FREQUENCY AND TIME STANDARDS BASED ON STORED IONS†

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ABSTRACT

The method of ion storage provides a basis for excellent time and frequency standards. This is due to the ability to confine ions for long periods of time without the usual perturbations associated with confinement (e.g. wall shifts). In addition, Doppler effects can be greatly suppressed. The use of stored ions for microwave frequency standards and the future possibilities for an optical frequency standard based on stored ions are addressed.

INTRODUCTION

Since the pioneering work of Dehmelt and coworkers [1] it has been realized that the techniques of ion storage provide some fundamental advantages over other devices for improved frequency and time standards. This assertion is based largely on the ability to confine ions for long periods of time without the usual perturbations associated with confinement. Samples of ions have been stored in electromagnetic traps for as long as days. [1-3] This means that the interaction time for the ions can be quite long which gives rise to large line Q (transition frequency divided by the linewidth) and high spectral resolution. For example, the linewidth of a cesium beam is limited by the transit time between the two ends of the Ramsey cavity. Linewidths of 0.01 Hz have already been observed for stored Mg^+ ions. [4] This would correspond to a cesium beam tube of about 10 km length. The long term confinement also implies that the average velocity $\langle\vec{v}\rangle$ of the ions approaches zero and first order Doppler shifts can be made very small. [5] This characteristic, which is also shared by rubidium clocks and hydrogen masers, gives an advantage over atomic beam devices where a correction must be made for cavity phase shift errors which are a form of residual first order Doppler effects. In addition, typical confinement dimensions of < 1 cm imply that the Dicke criterion [6] (confinement dimensions < wavelength) can be easily satisfied in the microwave region of the spectrum. This nearly eliminates any first order Doppler broadening of the microwave spectrum. It also appears that the Dicke criterion can be met in the optical region of the spectrum with laser cooling (to be described) on a single stored ion.

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The ion storage technique has the advantage that it lacks the usual perturbations associated with confinement. For example, the frequency shifts associated with collisions of atoms with identical atoms, buffer gases, or container walls such as in rubidium clocks or hydrogen masers are very small. Ions are often stored under conditions of ultrahigh vacuum so that frequency shifts due to ion collisions with background neutrals are negligible. Frequency shifts due to ion-ion collisions are caused by the electric fields of the Coulomb repulsion. These shifts as well as frequency shifts due to the electric fields of the trap can, in many cases, be made extremely small ($< 10^{-15}$). [5,7,8]

Two types of traps have so far been used for atomic clock experiments. The Paul [9] or rf trap uses inhomogeneous rf electric fields to provide confinement in a pseudopotential well [1]. It is the three dimensional analog of the Paul quadrupole mass filter. To see how it works we first note that in a (homogeneous) sinusoidal rf electric field, ion motion is sinusoidal but is 180 degrees out of phase with respect to the electric force. If the field is somewhat inhomogeneous, it is easy to show that the force on the ion averaged over one cycle of the driven motion is towards the region of weaker field. [1] Since an electric field minimum can exist in a charge free region, stable trapping can be accomplished. Such a trap is shown schematically in Fig. 1 where the three trap electrodes are shaped to provide an electric potential of the form (r^2-2z^2) inside the trap. For this "ideal" trap shape, an ion is bound in a nearly harmonic well.

The "ideal" Penning [10] trap uses the same electrode configuration as in Fig. 1 but uses static electric and magnetic fields. A harmonic potential well is provided along the "z" axis by static electric fields. This however results in a radial electric field which forces the ions towards the "ring" electrode. This effect can be overcome if a static magnetic field $\vec{B}$ is superimposed along the "z" axis. In this case the x - y motion of the ions is a composite of circular cyclotron orbits (primarily due to the $\vec{B}$ field) and a circular $\vec{E} \times \vec{B}$ drift ("magnetron" motion) about the trap axis.

FREQUENCY STANDARDS WITHOUT LASER COOLING

Several groups have sought to develop a microwave frequency standard based on the 40.5 GHz hyperfine splitting in $^{199}Hg^+$ ions stored in an rf trap. [11-15] The relatively small size of this device could make it a portable standard with potential commercial applications. The choice of the $^{199}Hg^+$ ion for a microwave frequency standard is based on its 40.5 GHz ground-state hyperfine separation, which is the largest of any ion which might easily be used in a frequency standard (hence high Q for given interrogation time), and its relatively large mass (hence small second order Doppler shift for a given temperature). In addition, a $^{202}Hg^+$ lamp source can be be used to optically pump the $^{199}Hg^+$ ground state. A fractional frequency stability comparable to that of commercial cesium

