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PROPOSED STORED $^{201}Hg^+$ ION FREQUENCY STANDARDS*

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Summary

In this paper, we discuss the performance potential and the problems of implementing a microwave frequency (and time) standard and an optical frequency standard utilizing $^{201}Hg^+$ ions stored in a Penning trap. Many of the discussions apply to ion storage-based frequency standards in general. Laser cooling, optical pumping, and optical detection of the microwave or optical clock transition could be achieved using narrowband radiation at the 194.2 nm 6p $^2P_{\frac{1}{2}} \leftarrow$ 6s $^2S_{\frac{1}{2}}$ transition, while selectively mixing the ground-state hyperfine levels with appropriate microwave radiation. A first-order field-independent microwave clock transition, which is particularly well-suited to the use of the Penning ion trap is the 25.9 GHz $(F,M_F) = (2,1) \leftrightarrow (1,1)$ hyperfine transition at a magnetic field of 0.534 T. The two-photon Doppler-free $5d^9\,6s^2\,^2D_{5/2} \leftrightarrow 5d^{10}\,6s\,^2S_{\frac{1}{2}}$ transition at 563 nm is a possible candidate for an optical frequency standard. Both standards have the potential of achieving absolute accuracies of better than one part in 10^{15} and frequency stabilities of less than 10^{-16}.

Introduction

In this paper, a specific proposal is made for a $^{201}Hg^+$ stored ion microwave frequency (and time) standard which could have an absolute fractional uncertainty of less than 10^{-15}. We also discuss the possibilities for a $^{201}Hg^+$ optical frequency standard. A future stored ion frequency standard may not take the exact form described here; nevertheless, it is useful to investigate a specific proposal, since many of the same generic problems will be encountered in any standard based on stored ions.

Since the pioneering work of Dehmelt and co-workers, who first developed the stored ion method for high-resolution spectroscopy,[1] it has been clear that these techniques provide the basis for an excellent time and frequency standard.[2-10] This conjecture is based primarily on the ability to confine the ions for long periods of time without the usual perturbations associated with confinement (e.g., "wall shift" as in the hydrogen maser). Starting with the work of Major and Werth[5] reported in 1973, groups at Mainz[7] and Orsay[10] and at least one commercial company[11] have sought to develop a frequency standard based on $^{199}Hg^+$ ions stored in an rf trap. The choice of Hg^+ ions for a microwave stored ion frequency standard is a natural one because its ground-state hyperfine structure is the largest of any ion which might easily be used in a frequency standard, and its relatively large mass gives a small second-order Doppler shift at a given temperature. This work has been developed to a fairly high level; the group at Orsay[10] has made a working standard whose stability compares favorably to that of a commercial cesium beam frequency standard. However, the full potential of the stored ion techniques has yet to be realized; this appears to be due to two problems: (1) Historically, it has been difficult to cool the ions below the ambient temperature; this is made more difficult in the rf trap by "rf heating"[1] —a process not clearly understood, but one that makes it difficult to cool even to the ambient temperature.[12] For both the rf and Penning traps, the inability to cool below the ambient temperature means that one must contend with the frequency shift from the second-order Doppler or time-dilation effect. Although it is possible to calculate this shift from the measured Doppler (sideband) spectra, to

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do so with the required accuracy may be difficult for ions near room temperature. (2) A second problem is that the number of ions that can be stored in a restricted volume (dimensions $\lesssim$ 1 cm) is typically rather small ($\lesssim 10^6$). This, coupled with the somewhat poor signal-to-noise ratios realized with conventional lamp sources, causes the short-term stability in a frequency standard based on ions to be degraded, even though the Q's realized are quite high.

In the past two or three years, both of these problems have been addressed in experiments and the results suggest viable solutions. In 1978, groups at the National Bureau of Standards (NBS) and Heidelberg demonstrated[13,14] that radiation pressure from lasers could be used to cool ions to temperatures < 1 K, thereby reducing the second-order Doppler shifts by 2-3 orders of magnitude below the room temperature case. As discussed below, the cooling is most favorable for very small numbers of ions (down to one ion), so that there is a trade-off between the maximum number of ions we can use and the minimum second-order Doppler shift that can be achieved.

To improve signal to noise, we note that in certain optical-pumping, double-resonance experiments, it is possible to scatter many optical photons from each ion for each microwave photon absorbed.[9] This can allow one to make up for losses in detection efficiency due to small solid angle, small quantum efficiency in the photon detector, etc., so that the transition probability for each ion can be measured with <u>unity</u> detection efficiency. This means that the signal-to-noise ratio need be limited only by the statistical fluctuations in the number of ions that have made the transition.[15] This is discussed in a simple example in Appendix A.

More recently, the narrow linewidths anticipated for the stored ion technique have been observed. A resonance linewidth of about 0.012 Hz at 292 MHz has been observed for the $(m_I, m_J) = (-3/2, 1/2) \leftrightarrow (-1/2, 1/2)$ hyperfine transition of $^{25}Mg^+$ at a magnetic field of about 1.24 T where the first derivative of the transition frequency with respect to magnetic field is zero.[16] (The Ramsey interference method was implemented by applying two rf pulses of 1 s duration separated by 41 s). These narrow linewidths should be preserved with hyperfine transitions of higher frequency, such as in $^{201}Hg^+$, but, of course, more attention must be paid to field homogeneity and stability.

