

# LASER COOLED ${}^9\text{Be}^+$ ACCURATE CLOCK

J. J. Bollinger, Wayne M. Itano, and D. J. Wineland  
 Time and Frequency Division  
 National Bureau of Standards  
 Boulder, Colorado 80303

## Summary

The use of laser cooled stored ions in an atomic frequency standard has the potential of very high accuracy because Doppler effects are greatly suppressed. A clock based on the ground-state hyperfine transition in  ${}^{201}\text{Hg}^+$  has potential accuracy and stability exceeding 1 part in  $10^{15}$ . However, laser cooled  ${}^9\text{Be}^+$  ions are experimentally easier to obtain. Therefore a  ${}^9\text{Be}^+$  based frequency standard is investigated in order to study the generic problems of laser cooled stored ion frequency standards. Approximately 300  ${}^9\text{Be}^+$  ions are stored in a Penning trap and laser cooled. The 303 MHz ground state  $(M_I, M_J) = (-3/2, 1/2) \rightarrow (-1/2, 1/2)$  nuclear spin flip hyperfine transition is observed at a magnetic field ( $\sim 0.82\text{T}$ ) where the transition frequency is independent of magnetic field to first order. The time domain Ramsey method of interrogation is used and yields a linewidth of 25 MHz. The stability of an oscillator locked to this transition has been measured for  $400\text{s} < \tau < 3200\text{s}$  to be  $\sigma(\tau) \approx 2 \times 10^{-11} \tau^{-2}$ . By measuring the velocity distribution of the ions, the second-order Doppler shift is determined to be on the order of  $5 \times 10^{-14}$ . The magnetic field instability contributes a  $3 \times 10^{-14}$  uncertainty in the present experiment. All other systematic uncertainties are estimated to be no greater than  $3 \times 10^{-14}$ .

## Introduction

Because stored ion techniques provide long confinement times with minimal perturbations, they provide the basis for improved time and frequency standards<sup>1</sup>. Several groups have sought to develop a microwave frequency standard based on  ${}^{199}\text{Hg}^+$  ions in an rf trap<sup>2-5</sup>. The choice of the  ${}^{199}\text{Hg}^+$  ion for a microwave stored ion 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 a given interrogation time), and its relatively large mass, (hence small second-order Doppler shift at a given temperature). In addition, a  ${}^{202}\text{Hg}$  lamp

source can be used to optically pump the  ${}^{199}\text{Hg}^+$  ground state. Experiments on Hg<sup>+</sup> ions to date have been unsuccessful in cooling the ions to the ambient room temperature. This results in a relatively large second-order Doppler or time-dilation shift which may be difficult to evaluate to better than about  $10^{-13}$ . NBS has proposed<sup>6</sup> an ion frequency standard based on a ground-state hyperfine transition in  ${}^{201}\text{Hg}^+$  ions stored in a Penning trap and cooled to below 1K by laser radiation pressure. The laser cooling of the Hg<sup>+</sup> ions is made difficult by the requirement for a 194 nm tunable, narrowband source and by depopulation optical pumping of the ground state. Laser cooling of  ${}^9\text{Be}^+$  ions is experimentally much easier. Therefore it is useful to investigate a  ${}^9\text{Be}^+$  based frequency standard in order to study the generic problems of stored ion frequency standards, even though the potential performance is not as high as for Hg<sup>+</sup> ions, due to the lower transition frequency.

## Experimental Method

Clouds of  ${}^9\text{Be}^+$  ions are confined by the static magnetic and electric fields of a Penning trap and stored for hours (see Fig. 1). The ions are created inside the trap by electron bombardment of Be atoms evaporated from an oven located outside the trap electrodes. During a run, the oven is turned off, and the residual pressure is estimated to be less than  $10^{-7}$  Pa. The trap is made of gold mesh endcaps and a molybdenum mesh ring electrode. The center of the trap is at one focus of an ellipsoidal mirror; the second focus is outside the vacuum system. A lens is used to collimate the fluorescence light into a photomultiplier tube. The ions are laser cooled, compressed, and pumped into the  $(M_I, M_J) = (-3/2, -1/2)$  ground state by a laser tuned to the  $2s^2S_{1/2}(-3/2, -1/2) \rightarrow 2p^2P_{3/2}(-3/2, -3/2)$  ( $\lambda = 313\text{nm}$ ) transition.<sup>7</sup> The 313 nm light is obtained by generating the second harmonic of the output of a single mode