standards has been demonstrated. [13] In these experiments, the second order Doppler shift can be reduced by cooling the ions with a light neutral buffer gas (e.g. helium or hydrogen). With buffer gas pressures up to 10^{-2} Pa the secular motion of the ions in the psuedopotential well can be thermalized to the ambient temperature.[14] For Hg^+ at room temperature, the second order Doppler shift is about 2×10^{-13}. Unfortunately, the second order Doppler shift due to the micromotion of the ions can be much larger. [1,14] The size of the micromotion contribution to the 2nd order Doppler shift depends on the size of the ion cloud, or, for a given ion number density, on the total number of ions. Consequently in the performance of the $^{199}Hg^+$ frequency standard there is a tradeoff between systematic errors due to the 2nd order Doppler shift and signal-to-noise ratio. For a cloud of $\sim 10^6$ ions an accuracy of 2×10^{-13} and fractional frequency stability of $\sigma_y(\tau) \cong 2 \times 10^{-12} \tau^{-1/2}$ appear accessible.[14] This would be about an order of magnitude improvement in accuracy and stability over commercially available cesium frequency standards.

In addition, optical microwave double resonance experiments on stored ions have been performed using tunable lasers as light sources. The ground-state hyperfine splittings of $^{137}Ba^+$, [16] $^{135}Ba^+$, [17] and $^{171}Yb^+$ [18] have been measured, using pulsed dye lasers and rf traps. Microwave resonances as narrow as 60 mHz were observed in $^{171}Yb^+$. This has a line Q of 2×10^{11}. In some cases, optical pumping out of the absorbing ground state prevents use of the double-resonance method. This problem may be overcome, however, with the use of collisional relaxation [16,19].

FREQUENCY STANDARDS WITH LASER COOLING

A fundamental limitation of the above ion trap experiments is the 2nd order Doppler shift. In 1975 proposals [20,21] were made which could further reduce the second order Doppler shift by a process called laser cooling (also called optical sideband cooling or radiation pressure cooling). Laser cooling is a method by which a beam of light can be used to damp the velocity of an atom or ion. The basic mechanism for cooling of a trapped ion by a laser beam tuned slightly lower in frequency than a strongly allowed resonance transition is as follows: when the velocity of the ion is directed against the laser beam, the light frequency in the ion's frame is Doppler shifted closer to resonance so that the light scattering takes place at a higher rate than when the velocity is along the laser beam. Since the photons are reemitted in random directions, the net effect, over a motional cycle, is to damp the ion's velocity, due to absorption of photon momentum. If the laser frequency is tuned above resonance, it causes heating. In certain cases laser cooling can reduce the ion temperature below 1 K. Because of rf heating, it may be more difficult to do significant laser cooling on a cloud of many ions in an rf trap than in a Penning trap. [2] Consequently laser cooling experiments with a cloud of many ions have primarily been done in Penning traps.

Laser cooling of Mg^+ [4,22-24] and Be^+ [25,26] ions in a Penning trap has been achieved. For both types of ions, the light sources were the second harmonics, generated in nonlinear crystals, of cw dye lasers. The ions were optically detected by monitoring the cooling laser light scattered by the ions. Because the photon scatter rates can be very large ($> 10^6$ s^{-1} per ion), the optical detection provides a very sensitive detection technique where the noise in the system can be limited to the statistical fluctuations in the number of ions that made the clock transition. [27]

As a step towards realizing a frequency standard based on laser cooled stored ions, a clock based on a hyperfine transition in $^9Be^+$ has been constructed [26]. The average frequency of an rf oscillator was locked to the $(M_I, M_J) = (-3/2, 1/2)$ to $(-1/2, 1/2)$ nuclear spin flip transition in the ground state of $^9Be^+$, near the magnetic field (0.8194 T) at which the first derivative of the frequency with respect to field goes to zero. The ions were cooled to less than 2K. The 303 MHz resonance was observed with 25 mHz linewidth by rf-optical double resonance (see Fig. 2). The frequency stability of the locked oscillator ($\sigma_y(\tau) \equiv 2 \times 10^{-11} \tau^{-1/2}$) was comparable to that of commercial Cs atomic beam frequency standards. The frequency accuracy was on the order of 10^{-13}, limited primarily by the uncertainty of the second-order Doppler shift due to heating of the ions during the rf resonance period, when the cooling radiation was shut off in order to avoid light shifts. At the end of the 20 s Ramsey interrogation period, the ion temperature had increased to ~ 30 K. The dominant heating mechanism may be due to axial asymmetries in the trap. [28,29] Reduction of the heating (and consequently the second order Doppler shift) by an order of magnitude should be possible by constructing a trap with better axial symmetry or by the use of a second type of ion (e.g. $^{24}Mg^+$) to "sympathetically" cool the $^9Be^+$ ions. [22,23] Primary cesium standards are slightly better than this first frequency standard based on a laser cooled ion, but future improvements with the $^9Be^+$ standard are anticipated.