These results have encouraged us to begin studies on the $^{201}Hg^+$ system and although this ion may not provide the "final answer," it appears to provide a case where inaccuracies significantly smaller than 10^{-13} can be achieved. The discussion here is largely devoted to a microwave frequency standard with a design goal of accuracy better than 10^{-15}; however, the possibilities for a stored ion optical frequency standard in $^{201}Hg^+$ are also briefly included.[17,32] For a given interaction time, the Q of a transition will scale with the frequency. Therefore, in principle, an optical frequency standard would have clear advantages over a microwave frequency standard. Our decision to work on a microwave frequency standard (as well as an optical standard) is motivated largely by practical considerations: (1) Before the full potential of an ion-storage optical frequency standard could be realized, tunable lasers with suitable spectral purity must become available. This problem may be nearing solution.[20] (2) If an optical frequency standard is to provide time, the phase of the radiation must be measured. This appears to present a much more formidable problem.[18,19] Both of these problems are already solved in the microwave region of the spectrum—thus, the attraction for investigating a microwave frequency standard.

<u>$^{201}Hg^+$ Stored Ion Microwave Frequency Standard</u>

$^{201}Hg^+$ ions will be stored in a Penning trap. The choice of the Penning trap over the rf trap is motivated primarily because it appears easier to cool a <u>cloud</u> of ions in a Penning trap than in an rf trap. Residual heating mechanisms in the Penning trap are quite small[13] whereas in the rf trap (where "rf heating" occurred), they can be substantial.[14] Of course, this problem does not exist for the rf trap if single ions are used, but for a microwave frequency standard, it is desirable to use as many ions as possible in order to increase signal-to-noise ratio. Use of a Penning trap means that one must use transitions that are independent of magnetic field to first order[8,9,16]; this limits the number of transitions available, but need not be an absolute restriction. The $^{199}Hg^+$ isotope is therefore not considered, since there are no frequency extrema at practical fields.

The energy level structure[21] of the $^{201}Hg^+$ ion vs. magnetic field is shown in Fig. 1. The

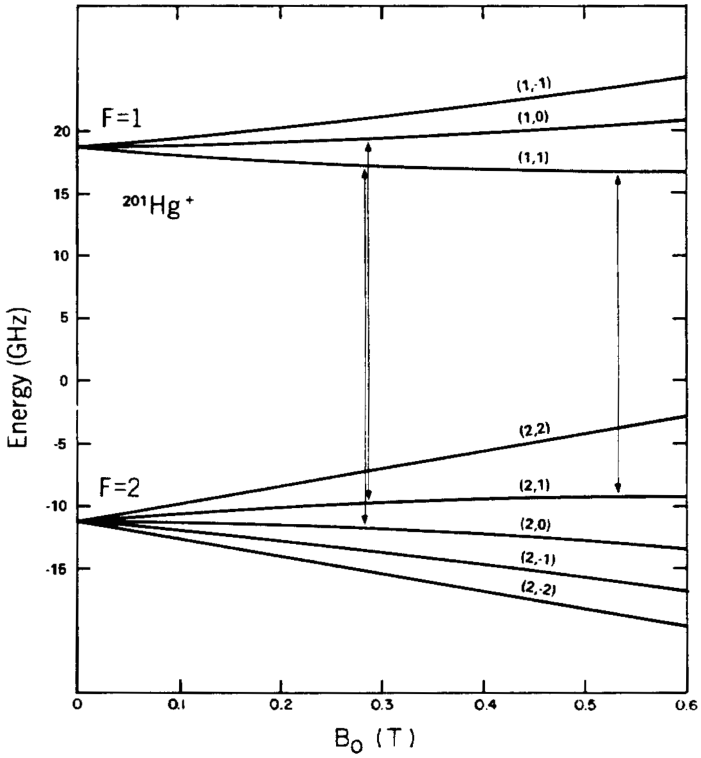

Figure 1. Ground state hyperfine energy levels of $^{201}Hg^+$ vs. magnetic field. States are designated by the (F, M_F) representation. Three transitions are indicated at the fields where the transition frequencies are independent of magnetic field to first order.

possible field independent "clock transitions" are the (F, m_F) = (2,0) ↔ (1,1) and (2,1) ↔ (1,0) transitions at about 0.29 T, the (2,1) ↔ (1,1) transition at 0.534 T, the (1,1) ↔ (1,0) transition at 3.91 T and the (1,0) ↔ (1,-1) transition at 28.1 T. In principle, one desires to work at the highest microwave frequency possible (for highest Q) at the highest attainable magnetic field (to maximize the number of ions). The choice seems to be between the 25.9 GHz (2,1) ↔ (1,1) transition at 0.534 T and the 7.72 GHz (1,1) ↔ (1,0) transition at 3.91 T. At these magnetic fields, where the transition frequency is independent of magnetic field to first order, the second-order field dependence is given by $\Delta\nu/\nu_0$ ((2,1) ↔ (1,1)) = $^1/_6\,(\Delta H/H_0)^2$ and $\Delta\nu/\nu_0$ ((1,1) ↔ 1,0)) ≅ $.04(\Delta H/H_0)^2$. The remaining part of the proposal is modeled around the (2,1) → (1,1) transition at 0.534 T because of its higher frequency. From the second-order field dependence noted above, it is necessary to control the field stability and homogeneity over the ion cloud to better than 10^{-7} in order to achieve 10^{-15} accuracy. This is an important problem, of course, but not insurmountable—we note that the free-running stability of the magnetic field must be better than 10^{-7} over the clock transition time; in longer term it can be stabilized to this level by locking the field to a Zeeman transition in the ions.[16]