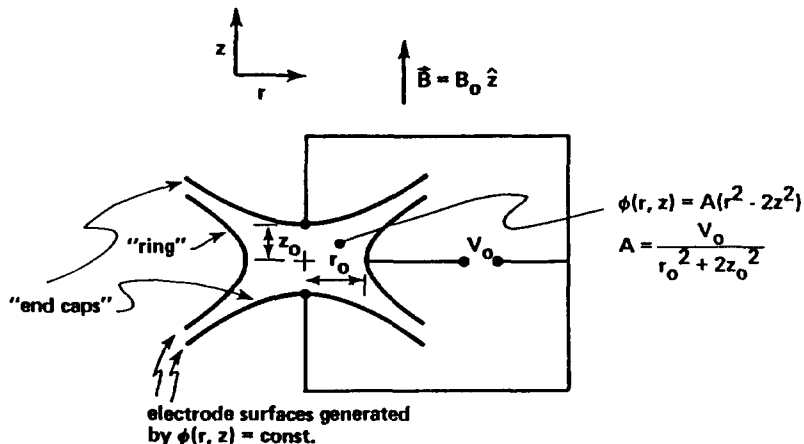


Figure 1. Electrode configuration for a Penning trap. In this experiment  $B_0 = 0.8194\text{T}$ ,  $1.64z_0 = r_0 = 0.417\text{cm}$ , and  $V_0 = 1\text{V}$ .

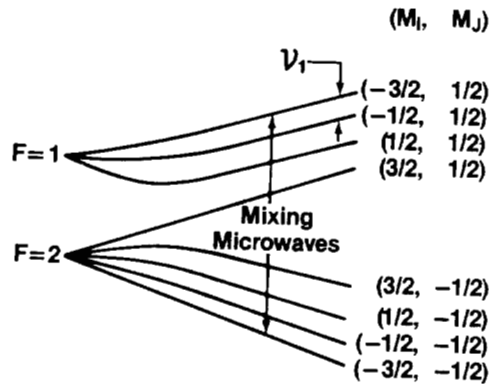


Figure 2. Hyperfine structure of the  ${}^9\text{Be}^+ 2s^2S_{1/2}$  ground state as a function of magnetic field.  $\nu_1$  is the clock transition.

cw dye laser in a  $90^\circ$  phase-matched crystal of rubidium dihydrogen phosphate (RDP). The power is typically  $20 \mu\text{W}$ . The size, density, and temperature of the ion clouds are determined by using a second probe laser.<sup>8</sup> Typical clouds consist of at most several hundred ions with cloud densities of  $1\text{-}2 \times 10^7$  ions/cm<sup>3</sup> and cloud diameters ranging from 100 to 300  $\mu\text{m}$ . Ion temperatures of around 100 mK are obtained with the laser on continuously.

At a magnetic field of about 0.8194 T, the  $(-3/2, 1/2) \rightarrow (-3/2, -1/2)$  ground state hyperfine transition  $\nu_1$  (see Fig. 2), depends only quadratically on the magnetic field deviation  $\Delta B$ .

$$\frac{\Delta\nu_1}{\nu_1} = -0.017 \left( \frac{\Delta B}{B} \right)^2$$

Linewidths which, to a high degree, are independent of the magnetic field homogeneity and stability are obtained by using a first order magnetic field independent transition like  $\nu_1$  as the clock transition.<sup>9</sup> Microwave radiation (tuned to the electron spin flip resonance ( $\sim 23\,914.01$  MHz) transfers half of the ion population from the optically pumped  $(-3/2, -1/2)$  state to the  $(-3/2, +1/2)$  state. Some of the  $(-3/2, +1/2)$  state population is transferred to the  $(-1/2, +1/2)$  state by application of rf near the 303 MHz clock transition frequency. This results in a decrease in the optically pumped  $(-3/2, -1/2)$  state population because of the microwave mixing and a decrease in the fluorescence light detected by the photomultiplier tube.

