PROSPECTS FOR STORED ION FREQUENCY STANDARDS

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ABSTRACT

Fundamental limitations of possible frequency standards based on stored ions are examined. Practical limitations are also addressed but without regard to size, power consumption, and cost. With these guidelines, one can anticipate that a stored ion frequency standard with accuracy and stability better than 10^{-15} is now possible.

INTRODUCTION

Since the pioneering work of Dehmelt and co-workers, who first observed high resolution microwave spectra on stored $^3He^+$ ions, it has been clear that the ion-storage technique [1] provides the basis for an excellent frequency standard [2-20]. The goals of various groups in this field seem to be determined largely by a trade-off between desired performance and limitations on equipment, such as size, power consumption, cost, etc. In this paper, the fundamental limitations of stored ion frequency standards are addressed. Experiments possible with "available" technology are discussed, but restrictions on experimental equipment, such as size, power consumption, cost, etc., are not made.

In any case, the following assumptions will be made:

(1) The only restriction on experimental equipment is that it be available at a "reasonable cost."

(2) Only experiments where inaccuracy $\leq 10^{-15}$ can be potentially achieved will be discussed.

(3) With this in mind, we will assume that "laser cooling" is employed in all experiments in order to suppress Doppler shifts.

(4) Both rf and Penning traps are considered with advantages and disadvantages of each noted.

Optical frequency standards as well as microwave frequency standards will be discussed. In a meeting on "precise time and time interval," this might seem a bit out of place because of the difficulty of providing time from frequency standards which operate much above 100 GHz [14]. However, these devices will also be discussed because of the other uses for frequency standards and because of the remarkable accuracies potentially achievable.

MICROWAVE FREQUENCY STANDARDS

The dominant choice for a microwave frequency standard is one based on hyperfine transitions in the ground state of a singly ionized atom. Fine structure transitions in an ion such as B^+ might be used [14], but here there are difficult problems with state selection and detection. Exotic choices such as Bk^+ are interesting because of the large hyperfine structure, but this ion has other obvious practical drawbacks.

If we assume that the transition linewidth is fairly independent of the ion species (for example, this is true if the linewidth is determined only by the fundamental limit of interrogation time), then we would like to use an ion with as high a hyperfine frequency as possible. This is why Hg^+ ions are attractive since $\Delta\nu_{hfs}$ ($^{199}Hg^+$) $\cong$ 40 GHz and $\Delta\nu_{hfs}$ ($^{201}Hg^+$) $\cong$ 30 GHz. Simple schemes for laser cooling and optical pumping/detection of hyperfine transitions such as was realized [21] in Mg^+ are possible [22] in other ions like He^+, Be^+, Zn^+, and Cd^+. However, in the case of He^+, the required laser wavelength is too short and for all of these ions, the hyperfine frequencies are about three or more times lower than for Hg^+.

Because of its high hyperfine frequency, large mass (which gives a small second order Doppler shift at a given temperature) and availability of a $^{202}Hg^+$ pumping lamp, $^{199}Hg^+$ has so far received the most attention as a possible microwave frequency standard [2, 4, 5, 7, 10, 11, 19, 20]. If "laser cooling" is employed, the simple schemes [21] using only a laser for cooling and state selection are not possible. However, if the ground state energy levels are "mixed" [9, 16], then laser cooling and optical pumping/detection in Hg^+ ions are possible. Using this "mixing," Ba^+ also becomes a possible choice [6, 9, 13], but the mixing schemes are more complicated than for ^+Hg. Also, since the Ba^+ hyperfine frequency is smaller than for Hg^+, then Hg^+ still seems a better choice. Unfortunately, the first resonance line for Hg^+ which would be used for laser cooling and pumping/

detection is at a wavelength $\lambda = 194.2$ nm. Generating this wavelength in a c.w., narrow band (< 10 MHz) way is difficult but possible using state-of-the-art techniques [16]. With this in mind, experiments have been initiated at NBS to realize a microwave frequency standard based on the 25.9 GHz $(F, M_F) = (2,1) \leftrightarrow (1,1)$ transition in $^{201}Hg^+$ at a magnetic field of 0.534 T [16]. For operation in a Penning trap, this transition is chosen because at 0.534 T, the transition frequency is independent of magnetic field to first order. Therefore, systematic effects due to magnetic field instabilities and inhomogeneities are reduced (see below). If a similar experiment is done in an rf trap, then the 40.5 GHz $(F,M_F) = (1,0) \rightarrow (0,0)$ transition in $^{199}Hg^+$ at low magnetic field would probably be a better choice.