We will assume that a laser can be tuned to the 6p $^2P_{1/2}$ ← 6s $^2S_{1/2}$ transition at 194.2 nm with sufficient power to provide laser cooling, optical pumping, and fluorescence detection. A specific scheme for observing the clock transition might be the following: (we will assume that when the ions are cold, the widths of the Doppler-broadened optical lines are approximately equal to the natural width (~ 70 MHz)).

(1) We tune the laser about 35 MHz below the 6p $^2P_{1/2}$ (1,-1) ← 6s $^2S_{1/2}$ (2,-2) optical transition. This laser tuning gives the maximum cooling possible,[22] but rapidly pumps the ions out of the (2,-2) ground state. (For example, the (1,-1) excited state decays to the (1,-1) ground state with a probability equal to 0.329.) For cooling and detection, we require each ion to scatter many photons. Unfortunately, the simple schemes[15,16,23] for multiple scattering that have been realized with ions like $^{25}Mg^+$ do not exist for Hg^+. There-

fore, for the initial cooling, we must mix all the ground states with microwave radiation. This effectively reduces the cooling by a factor of 8 from the two-level ion case, since only $^1/_8$ of the ions are in the (2,-2) ground state, but should not be a problem given enough laser power (see below).

(2) Once cooling is accomplished, we can pump all the ions into one of the clock levels by, for example, mixing all the ground-state levels except the (1,1) level. This pumping occurs because we are exciting the ions in the wings of other optical transitions; for example, we are pumping in the wings of the (1,0) ← (2,-1) optical transition (about 3.5 GHz away) and the (1,0) excited state decays into the (1,1) ground state with a probability of 0.039. (Note that the (1,-1) excited state cannot decay to the (1,1) ground state.) We remark that we have neglected the decay of the ions from the $^2P_{1/2}$ excited state to either the $^2D_{3/2}$ or $^2D_{5/2}$ states. That this is a good approximation is shown in Appendix B. The ratio of the scattering rate on the (1,-1) ← (2,-2) optical transition to the pumping rate into the (1,1) ground-state level is about 6×10^4, i.e., about 6×10^4 photons are scattered by each ion before it is pumped into the (1,1) ground-state clock level. This number can be reduced by tuning the laser to another optical transition; this may be necessary if the laser power is too small. As discussed below, it is desirable for detection purposes for this number to be large.

(3) After the pumping is achieved, the laser and ground-state mixing rf are turned off (to avoid ac Stark shifts and relaxation) and the clock transition is driven. If we use the Ramsey interference method as was done for $^{25}Mg^+$,[16] the resulting linewidth is about one-half as wide as if we use continuous excitation (the Rabi method). Therefore, the Ramsey method will be assumed.

(4) After the rf cycle is complete, the laser is turned back on as well as the mixing rf (excluding the (1,1) ground-state level) and the fluorescence scattering is observed. It should not be too difficult to detect the scattered photons with about 10^{-3} overall efficiency. This would mean that about 60 photons would be collected for each ion that had made the transition (before it is repumped to the (1,1) level) insuring that the noise would only be due to the statistical fluctuations in the number of ions that had made the transition (see Appendix A).

In order to lock a local oscillator to the center of the clock transition, one would first complete one of the above cycles with the local oscillator tuned to the half-intensity point (where the transition probability for each ion is one half) on, say, the low side of the central Ramsey peak. These photon counts could then be stored and the cycle repeated with the local oscillator tuned to the high-frequency side of the line. These resulting counts could then be subtracted from the first to give an error signal which can be used to force the mean frequency of the local oscillator to be at the center of the clock transition. We will assume that the time for fluorescence observation and repumping is much less than the clock transition time.

Systematic Frequency Shifts

We have already mentioned the stringent requirements on magnetic field stability and homogeneity. Using a superconducting magnet, it is possible to stabilize the magnetic field to better than 10^{-7}. (This may require locking the field to an NMR probe adjacent to the trap over the time of the clock transition). The field homogeneity requirements will be more difficult to satisfy (assuming a 1 cm diameter spherical working volume inside the trap), but are still feasible.