The time domain Ramsey method is used to probe the  $\nu_1$  clock transition. First a 0.5 s rf pulse is applied to the trapped ions. This is followed by a 19 s free precession period and then the second 0.5 s rf pulse, coherent with the first one. The laser and mixing microwaves are on for a period of 3 to 5 s during which the  ${}^9\text{Be}^+$  ions are prepared in the  $(-3/2, -1/2)$  and  $(-3/2, +1/2)$  states. The laser and mixing microwaves are then turned off during the 20 s rf Ramsey interrogation period in order to avoid light and ac Zeeman shifts. After the Ramsey interrogation period, the laser and mixing microwaves are turned back on, and the signal is obtained from the photomultiplier tube count rate during the first 0.3 to 0.5 s of the laser and mixing microwaves on period. The theoretical line shape for these conditions is shown in Fig. 3. Figure 4 gives the signal obtained by averaging 10 sweeps across a 100 MHz width centered near the clock transition

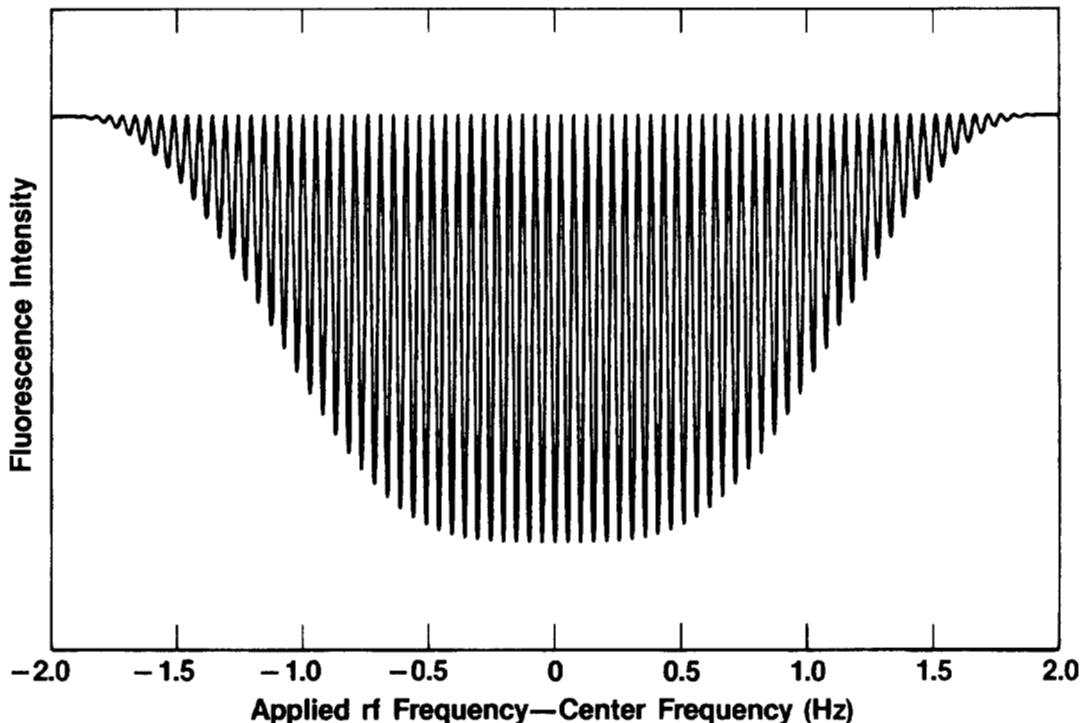


Figure 3. Theoretical resonance curve for the clock transition.