Because $^9Be^+$ is experimentally easy to cool with a laser, it was used to investigate the generic problems of a laser cooled stored ion frequency standard. As a microwave frequency standard, $^9Be^+$ is limited because of the low 303 MHz frequency of the clock transition. Clock transition linewidths are probably independent of the species of the trapped ion used. Therefore an ion with as high a clock transition frequency as possible should be used in order to increase the line Q and reduce the measurement imprecision. For this reason a better ion for a laser cooled microwave clock is Hg^+. Unfortunately laser cooling is much harder to achieve with Hg^+ than with Be^+ (partly because the 194 nm cooling radiation is difficult to produce), and has not yet been demonstrated. A proposal for a frequency standard based on a 25.9 GHz magnetic field independent transition in $^{201}Hg^+$ has the potential of achieving absolute accuracies of better than one part in 10^{15} and frequency stabilities of less than 10^{-16}. [27]

OPTICAL FREQUENCY STANDARDS

In order to increase the Q even further, one could go to a much higher frequency; for example, use a narrow optical transition. The anticipated Q in this case can be extremely high, 10^{15} or more. A number of transitions in various ions have been proposed [2]; Dehmelt [30] was the first to suggest that such extremely high resolution spectroscopy could be carried out using one photon transitions in, for example, single group IIIA ions. For instance the $^1S_0 \leftrightarrow {}^3P_0$ transition in $T\ell^+$ (λ = 202 nm) has as $Q \cong 5 \times 10^{14}$. [30] For such optical one photon transitions, it is desirable to approximately satisfy the Dicke criterion; this is most easily accomplished with single trapped ions [2,30]. Others [31] have proposed using Doppler free two photon transitions, for example the $^2S_{1/2} \rightarrow {}^2D_{5/2}$ transition in Hg^+ (λ = 563 nm, $Q \cong 7 \times 10^{14}$). Optical two photon transitions using equal frequency photons have the potential of completely eliminating the first order Doppler effect for a cloud of many ions where it is impossible to satisfy the Dicke criterion. They ultimately have the disadvantage that the rather large optical fields necessary to drive the transition cause undesirable ac Stark shifts [27,31].

The projected accuracy for optical frequency standards using single ions is extremely high. Second order Doppler shifts of 10^{-19} or lower are possible. [2] Other systematic shifts can occur [1,2,7,27,30,31] but it is possible that they can be controllable to this level. These extreme accuracies make important the problem of measurement imprecision since the signal-to-noise ratio on a single ion will at best be about one for each measurement cycle. Practically speaking, this means that a long averaging time will be required to reach a measurement precision equal to these accuracies. In fact, for a while, the accuracy and resolution may be limited by laser linewidth characteristics (linewidth and linewidth symmetry). However, the potential for extremely narrow lasers also exists [32].

Unfortunately, to use such laser devices as clocks one must count cycles of the radiation, that is, measure its phase. At microwave frequencies this is straightforward. At optical frequencies it is technically feasible but very hard [33]. In any case, the potential accuracy for stored ion spectroscopy in all spectral regions seems extremely high. Frequency standards and clocks with inaccuracy of one part in 10^{15} appear very reasonable, eventually they could be orders of magnitude better than this.

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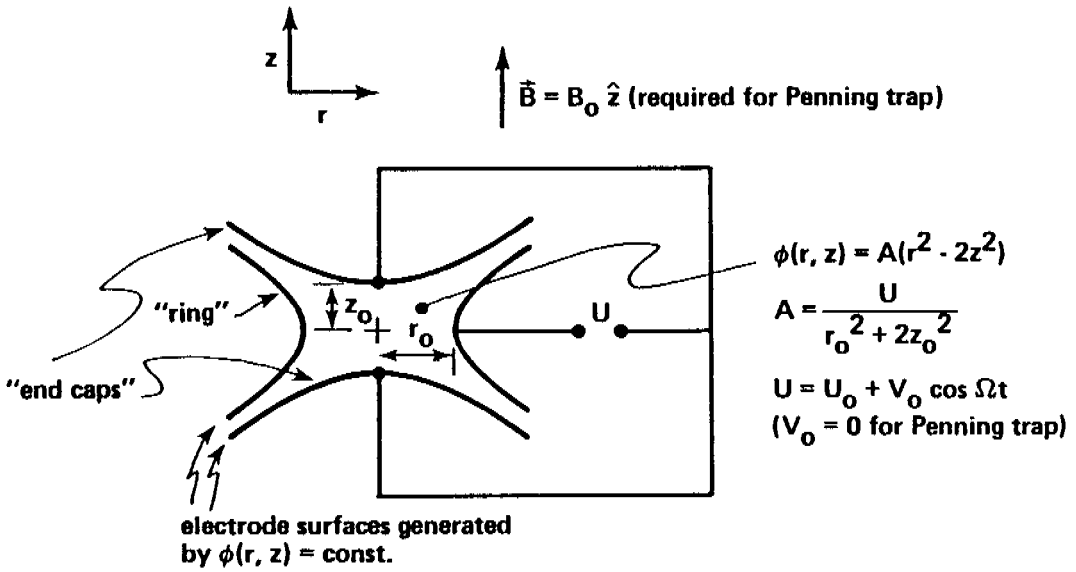