Regardless of the transition or ion used, the prospects for obtaining high Q look very good. Transition linewidths of 0.012 Hz have already been observed [21] in Mg^+; it is anticipated that these narrow linewidths and linewidths even smaller should be observable in Hg^+ which would yield a Q significantly greater than 10^{12}.

In addition to the high Q possible, we note that by observing the scattering of many optical photons (or the absence of many scattered photons) for each microwave photon absorbed, it should be possible to achieve the maximum signal-to-noise -- that is, where the limit is governed by the statistical noise in the number of ions that have made the transition [16, 17]. This will be important in any stored ion experiment, since the number of stored ions are typically rather low.

OPTICAL FREQUENCY STANDARDS

Because of the practical difficulty [14] of producing time from an optical frequency standard, the utility of such a device is restricted. Nevertheless, there would be many uses for such a device used only as a frequency standard; this fact coupled with the potential performance make it interesting to examine. In this discussion, the term "optical" frequency standard is used loosely and will include frequencies above about 1 THz.

When we consider "optical" frequency standards, if we carry our thoughts to their logical conclusion, the obvious choice is to build a γ-ray clock based on a recoilless ("Mossbauer") nuclear transition. The reason we don't think about such things yet is that we don't have the required narrowband, tunable, γ-ray local oscillator. (Not to mention the problems of frequency comparison.) The optical frequency standard problem is similar, but it now appears that very narrow band, tunable, laser sources will be available in the not-too-distant future. Hopefully, in the next few years, tunable lasers will achieve linewidths less than 1 Hz and stabilities over short times of < 10^{-15} [22,23]. With this in mind, the prospects for an optical ion storage frequency standard look very promising.

Again, our choices are somewhat restricted because for the laser cooling and optical pumping/detection functions we require a fairly strongly allowed electric dipole transition. For the frequency standard transitions, however, we desire a very weakly allowed transition in order to obtain a narrow bandwidth. Dehmelt [24] was first to suggest using the intercombination lines of group III B singly ionized atoms for an optical frequency standard; an experiment based on a $T\ell^+$ was suggested. (Note that the highest resolution so far obtained in the visible is on the $^1S_0 \leftrightarrow {}^3P_1$ (657 nm) intercombination line in neutral calcium [25,26].) For $T\ell^+$, laser cooling and optical pumping/detection could be accomplished using the fairly strong $^1S_0 \leftrightarrow {}^3P_1$ line (191 nm). The optical frequency standard would be obtained on the $^1S_0 \leftrightarrow {}^3P_0$ line which has a Q of 5 x 10^{14}! Although difficult, it is certainly within the state of the art to produce these wavelengths by doubling and mixing tunable dye lasers. (With this in mind, a more favorable choice appears to be In^+.) Another possibility which is more attractive from the standpoint of available lasers is to drive the two-photon $^2S_{\frac{1}{2}} \leftrightarrow {}^2D_{5/2}$ transition in Ba^+ [18, 27] or Hg^+ [15, 16]. These transitions have comparable Q to $T\ell^+$ but can suffer from the problem of ac Stark shifts. For example, in Hg^+ if the S→D transition is driven with two photons of equal wavelength (λ = 563 nm), then the ac Stark shift is about 10^{-15} [16]. To make the ac Stark shift negligible, one could drive the $^2S_{\frac{1}{2}} \leftrightarrow {}^2D_{5/2}$ single photon quadrupole transitions [17].