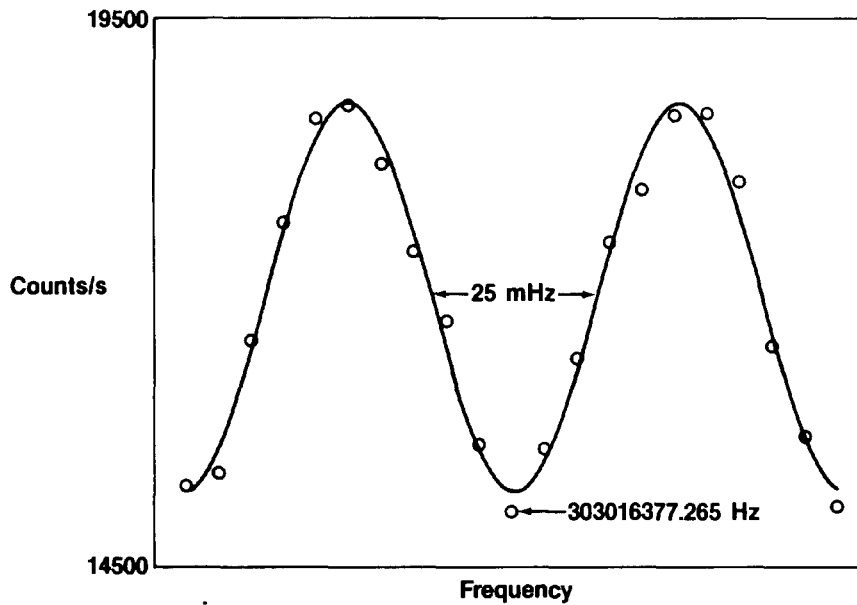


Figure 4. Ramsey signal obtained on the clock transition. The sweep width is 100 MHz and the frequency interval between points is 5 MHz. The dots are experimental; the curve is a least squares fit.

frequency (i.e., the central portion of Fig. 3). The 25 MHz linewidth gives a Q of  $1.2 \times 10^{10}$  on the 303 MHz clock transition.

Figure 5 is a block diagram of the system used to lock an rf oscillator to the clock transition. The 303 MHz is obtained by sum-mixing the 294 MHz frequency doubled output of a frequency synthesizer (SYNTH 1) with the output of another frequency synthesizer (SYNTH 2) near 9 MHz. A 5 MHz voltage controlled crystal oscillator (VCXO) is used as a reference for SYNTH 2. A passive hydrogen maser ( $\sigma_y(\tau) = 1.5 \times 10^{-12} \tau^{-1/2}$ , frequency drift  $< 3 \times 10^{-16}/\text{day}$ )<sup>10</sup> is used as a reference for SYNTH 1. SYNTH 2, the counter, the laser light shutter, and the microwave and rf switches are interfaced to a computer which controls the data acquisition sequence. The computer steps SYNTH 2 by  $\pm 13$  MHz about a frequency near the 303 MHz clock transition frequency minus 294 MHz. If  $C_1$ ,  $C_2$ , and  $C_3$  are three successive signals, the computer calculates an error signal equal to  $C_1 + C_3 - 2C_2$ .<sup>4,5</sup> This error signal is independent of a linear drift in the total count rate. The computer simulates an analog integrator by incrementing a register by a number proportional to the error signal after each measurement. The contents of this register are output to a digital-to-analog converter (D/A), which steers the VCXO so as to keep the mean interrogation frequency (the average of the high and low frequency half-cycles) as close as possible to the clock transition frequency. The frequency of the VCXO is compared with the frequency of the passive hydrogen maser.

### Results

The Allan variance of the VCXO was measured out to  $3.2 \times 10^3$  s. The VCXO is used as a reference for SYNTH 2 which provides only 9 MHz of the 303 MHz rf. By dividing the Allan variance of the VCXO by the ratio of the clock transition and SYNTH 2 frequencies (equal to 33.6), the Allan variance,  $\sigma_y(\tau)$ , of an oscillator stabilized by the entire 303 MHz clock transition is obtained, since frequency