The electric fields from the applied trap potentials and from Coulomb interactions between ions can cause second-order Stark shifts, but the resulting fractional frequency shifts are estimated to be much less than 10^{-15}. The black body ac Zeeman shift[38] is estimated to be $\Delta\nu/\nu_0 \cong 1.3 \times 10^{-17}\ (T/300)^2$ and is therefore neglected. The black body ac Stark shift[39] is estimated to be $\Delta\nu/\nu_0 \cong 2 \times 10^{-16}\ (T/300)^4$ and therefore must be accounted for or reduced environmental temperatures are required.

In spite of the laser cooling that has been achieved, we must still be concerned with the second-order Doppler frequency shift. In the Penning trap, the cyclotron-axial temperature of $^{201}Hg^{+}$ ions needs to be cooled to below 1.45 K to insure that the second-order Doppler shifts (on these degrees of freedom) is less than 10^{-15}. These low temperatures should be easy to obtain. A more serious problem exists for the magnetron rotation of the cloud; the kinetic energy in this degree of freedom will probably limit how small the second-order Doppler shift can be.[24] In the limit of very small numbers of ions, the magnetron kinetic energy should be negligible,[25] but in the case discussed here, we would like to use the maximum number of ions possible and this will cause problems as described below.

The "magnetron" rotation in a Penning trap is simply a form of circular $\vec{E} \times \vec{B}$ drift; that is, the radial electric fields in the trap from the applied potentials on the electrodes and from space charge act in a direction perpendicular to the magnetic field. This causes the ions to drift in circular "magnetron" orbits about the axis of the trap. If we assume that we must keep the ions inside a 1 cm diameter spherical working volume, then qualitatively, the nature of the problem is as follows: if we add more ions to this volume, then the magnetron frequency increases due to two effects. First, we must increase the applied trap potentials to overcome the increased space charge repulsion along the z axis, which tends to elongate the cloud in this direction. Consequently, the magnetron frequency increases due to the increased potentials and the increased space charge fields in the radial direction. In Appendix C, we estimate the maximum number of ions contained in a 1 cm diameter sphere assuming that the second-order Doppler shift is 10^{-15} for ions on the perimeter of the cloud at z = 0. We obtain $N_{max} = 8.2 \times 10^{4}$ and note that the applied trap voltage is only 71 mV for $z_o = r_o/1.64 = 0.8$ cm.[1]

Frequency Stability

At optimum power (transition probability is equal to one at line center), we can closely approximate the number of detected photon counts for each experimental cycle as

$$N = N_i \, n_d \left(\frac{1 + \cos(\omega - \omega_o)\, T}{2} \right) \qquad (4)$$

where, as in Appendix A, N_i is the number of ions in the trap and n_d is the average number of detected photons for each ion that has made the transition. ω and ω_o are the frequency of the applied rf and "clock" center frequency; T is the time between the rf pulses at the beginning and end of the rf period. (We assume that the time of the rf pulses is much less than T.) As described above, the interrogating oscillator is switched between $\omega - \pi/2T$ and $\omega + \pi/2T$ (where we assume $|\omega - \omega_o| \ll \pi/2T$), and the resulting counts subtracted to give an error signal. The sensitivity to mistuning of ω is given by calculating the slope of the signal in Eq. 4 at the half-intensity points ($|\omega - \omega_o| = \pi/2T$). We have

$$\left|\frac{dN}{d\omega}\right|_{|\omega-\omega_o| = \frac{\pi}{2T}} = \frac{N_i n_d \, T}{2}$$

After one full switching cycle (taking the difference of the counts from both sides of the line), the error signal is

$$\delta N = 2 \left|\frac{dN}{d\omega}\right|_{|\omega-\omega_o| = \pi/2T} \times \delta\omega$$

where $\delta\omega \equiv \omega - \omega_o$. Since N fluctuates statistically, these fluctuations (δN) give rise to frequency fluctuations in the locked local oscillator:

$$(\delta\omega)^2_{rms} = \frac{\delta N^2}{4 \left|\frac{dN}{d\omega}\right|^2_{|\omega-\omega_o| = \pi/2T}}$$

Maximum frequency stability is thus given by

$$\sigma_y^2(2T) \equiv \frac{1}{2} \frac{(\delta\omega)^2_{rms}}{\omega_o^2}$$

Assuming $n_d \gg 2$ (Appendix A), we have[26]

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

For $N_i = 8.2 \times 10^4$ and $T = 50$ s,

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

and

$$Q = \frac{\omega_o T}{\pi} \cong 2.6 \times 10^{12}$$

Lasers

Perhaps the single reason that such a proposal has not been made previously is that the required narrowband tunable laser has not been available at 194.2 nm. However, it appears that two possible approaches lend themselves to initial experiments in this system. Briefly, the first approach might be to use an externally narrowband filtered (≅ 100 MHz) pulsed ArF excimer laser. We estimate that it should be possible to achieve saturating intensity from such a filtered laser. However, the pulse lengths of these lasers are quite short ($\lesssim$ 10 ns), so that only about two photons per ion will be effective in each laser shot to drive the optical transition. (The lifetime of the upper $^2P_{\frac{1}{2}}$ (1,-1) state is about 2.3 ns and decays with 0.46 probability to the (2,-2) ground state.) The potential advantage of excimer laser systems is that the repetition rates can be quite high (KrF lasers have been built with 1 kHz repetition rates). However, at the present time, it is probably only feasible to realize 150 Hz repetition rates for ArF lasers. This should allow reasonable signal to noise in the above scheme when the ions are cold; however, we estimate cooling times of order 20 minutes, which is uncomfortably long.