For single photon transitions, it will be desirable to achieve or approximately satisfy the Dicke criterion (confinement dimensions $\lesssim$ $\lambda/2\pi$). This condition is most easily satisfied for a single trapped ion. A single ion is also the most desirable case from the point of reducing systematic effects (see next Section), but suffers, of course, from the standpoint of signal-to-noise ratio. For a single ion which approximately satisfies the Dicke criterion, it is interesting to note that the power required to saturate a transition (assuming the natural radiation decay process is the same as the excitation process) is given by assuming the ion has an absorption cross-section of about $\lambda^2/2\pi$ (case for ions unpolarized). If the laser is focused to about a 1 μm diameter, then a power of only 2 x 10^{-15} W is required to saturate the 202 nm transition in $T\ell^+$. These small required powers may make practical the possibility of producing these short wavelengths by very weak nonlinear processes. Unfortunately, the initial preparation of laser cooled single ions would require substantially higher powers.

For the experiments on single ions, the rf trap may ultimately have an advantage because the confinement can be tighter. It should be noted, however, that even if the Dicke criterion cannot be satisfied, the performance is not severely degraded, since the line is only slightly broadened [24] and more noisy.

SYSTEMATIC SHIFTS

Here we discuss the more important fundamental systematic shifts in possible ion storage frequency standards. They are basically the same in microwave and optical frequency standards, but may differ in magnitude.

(1) Magnetic Fields: In the rf trap, very low magnetic fields would be desirable, and although there would be slight field sensitivities [12, 19], these could be stabilized and calibrated out of the system by locking the field to a Zeeman transition. For example, in the case of the $^{199}Hg^+$ microwave frequency standard, the problem would be the same as in the hydrogen maser. It is sometimes noted that a drawback of the Penning trap is the required large magnetic fields, and the influence these fields have on transition frequencies. These problems can be made very small, however, by operating at a magnetic field where the transition frequency is independent of field to first order. For the $(F,M_F) = (2,1) \leftrightarrow (1,1)$ transition in $^{201}Hg^+$ discussed above [16], the second order field dependence is given by $\Delta\nu/\nu_{hfs} = (\Delta H/H)^2/6$. For the $S\leftrightarrow D$ optical transitions, we obtain a further reduction in sensitivity by approximately the ratio of the hyperfine frequency to the optical frequency ($\sim 10^{-4}$). Since a good magnet system has drift rates $\leq 10^{-8}$ and inhomogeneities $\leq 10^{-8}$ over 1 cm dimensions, field instabilities should not be a problem until well below the 10^{-15} level of accuracy.

(2) Second Order Doppler and Electric Field (Stark) Shifts: The fundamental limits on these two effects will scale together, so they are treated at the same time. Usually only second order "Stark" shifts will be important; therefore, we will be interested only in $\langle E^2 \rangle$.

For single ions, laser cooling has already achieved temperatures between 10 mK and 100 mK [18, 28]. Theoretically, when the motional oscillation frequencies Ω_i (ω_z and ω_r for the rf trap and ω_z, ω_c, and ω_m for the Penning trap) are less than the natural linewidth (γ) of the optical cooling transition, then the limiting "temperature" in each degree of freedom is given by $k_B T \cong \frac{1}{2}\hbar\gamma$ [18, 29, 30], where k_B is Boltzmann's constant. (For a single ion, the precise minimum temperature depends on the angle of incidence of the laser beam and on the spatial distribution of recoil photons [30].) For strongly allowed transitions as in Ba^+ or Hg^+, this limiting temperature is about 1 mK. For more weakly allowed transitions, the temperature is correspondingly less, but other limits such as recoil can come into play, limiting the temperature to about 10^{-6} K.