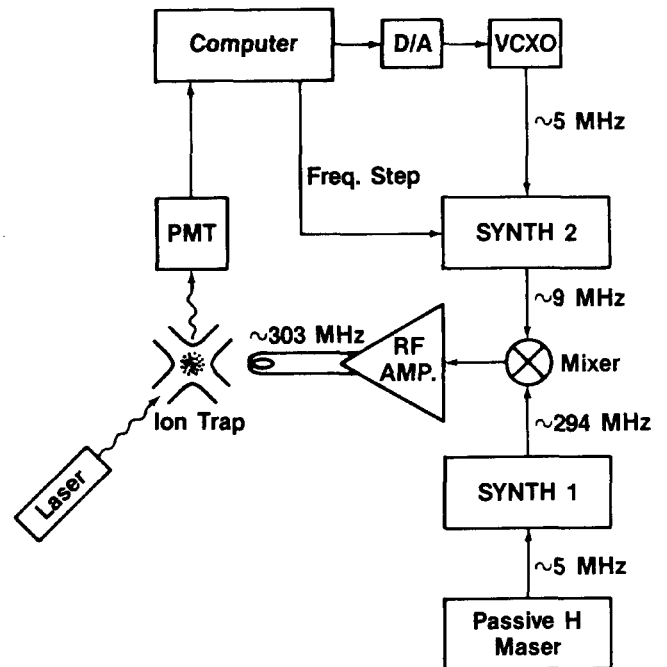


Figure 5. Block diagram of the frequency servo.

fluctuations in the frequency doubled 294 MHz output of SYNTH 1 are negligible. Figure 6 shows  $\sigma_y(\tau)$  for six runs of this experiment. The attack time is between 300 and 400 s. For times greater than the attack time,  $\sigma_y(\tau)$  is falling off as  $2 \times 10^{-11} \tau^{-1/2}$ . A measurement of the  $\nu_1$  clock transition frequency is obtained from each run. A weighted average frequency,  $\bar{f}$ , is obtained for fifteen 6400 s runs by weighting each run according to its stability  $\sigma_y(\tau)$  at  $\tau=1600$  s.

$$\bar{f} = \frac{\sum (f_i / \sigma_i^2)}{\sum (1 / \sigma_i^2)}$$

$$= 303\,016\,377.265\,077 \text{ Hz}$$

The standard deviation of this average is

$$\sigma = \left[ \frac{\sum (f_i - \bar{f})^2 / \sigma_i^2}{(N-1) \sum (1 / \sigma_i^2)} \right]^{1/2}$$

$$= 44 \text{ } \mu\text{Hz} (1.5 \times 10^{-13}).$$

As noted above, the frequency of the stabilized VCXO was measured relative to the passive hydrogen maser. The uncertainty of the measured frequency of the passive hydrogen maser relative to the international definition of the second, based on the Cs hyperfine separation, is 64  $\mu\text{Hz}$  ( $2.1 \times 10^{-13}$ ).

Various experimental parameters were varied to test for possible systematic errors. The mixing microwave power was varied by over 20 dB. In the initial stages of this experiment, a  $1 \times 10^{-12}$  shift in the measured clock transition frequency was observed when the mixing microwave power was lowered by 20 dB. It was determined that this shift was

related to the small rf leakage during the repumping (laser, mixing microwaves on) part of the data cycle. By switching the SYNTH 2 frequency by +1 kHz during the repumping part of the data cycle, this shift was made to disappear. Other parameters which were varied in order to test for systematic frequency shifts were the laser power and the number of ions, each of which was varied by about a factor of two, and various dead times which were inserted between parts of the data cycle. In order to test for frequency offsets due, for example, to an asymmetric component of the resonance line, the frequency steps were changed to  $\pm 0.47$  Hz, so that the sides of the ninth sidelobes, rather than of the central lobe, were sampled (See Fig. 3). No systematic shifts were observed at the level permitted by the signal-to-noise ratio (a few parts in  $10^{13}$ ).