A second scheme using frequency mixing of cw lasers in nonlinear crystals is presently being pursued at NBS. Tunable coherent radiation in the 194 nm region has previously been produced by phase-matched sum frequency mixing of pulsed lasers in a potassium pentaborate (KB5) crystal.[27] The second harmonic of the 514 nm Ar^+ single frequency cw laser line, when mixed with radiation near 790 nm from a cw dye laser, will generate single frequency cw radiation at 194 nm. The estimated efficiency of this process for a 3 cm crystal is given by

$$P_3 \cong 8 \times 10^{-5} P_1 P_2,$$

where P_3 is the output power at 194 nm, P_1 is the input power at 257 nm from the doubled Ar^+ laser, and P_2 is the input power at 790 nm.[28,29] All powers are expressed in watts. The second harmonic of 514 nm radiation can be generated in 90° phase matched temperature tuned KDP or ADP crystals with an efficiency (for a 5 cm crystal) given by

$$P_1 \cong 2.5 \times 10^{-3} P_o^2,$$

where P_1 is the output power at 257 nm and P_o is the input power at 514 nm. Thus, about 10 mW can be produced with a 2 W input. The output power can be increased considerably by using a cavity to build up the circulating power. As much as 300 mW of cw power at 257 nm has been produced in this way.[30] Assuming that 200 mW at 257 nm and 500 mW at 790 nm are available, about 8 μW at 194 nm could be produced. From the experience with Mg^+ ions,[13,15,16] this should be adequate power for initial experiments. The output could be further increased by building a cavity around the KB5 crystal to increase the circulating power at 790 nm, 257 nm, or both. Finally, the output power at the ions could be increased by building a ring cavity around the trap. The frequency of the Ar^+ laser can be stabilized to an I_2 absorption. The dye laser can be stabilized and tuned using standard techniques. The temperature of the KB5 may have to be shifted slightly from room temperature in order to satisfy the phase-matching conditions. We estimate from the observed temperature tuning at 201.6 nm[31] that the required shift is less than 25 °C.

Two-Photon Optical Frequency Standard in $^{201}Hg^+$

We will briefly describe the properties of a $^{201}Hg^+$ optical frequency standard that has been suggested previously.[32] ($^{199}Hg^+$ should, of course, also be considered because of its simpler structure.) Using two-photon Doppler-free spectroscopy,[33] it should be possible to excite $^{201}Hg^+$ ions from a ground-state sublevel to a particular $^2D_{5/2}$ sublevel using a dye laser tuned to approximately 563.2 nm. An excited state and magnetic field could be chosen so that the transition frequency could be independent of magnetic field to first order. For the case of this optical transition, the second-order dependence of fractional frequency offset due to magnetic field would be reduced by approximately the ratio of the optical frequency (5.33×10^{14} Hz) to the ground-state hyperfine frequency. This represents a reduction in sensitivity of about four orders of magnitude which would greatly relax the constraints on magnetic field. The ground and excited states could be chosen such that the ground-state level would be depopulated by the two-photon transition; therefore, detection could be accomplished in essentially the same manner as described for the microwave case above. The lifetime of the excited states[35] is about 0.11 s, so that the Q of this transition is about 7.4×10^{14}!

A drawback to this scheme is the accompanying ac Stark shift[33]; this shift is formally equivalent to the "light shift" in rubidium frequency standards. We have roughly estimated the transition probability per unit time to be $W \cong 0.3\ I^2/\delta\nu$ where $\delta\nu$ is the larger of the (doubled) laser linewidth or the natural linewidth (1.4 Hz) and I is the laser intensity in each direction in W/cm². We have also estimated the accompanying light shift to be $\Delta\nu \cong -I$ (Hz). If the (doubled) laser linewidth is less than the natural width and if we drive the transition near saturating intensity ($W \cong 1/s$), then $I \cong 2.2$ W/cm², which implies a fractional frequency shift of 2×10^{-15}.

The black body ac Stark shift[38] is estimated to be $\Delta\nu/\nu_0 \cong 10^{-16}\ (T/300)^4$. The frequency shift[40] due to the interaction of the quadrupole moment of the atomic D levels with the applied quadrupole field and fields due to ion-ion collision is estimated to be less than 10^{-16} for the conditions described here.

In initial experiments, it will very likely not be possible to obtain (doubled) laser linewidths less than 1.4 Hz; however, laser linewidths of a few tens of hertz should be achievable. Therefore, the above projections may not be too optimistic.

Assuming the same conditions as for the microwave case, predicted stabilities are also quite dramatic (assuming laser linewidths are sufficiently narrow). If we assume that the (Rabi) interaction time is 1 s and that the detection and repumping time is much less than 1 s, this would imply (for $N_i = 8.2 \times 10^4$)

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

For $N_i = 1$, $\sigma_y(\tau) \cong 6 \times 10^{-16}\ \tau^{-1/2}$.