When the opposite condition ($\Omega_i >> \gamma$) is fulfilled and $\hbar\Omega_i >>$ recoil energy, then the limiting energy [27, 29] is given by $E_i = \hbar\Omega_i\,(\langle n_i\rangle + \frac{1}{2})$ where $\langle n_i\rangle \cong 5\gamma^2/(16\,\Omega_i^2)$. Therefore the limiting kinetic energy is given by $E_{Ki} \cong \hbar\Omega_i/4 << \hbar\gamma/4$. For simplicity, we will assume only the case $\Omega_i << \gamma$ and $\hbar\gamma >>$ recoil energy below; however, even better results are potentially obtained for the opposite condition ($\gamma << \Omega_i$).

For a single ion in an rf trap, when U_0 (D.C. applied potential) = 0, the nonthermal micromotion has an average kinetic energy equal to that of the secular motion (1); this is approximately true in the spherical trap. In the Penning trap the kinetic energy in the nonthermal magnetron motion can be much less than in the cyclotron or axial modes [30]. Therefore, the minimum second order Doppler shifts are given approximately by [31]:

$$\frac{\Delta\nu_D}{\nu_0} = \frac{E_K}{Mc^2} = \begin{cases} \frac{3}{2}\,\hbar\gamma/Mc^2 & \text{rf trap} \\ \frac{3}{4}\,\hbar\gamma/Mc^2 & \text{Penning trap} \end{cases}$$

For a single ion in a spherical rf trap, $\langle E^2\rangle$ is primarily due to the oscillating rf fields and is largest for the z motion. A simple calculation gives $\langle E^2\rangle_z = M\Omega^2\hbar\gamma/e^2$ for maximum laser cooling <u>or</u> $\langle E^2\rangle_z = 2M\Omega^2 k_B T/e^2$ for a given temperature in the z secular motion. For a single ion in a Penning trap, it is usually possible to make r_m, $r_c <<$ z [28, 30], therefore Stark shifts from the static fields are primarily due to the z motion. We find $\langle E^2\rangle_z = \hbar\gamma M\omega_z^{\,2}/(2e^2)$ for maximum laser cooling or $\langle E^2\rangle_z = k_B T M\omega_z^{\,2}/e^2$ at temperature T. In the Penning trap, a larger effect can be caused by the motional electric field $\vec{E}_M = \vec{v} \times \vec{B}/c$. We have $\langle E^2\rangle_M = \hbar\gamma B^2/(Mc^2)$ (maximum laser cooling) and $\langle E^2\rangle_M = 2k_B T\,B^2/(Mc^2)$. In table I are shown examples of the second order Doppler shift and $\langle E^2\rangle$ for single ions in rf and Penning traps.

To get an idea of the effect of electric fields, we note that the fractional Stark shift of Hg^+ hyperfine structure has been estimated to be [32]

$$\Delta\nu/\nu_{hfs} \cong 1.43 \times 10^{-18}\,E^2$$

where E is in volts/cm. (The shift for Ba^+ is about 24 times higher [32].) For the $^2S_{\frac{1}{2}} \leftrightarrow {}^2D_{5/2}$ transitions in Hg^+ we have [16]

$$\Delta\nu/\nu \cong 1.4 \times 10^{-18} E^2.$$

This shift should be similar in magnitude in other optical transitions. We note that in many cases, the electric fields from blackbody radiation ($\langle E^2 \rangle_{bb} \cong (8.3 \text{ V/cm})^2$ at T = 300 K) [32] can be much larger than those due to trapping conditions. Therefore, operations at reduced environmental temperatures may ultimately be required.

For clouds of identical ions, we first consider the electric fields due to collisions between ions. For the rf trap, we neglect the energy in the micromotion since the ions are driven in phase, therefore ion collisional effects in the rf and Penning traps are treated the same. $\langle E^2 \rangle$ due to collisions will, of course, depend on the cloud density and temperature, but some idea of the magnitude can be given by calculating the electric field for one ion on another at the distance of closest approach. Assuming the maximum energy available for closest approach is given by $3k_BT$, we have $E_{max} = 6.7 \times 10^{-8}$ V/cm ($\gamma/2\pi$ = 10 MHz and maximum laser cooling) and $E_{max} = 7.4$ V/cm at T = 4K. Therefore at modest temperatures, ion-ion collision induced Stark shifts can be quite small.