FIG. 1 Schematic representation of the electrode configuration for the "ideal" Paul (rf) or Penning trap. Electrode surfaces are figures of revolution about the z axis and are equipotentials of $\phi(r,z) = A(r^2-2z^2)$. (Cylindrical coordinates are used with the origin at the center of the trap.) Typical dimensions are $\sqrt{2}\, z_0 = r_0 \cong 1$ cm. Typical operating parameters are: for the Paul trap, $V_0 = 300$ V/cm, $\Omega/2\pi \cong 1$ MHz; for the Penning trap, $U_0 \cong 1$V, $B \cong 1$T.

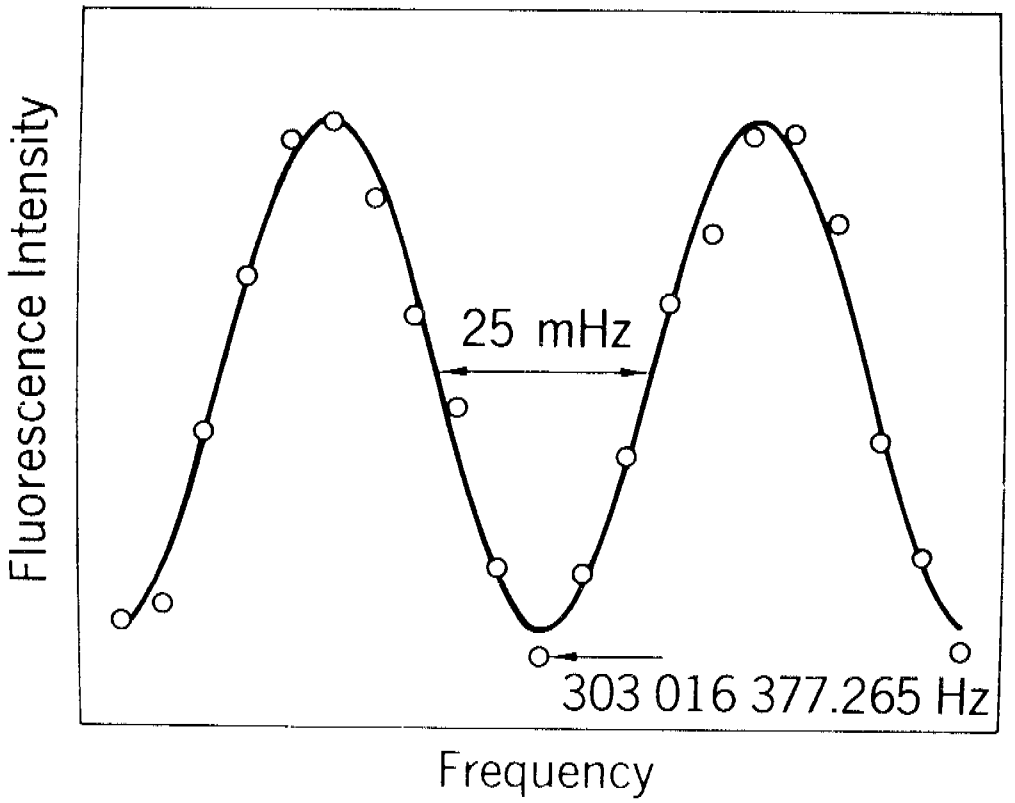

FIG. 2. Signal obtained with two 0.5s Ramsey pulses separated by a 19 s free precision interval on the clock transition in $^{9}Be^{+}$ (see text). The sweep width was 100 mHz and the frequency interval between points was 5 mHz. The dots are experimental and are the average of 10 sweeps; the curve is a least squares fit.

QUESTIONS AND ANSWERS

MR. HELLWIG: What is the difference between mercury 199 and mercury 201? It has to do with the F numbers, right?

MR. BOLLINGER: Since we wanted to do an experiment in the Penning trap, because the laser cooling appears easier, we have to find a field independent transition at a large magnetic field, and one exists in mercury 201 at around 29.5 GHz. That's the reason the proposal is made for mercury 201 as opposed to mercury 199.