Conclusions

Some of the problems associated with a stored-ion frequency standard have been addressed by making a specific proposal around the $^{201}Hg^+$ ion. Although other interesting candidates exist, this system appears feasible enough that experimental work has begun at NBS. Current efforts are aimed at producing the 194.2 nm laser light, producing narrowband 563.2 nm laser light for the two-photon transition, studying the ion cloud dynamics in order to produce the rather diffuse, spatially stable ion clouds and making ion traps with significantly increased collection efficiency.

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APPENDIX A

Statistical Fluctuations in Detected Photon Counts

For simplicity, we will assume the conditions shown in Fig. 2. The actual conditions discussed in this paper and in Refs. 15 and 16 are somewhat

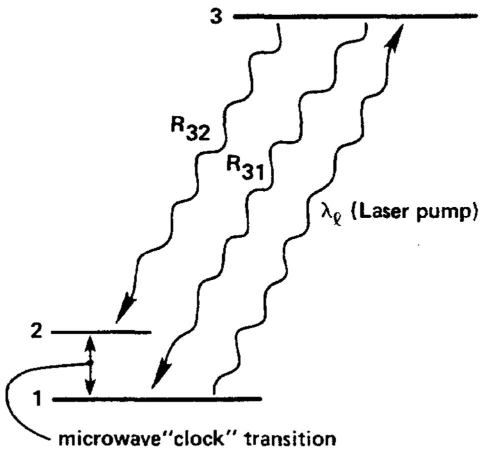

Figure 2. "Model" 3 level system to show the effects of statistical fluctuations in an optical pumping, double resonance experiment.

more complicated, but the basic result still applies. We assume that a laser is tuned to the $3 \leftarrow 1$ transition wavelength and that we can neglect excitation in the wings of the $3 \leftarrow 2$ transition by this laser. We also assume that the decay branching ratios R_{32} and R_{31} are such that $R_{31} >> R_{32}$.

The basic scheme for an optical-pumping double-resonance experiment in this simple example is the following: (1) If the number of scattering events (for each ion) is sufficiently large, then essentially all the ions are pumped into the 2 level. (2) The laser is now turned off and the "clock" $(1 \leftarrow 2)$ transition is driven. (3) The ions that have made the transition are then detected by turning the laser back on and observing the fluorescence scattering by collecting the light in a phototube.

For each ion that has made the clock transition, let the average number of detected photons be n_d; the rms fluctuation in the number of detected photons per ion is $\sqrt{n_d}$. Because of inefficiencies in collection and detection, typically $n_d << R_{31}/R_{32}$, but the interesting case will be when n_d is still significantly greater than one. We will assume that the clock transition is driven with optimum power, so that when we are tuned to the half-intensity point on the line (for maximum frequency sensitivity), the probability, p, that each ion has made a transition is 0.5. In this case, if the total number of ions is N_i, then the average number that have made the transition is $pN_i = N_i/2$ and the rms fluctuations in the number of ions that have made the transition on each experimental cycle is

$$\Delta N_i = \sqrt{N_i\ p(1-p)} = \sqrt{N_i}/2.$$

For one photon counting cycle, the average number of photons detected is $N_{TOT} = n_d\ N_i/2$. The fluctuations in the number of detected photons (ΔN_{TOT}) is due to two causes: (1) The fluctuations in the number of photons counted $(\Delta n_{TOT})_d$ due to fluctuations in the counted photons for each ion. Since these are statistically independent:

$$(\Delta n_{TOT})_d = \sqrt{\sum^{N_i/2} (\Delta n_d)^2} = \sqrt{n_d\ N_i/2}$$

(2) The fluctuation in the number of photons counted $(\Delta n_{TOT})_i$ due to the fluctuations in the number of ions that have made the transition:

$$(\Delta n_{TOT})_i = \Delta N_i\ n_d = n_d \sqrt{N_i}/2$$

Since these two processes are also statistically independent, the total fluctuations in N_{TOT} are given by:

$$\Delta N_{TOT} = \sqrt{(\Delta n_{TOT})_d^2 + (\Delta n_{TOT})_i^2} = \frac{n_d \sqrt{N_i}}{2} \sqrt{1 + \frac{2}{n_d}}$$

Therefore when $n_d >> 2$, the fluctuations in the photon counts are given by the statistical fluctuations in the number of ions that have made the transition. We must note, of course, that the above arguments assume that the laser intensity and cloud of ions is stable, and therefore, fluctuations in signal due to changes in laser-cloud spatial overlap are negligible.