Table 1 lists the estimated systematic errors for this experiment. The 3 parts in  $10^6$  peak-to-peak fluctuations in the magnetic field contribute a  $3 \times 10^{-14}$  uncertainty in the clock transition frequency. State of the art superconducting magnets have stabilities 1,000 times better than the magnet used in this experiment, which could lower this uncertainty by a factor of  $10^6$ . Nonlinearities and frequency drift in the VCXO could cause servo offsets at the  $3 \times 10^{-14}$  level. By improving the servo system (for instance, linearizing the VCXO response) this uncertainty could be substantially reduced. The temperature of the ions with the laser on was measured to be approximately 100 mK which corresponds to a second-order Doppler shift of  $1.5 \times 10^{-15}$ . With the laser off, some heating of the ions was observed. During the 20 s rf Ramsey period while the laser was off, the ion temperature increased to about 3 K. This results in a second-order Doppler shift of about 5 parts in  $10^{14}$ . We believe that the heating observed in this experiment can be

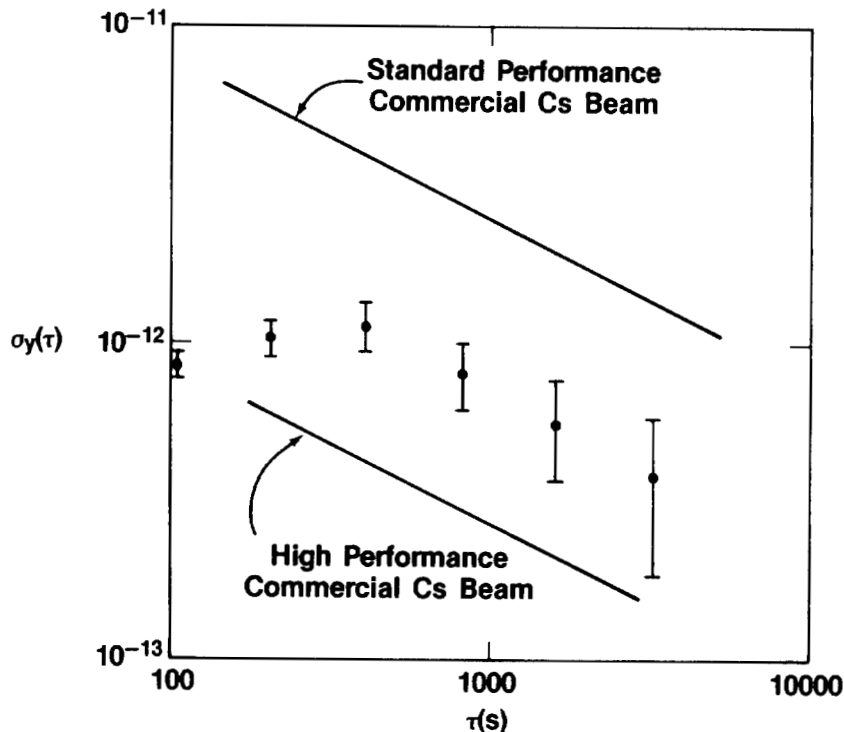


Figure 6.  $\sigma_y(\tau)$  in this experiment compared with standard and high performance commercial Cs beam tubes.

TABLE I. Estimated systematic errors

| Systematic effect               | Size of effect            | Uncertainty         |
|---------------------------------|---------------------------|---------------------|
| Magnetic field freq. shift      | $\nu_1 = 303 \text{ MHz}$ | $3 \times 10^{-14}$ |
| Servo offsets                   | $3 \times 10^{-14}$       | $3 \times 10^{-14}$ |
| 2nd order Doppler               | $5 \times 10^{-14}$       | $5 \times 10^{-14}$ |
| Pulling due to spurious signals | $<10^{-15}$               |                     |
| Stark shifts                    | $<10^{-15}$               |                     |
| 1st order Doppler               | $<10^{-15}$               |                     |
| Collisions with background gas  | $<10^{-15}$               |                     |
| Background slopes               | $<10^{-15}$               |                     |
| Coherence between cycles        | $<10^{-15}$               |                     |

understood and controlled in future experiments, and that second-order Doppler shifts at the  $1 \times 10^{-15}$  level will be obtained. A potential systematic effect for stored ion frequency standards is a light shift due to an incomplete reinitialization of the ion population during the repumping part of the data cycle<sup>11</sup>. Such a shift may be present even though the light is off during the rf interrogation period, if coherence survives the repumping period. In this experiment, the computer random function generator was used to randomize the phase of the rf before each rf Ramsey period. This eliminates any systematic light shift. All other systematic effects listed in Table I are less than  $1 \times 10^{-15}$ . Unfortunately, the signal-to-noise ratio in this experiment did not permit reaching the level of the anticipated systematics of Table I in a reasonable amount of time.