For clouds of ions, other effects can contribute to Stark and second order Doppler shifts. We will consider only theoretical limits and therefore neglect effects such as rf heating in an rf trap, which may be the real limitation in a practical experiment. We will assume that the secular motion in an rf trap and the axial and cyclotron modes in a Penning trap have been cooled to negligible values. For both traps we will assume that it is desirable to maximize the number of ions N.

In an rf trap we must consider the effects of the micromotion and corresponding electric fields for ions on the edge of the cloud. We impose the constraint that the maximum fractional second order Doppler shift ($\Delta\nu_0/\nu_0$) not exceed a certain value (ε). Therefore, for spherical clouds in an rf trap we find [31]

$$N_{max} = 6.48 \times 10^{15}\, r_i M\varepsilon$$

where M is in u (atomic mass units), and r_i is the cloud radius.

For a spherical cloud of ions in a Penning trap, the maximum second order Doppler effect is due to the magnetron motion of ions on the edge of the cloud ($r_m = r_i$, z=0). We find [31]

$$N_{max} = 1.96 \times 10^{13}\, B \sqrt{\varepsilon} \left[r^2_i - \frac{440\, M\sqrt{\varepsilon}}{B} r_i \right]$$

where B is in tesla, M in u. Negative solutions are not physical because they correspond to parameters where the magnetron second order Doppler shift cannot be made as large as ε (for a spherical cloud).

We can also calculate the corresponding electric fields. As before, for the rf trap, the maximum fields occur on ions for $z=r_i$ and $r=0$ and we have [31]

$$\langle E^2\rangle_{z(max)} = \frac{2M\Omega^2 N}{r_i}$$

In the Penning trap, the electric fields cancel along the z axis. Along the radial direction [31]

$$\langle E^2\rangle_{r(max)} = [2Mc^2\varepsilon/(er_i)]^2$$

In table II are shown some representative values of maximum numbers of stored ions and Stark shifts for various values of $\Delta\nu_D/\nu_o$ and r_i on clouds of ions. In certain configurations, second order Doppler and Stark shifts could still be a problem; however, with small enough numbers of ions these can be overcome.

The values in tables I and II are only examples, and of course each experiment would vary. However, table II seems to emphasize that in experiments on clouds of ions, one must be careful to account for Doppler shifts and electric fields due to either the forced micromotion in the rf trap or the magnetron motion in the Penning trap. We also note that in order to obtain very small second order Doppler shifts, very shallow well depths are required [31].

With these extremely low levels of anticipated systematic effects, the search for other effects continues. For example, Dehmelt has pointed out [12] that the shift due to atomic quadrupole moments must be accounted for. In nearly all of the microwave experiments, however, this small shift is negligible; moreover, in single ion experiments it can be calibrated to extremely high levels of precision ($\ll 10^{-17}$).

STABILITIES

With the anticipated high Q's, the expected stabilities are quite high even though the number of ions is rather small. If, as in the microwave case, the linewidths are limited by the interrogation time, then we could expect [16]:

$$\sigma_y(\tau) = (2\omega_o N_i T\tau)^{-\frac{1}{2}} \qquad \tau > 2T$$

where T is the interrogation time, assuming the time domain Ramsey method is used. For $^{201}Hg^+$ ($\omega_o = 2\pi \cdot 25.9$ GHz), assuming $T = 50$s and $N_i = 8.2 \times 10^4$, we obtain

$$\sigma_y(\tau) = 2 \times 10^{-15}\tau^{-\frac{1}{2}} \qquad \tau > 100s$$

which emphasizes the need for extremely stable oscillators to drive the transition.

In the optical domain, anticipated stabilities are even more dramatic. For the $^2S_{\frac{1}{2}} \leftrightarrow {}^2D_{5/2}$ transition in Hg^+ we expect [16]

$$\sigma_y(\tau) \cong 2 \times 10^{-18}\tau^{-\frac{1}{2}} \qquad \tau \geqq 2s$$

for $N_i = 8.2 \times 10^4$ and even for one ion:

$$\sigma_y(\tau) = 6 \times 10^{-16}\tau^{-\frac{1}{2}} \qquad \tau \geqq 2s$$

Of course, these anticipated stabilities would be limited by available lasers, but perhaps in the future, these theoretical limits may be approached.