Appendix B

$^2P_{1/2} \rightarrow {}^2D_{3/2,5/2}$ Decay Rates

Decay from the $5d^{10}\,6p\,{}^2P_J$ levels to the $5d^9\,6s^2\,{}^2D_J$ levels is allowed through configuration interaction, which can mix some $5d^{10}$ 6d amplitude into the D states and some $5d^9$ 6s 6p amplitude into the P states.[35] Crandell, et al.[36] have determined that the decay rate from the $^2P_{3/2}$ state to the $^2S_{1/2}$ ground state is 350 ± 30% times greater than the decay rate to the $^2D_{5/2}$ state. If LS coupling is valid, the $^2P_{1/2}$ state decays to the $^2D_{3/2}$ with probability 3×10^{-7}. This decay is highly suppressed because the energy difference is only 933 cm^{-1}, which corresponds to a wavelength of 11 μm. (The decay rate is proportional to the cube of the energy difference.) Decay of the $^2P_{1/2}$ state to the $^2D_{5/2}$ state is forbidden by the electric dipole selection rules, since it requires J to change by 2. Hyperfine and Zeeman interactions mix different J states, making this decay slightly allowed. However, we estimate this probability to be less than 10^{-11} at a magnetic field of 0.5 T. The $^2D_{3/2}$ state decays to the $^2D_{5/2}$ state at a rate of about 54 s^{-1} and to the $^2S_{1/2}$ state at a rate of about 42 s^{-1}. The $^2D_{5/2}$ state decays to the $^2S_{1/2}$ at a rate of about 9.5 s^{-1}.[35] If the laser-induced $^2S_{1/2}$ to $^2P_{1/2}$ transition is denoted by γ_ℓ, the ions reach the $^2D_{3/2}$ state at a rate of $3 \times 10^{-7} \cdot \gamma_\ell$ from which they decay, with probability about 0.56, to the $^2D_{5/2}$ state, where they stay for an average of about 0.11 s before decaying back to the $^2S_{1/2}$ state. If γ_ℓ is less than 6×10^6 s^{-1}, the ions spend less than 10% of their time in the metastable D states, so that this trapping does not cause a problem for the cooling. Transitions to the D states occur at a rate which is much less than the optical pumping rates between ground-state sublevels, so their neglect in the previous discussion on optical pumping is justified.

Appendix C

Maximum Number of Stored Ions

Since the cyclotron and axial kinetic energies are assumed cold, we will assume a uniform charge distribution for the ions.[37] For a spherical ion cloud, the magnetron rotation frequency of the cloud (ω_m) is given from the equations of motion as:

$$\omega_m = \frac{\omega_c}{2} - \sqrt{\left(\frac{\omega_c}{2}\right)^2 - \left(\frac{\omega_z^2}{2} + \frac{4\pi\, e\rho}{3M}\right)} \qquad (C.1)$$

where ω_c is the unperturbed ion cyclotron frequency $\omega_c = eB/Mc$, ω_z is the axial oscillation frequency derived from the applied trap voltage (V_o) as:[1]

$$\omega_z^2 = 4e\, V_o/M(r_o^2 + 2z_o^2)$$

and ρ is the space charge density. We have for the space charge potential from the ions (inside the cloud)

$$\phi_i = \frac{2}{3}\pi\rho\,(r^2 + z^2)$$

and the trap potential may be expressed as

$$\phi_T = \frac{V_o\,(r^2 - 2z^2)}{r_o^{\,2} + 2z_o^{\,2}}$$

When the axial-cyclotron motion is cold, we have $\phi_i(z) = \phi_T(z) = 0$; this implies

$$\rho = \frac{3}{4\pi}\;\frac{M}{e}\,\omega_z^2 \qquad (C.2)$$

If we assume we want the maximum second-order Doppler shift $\left(\frac{1}{2}\left(\frac{v}{c}\right)^2\right)$ less than ε, then we require $\omega_m = c\,\sqrt{2\varepsilon}/r_{c\ell}$ where $r_{c\ell}$ is the radius of the cloud. If we want to maximize the number of ions $N = 4\pi\rho(r_{c\ell})^3/(3e)$, then we want to maximize this expression subject to the above contraints on ω_m. Substituting Eqs. (C.1) and (C.2) into this expression for N we have:

$$N = 2\;\frac{\sqrt{2}\,Mc}{3e^2}\; r_{c\ell}\,\sqrt{\varepsilon}\left(\omega_c\, r_{c\ell} - c\,\sqrt{2\varepsilon}\right) \qquad (C.4)$$

(Values of $r_{c\ell}$ where this expression is negative are unphysical because we assume that ε is a fixed value on the perimeter of the cloud (at $z = 0$) no matter what its size is. For very small clouds, this requires V_0 large enough that the ions are unbounded—the case for $N \leq 0$.)

From (C.2) and the expression for for ω_z^2,

$$V_0 = \pi(r_0^2 + 2z_0^2)\,\rho/3 \qquad \text{(C.5)}$$

For the conditions assumed in the text, $r_{c\ell}$ = 0.5 cm, ω_c = 40.5 kHz, and $\varepsilon = 10^{-15}$ we have $N \cong 8.2 \times 10^4$. Assuming $z_0 = r_0/1.64 = 0.8$ cm, then $V_0 = 0.071$ V.

