fact is that bulky temperature controlling shields and ovens are not required. This feature simplifies the structure and reduces the associated mass of the device.

V. Trapped-Ion Technical Aspects

In the preceding sections, general features of atomic frequency standards were considered, and comparisons were made between the hydrogen maser and the trapped mercury ion device. It was indicated that because of its inherent properties, the trapped-ion device offers the potential for a frequency standard which is simpler in structure, has less mass and size, is more reliable, and has better stability performance.

Nevertheless, the trapped mercury ion standard has a few features which pose technological challenges that must be met before the full potential of this device can be realized. Some of the features unique to the trapped mercury ion are described in this section.

The confinement time of ions depends on a number of parameters, the most significant of which is the collision rate of mercury ions with background mercury vapor and with residual impurities in the vacuum system. These collisions may also redistribute the population of levels prepared by optical pumping. Consequently, a background pressure of less than $10^{-9}$ torr is desired to diminish the influence of background particles. This level of vacuum is not difficult to realize but requires care appropriate to ultra-high vacuum practices.

Another unique requirement for the trapped mercury ion standard relates to the lamp used for optical pumping. While much has become known about mercury lamps through the experience of the lighting industry over the years, the requirements of the light source for optical pumping are rather different. Here the radiation of interest is due to ion emission, while the mercury neutral emission copiously produced and used in fluorescent lights is the unwanted background. Light emission from the ions implies a higher plasma temperature in the lamp, which in turn requires higher input power. Thus, about 20 watts of rf power is coupled inductively to an rf excited lamp, resulting in accelerated damage to the glass and ultimately in short operating life. A number of design approaches, potential solutions, and candidate technologies are being used to address the problem of lamp life. Nevertheless, the development of efficient and reliable lamps continues to challenge groups active in this work. The frequency stability of the trapped-ion device depends on the signal-to-noise ratio of the scattered 194 nm radiation. An increase in this parameter directly improves the ultimate stability. Improvement of the input optics and collection optics, together with all other steps that enhance the signal-to-noise ratio, will continue to be pursued by workers in this field. A number of approaches leading to increased efficiency of microwave photon detection through the detection of the UV radiation at 194 nm are currently under investigation.

Finally, mention was made of the sources of drift in the device, particularly the offset due to the second order Doppler effect. Cooling of the ions is an approach which reduces most unwanted drift. At the present time, however, a complete and full understanding of all sources of ion heating, both collisional and that due to rf fields, is lacking.
References