CONCLUSIONS

From the above, it is not unrealistic to contemplate frequency standards with inaccuracies << 10^{-15}. These projections have assumed that the experiments would not be limited by local oscillators; however, this clearly is an important limit -- particularly in the case of optical frequency standards. Since this limitation may well be overcome, the future of ion frequency standards looks very promising indeed.

ACKNOWLEDGEMENTS

The author wishes to thank both the Office of Naval Research and the Air Force Office of Scientific Research for continued support.

	$T(K)$	300	4	2.4×10^{-4}
	$\gamma/2\pi$	----	----	10 MHz
rf	$\Delta\nu_D/\nu_o$	8.3×10^{-13}	1.1×10^{-14}	6.6×10^{-19}
rf	$\langle E^2\rangle_z$ (V^2/cm^2)	200	2.67	1.6×10^{-4}
rf	z_{rms} (μm)	170	20	0.15
Pen.	$\Delta\nu_D/\nu_o$	4.2×10^{-13}	5.5×10^{-15}	3.3×10^{-19}
Pen.	$\langle E^2\rangle_z$ (V^2/cm^2)	4.0×10^{-2}	5.4×10^{-4}	3.2×10^{-8}
Pen.	$\langle E^2\rangle_M$ (V^2/cm^2)	499	0.067	4.0×10^{-6}
Pen.	z_{rms} (μm)	1300	145	1.1

TABLE I.

Fractional second order Doppler shifts ($\Delta\nu_D/\nu_o$), Stark fields ($\langle E^2\rangle$), and classical r.m.s. axial amplitudes (z_{rms}) for single ions in rf and Penning traps. When $\gamma/2\pi$ is given, we assume maximum theoretical laser cooling ($\Omega_i \ll \gamma$). For both traps we assume M = 100u. For the rf trap where the trap potential is $\phi(r,z) = A_o \cos \Omega t(r^2 - 2z^2)$, we assume $\Omega/2\pi$ (rf drive frequency) = 1 MHz, A_o = 300 V/cm^2. For the Penning trap, $\omega_z/2\pi$ = 20 kHz, B = 1 T. T is the temperature of the secular motion for the rf trap and the temperature of the cyclotron and axial motion for the Penning trap. $\langle E^2\rangle_z$ is the mean square electric field for motion along the z axis, $\langle E^2\rangle_M$ is the mean square "motional" electric field for the $\vec{v}\times\vec{B}/c$ force in the Penning trap. Note that z_{rms} for the Penning trap can be reduced at expense of increasing $\langle E^2\rangle_z$.

	$\Delta\nu_D/\nu_o$	10^{-12}	10^{-12}	10^{-15}	10^{-15}
	r_i(cm)	0.5	0.01	0.5	0.01
rf	N_{max}	3.2×10^5	6500	320	~ 6
rf	$\langle E^2\rangle_z$ (V^2/cm^2)	760	760	0.76	0.76
Pen.	N_{max}	4.9×10^6	---	1.5×10^5	53
Pen.	$\langle E^2\rangle_{r_i}$ (V^2/cm^2)	0.14	---	1.4×10^{-7}	3.5×10^{-4}

TABLE II

Maximum numbers, (N_{max}) and electric fields ($\langle E^2\rangle$) for "cold" spherical ion clouds in rf and Penning traps. A maximum fractional second order Doppler shift $\Delta\nu_D/\nu_o$ is assumed. The secular motion for the rf trap and the axial and cyclotron motion for the Penning trap are assumed to be frozen out (i.e., cooled to negligible values). r_i = ion cloud radius; M = 100u, $\Omega/2\pi$ = 1 MHz for the rf trap and B = 1 T for the Penning trap.

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