References

1. H. G. Dehmelt, Advan. Atomic and Mol. Physics 3, 53 (1967) and 5, 109 (1969).
2. F. G. Major, NASA Report X-521.69.167, Goddard Space Flight Center (1969).
3. H. A. Schuessler, E. N. Fortson, and H. G. Dehmelt, Phys. Rev. 187, 5 (1969).
4. H. A. Schuessler, Metrologia 7, 103 (1971).
5. F. G. Major and G. Werth, Phys. Rev. Lett. 30, 1155 (1973) and Appl. Phys. 15, 201 (1978).
6. R. Ifflander and G. Werth, Metrologia 13, 167 (1977).
7. M. D. McGuire, R. Petsch, and G. Werth, Phys. Rev. A 17, 1999 (1978).
8. F. Strumia, Proc. 32nd Ann. Symp. on Freq. Control (Fort Monmouth, NJ, May 1978) p. 444.
9. F. L. Walls, D. J. Wineland, and R. E. Drullinger, ibid. p. 453.
10. M. Jardino, M. Desaintfuscien, R. Barillet, J. Viennet, P. Petit, and C. Audoin, Proc. 34th Ann. Symp. on Freq. Control (Fort Monmouth, NJ, June 1980) p. 353 and Appl. Phys. 24, 1 (1981).
11. M. D. McGuire, Bull. Am. Phys. Soc. 26, 615 (1981).
12. D. A. Church and H. G. Dehmelt, J. Appl. Phys. 40, 3421 (1969).
13. D. J. Wineland, R. E. Drullinger, and F. L. Walls, Phys. Rev. Lett. 40, 1639 (1978).
14. W. Neuhauser, M. Hohenstatt, P. Toschek, and H. Dehmelt, Phys. Rev. Lett. 41, 233 (1978).
15. D. J. Wineland, J. C. Bergquist, Wayne M. Itano, and R. E. Drullinger, Opt. Lett. 5, 245 (1980).
16. Wayne M. Itano and D. J. Wineland, Phys. Rev. A, to be published.
17. See also: H. G. Dehmelt, Bull. Am. Phys. Soc. 18, 1521 (1973) and 20, 60 (1975), and D. J. Wineland and H. G. Dehmelt, Bull. Am. Phys. Soc. 20, 637 (1975).
18. J. L. Hall, Science 202, 147 (1978).
19. D. J. Wineland, Proc. 11th PTTI, (NASA Publ. 2129, Nov., 1979) p. 81.
20. J. L. Hall, Proc. Int. Conf. on Atomic Phys., Boston, Mass. (Aug. 1980).
21. The ground-state hyperfine separation was taken from the measurements of Y. Guern, A. Bideau-Méhu, R. Abjean, and A. Johannin-Gilles, Physica Scripta 14, 273 (1977).
22. D. J. Wineland and Wayne M. Itano, Phys. Rev. A 20, 1521 (1979).
23. Wayne M. Itano and D. J. Wineland, Bull. Am. Phys. Soc. 24, 1185 (1979).
24. A similar limitation for the rf trap is imposed by the kinetic energy in the forced "micromotion" of ions not at the center of the trap. See D. J. Wineland, Proc. 2nd Int. Conf. on Precision Meas. and Fundamental Constants, Gaithersburg, MD, June 1981, to be published.
25. D. J. Wineland and Wayne M. Itano, Phys. Lett. 82A, 75 (1981).
26. The basic scheme proposed for $^{137}Ba^+$ in Ref. 9 should be possible; however, two important errors in this paper should be noted: first, the number of scattered photons before re-pumping was overestimated because pumping in the wings of the other optical lines was not considered. Second, although the improvement in signal-to-noise ratio due to multiple scattering was noted, it was overestimated because it was not assumed that the fluctuations would be limited to the statistical variations in the number of ions that made the clock transition. (See Appendix A).
27. R. E. Stickel, Jr. and F. B. Dunning, Appl. Opt. 17, 981 (1978).
28. G. D. Boyd and D. A. Kleinman, J. Appl. Phys. 39, 3597 (1968).
29. F. B. Dunning and R. E. Stickel, Jr., Appl. Opt. 15, 3131 (1976).
30. P. Huber, Opt. Commun. 15, 196 (1975).
31. R. E. Stickel, Jr. and F. B. Dunning, Appl. Opt. 16, 2356 (1977).
32. P. L. Bender, J. L. Hall, R. H. Garstang, F. M. J. Pichanick, W. W. Smith, R. L. Barger, and J. B. West, Bull. Am. Phys. Soc. 21, 599 (1976).
33. See for example: V. S. Letokhov and V. P. Chebotayev, Nonlinear Laser Spectroscopy, (Springer-Verlag, New York) 1977.
34. Ye. V. Baklanov, V. P. Chebotayev, and B. Ya. Dubetskii, Appl. Phys. 11, 201 (1976).
35. R. H. Garstang, National Bureau of Standards J. Research 68A, 61 (1964).
36. D. H. Crandall, R. A. Phaneuf, and G. H. Dunn, Phys. Rev. A 11, 1223 (1975).
37. See for example: S. A. Prasad and T. M. O'Neil, Phys. Fluids 22, 278 (1979). T. M. O'Neil and C. F. Driscoll, Phys. Fluids 22, 260 (1979).
38. T. F. Gallagher and W. E. Cooke, Phys. Rev. Lett. 42, 835 (1979).
39. L. L. Lewis, Wayne M. Itano, and D. J. Wineland, to be published.
40. H. G Dehmelt, paper in these prodeedings.