#### Future Work

As stated in the introduction, the purpose of this experiment was to study the generic problems of stored ion frequency standards with cold ions. Ultimately, the same experiment could be performed on the 26 GHz field independent ground state hyperfine transition in  $^{201}\text{Hg}$ .<sup>6</sup> Based on previous work with  $\text{Mg}^+$  ions<sup>7</sup>, 10 mHz linewidths should be obtainable. This would give a line Q of  $2.6 \times 10^{12}$ . In future experiments, we also expect to load clouds with as many as  $10^5$  ions.<sup>12</sup> From the results obtained in this experiment on small clouds and a 303 MHz clock transition frequency, we anticipate that a stability of  $2 \times 10^{-15} \tau$  should be obtainable with a frequency standard based on large clouds of  $^{201}\text{Hg}$  ions.<sup>6</sup> The uncertainty of the anticipated systematic effects should be comparable to those discussed in the previous section; thus an accuracy on the order of  $1 \times 10^{-15}$  is expected. The development of the cw, tunable, narrowband source at 194 nm required for cooling the  $\text{Hg}^+$  ions has now been accomplished.<sup>13</sup> Experiments on trapping and cooling  $\text{Hg}^+$  ions are being initiated.

#### Acknowledgments

We wish to thank the Air Force Office of Scientific Research and the Office of Naval Research for their support of this work. We would also like to thank S. R. Stein for his aid in using the time measurement system and for many helpful discussions, C. Manney for his work in designing the rf probe, and J. S. Wells and R. Blatt for carefully reading the manuscript.

#### References

1. H. G. Dehmelt, *Advan. Atomic and Mol. Physics* **3**, 53 (1967) and **5**, 109 (1969). D. J. Wineland, W. M. Itano, and R. S. Van Dyck, Jr., *Advan. Atomic and Mol. Physics* **19**, to be published.
2. F. G. Major and G. Werth, *Phys. Rev. Lett.* **30**, 1155 (1973).
3. M. D. McGuire, R. Petsch, and G. Werth, *Phys. Rev.* **A17**, 1999 (1978).
4. M. Jardino, M. Desaintfuscién, R. Barillet, J. Viennet, P. Petit, and C. Audoin, *Proc. 34th Ann. Symp. on Freq. Control*, 1980 (Electronic Industries Assoc., 2001 Eye St., NW, Washington D.C. 20006) p. 353 and *Appl. Phys.* **24**, 107 (1981).
5. L. S. Cutler, R. P. Giffard, and M. D. McGuire, *Proc. 13th Annual Precise Time and Time Interval Applications and Planning Meeting*, Washington, D.C. 1981 (NASA Conf. Publ. 2220, NASA Scientific and Tech. Info. Branch, 1982) p. 563 and paper in these proceedings.
6. D. J. Wineland, W. M. Itano, J. C. Bergquist and F. L. Walls, *Proc. 35th Ann. Symp. on Freq. Control*, 1981 (Electronic Industries Assoc., 2001 Eye St., NW, Washington D.C. 20006) p. 602.
7. W. M. Itano and D. J. Wineland, *Phys. Rev.* **A24**, 1364 (1981).
8. J. J. Bollinger and D. J. Wineland, *Bull. Am. Phys. Soc.* **28**, 782 (1983).
9. P. Kusch and H. Taub, *Phys. Rev.* **75**, 1477 (1949).
10. Private communication with F. L. Walls, Time and Freq. Div., NBS.
11. E. I. Alekseyev, Ye. N. Bazarov, and G. I. Telegin, *Radio Eng. Electron. Phys. (USSR)* **20**, 73 (1975).
12. Only small clouds of  $^9\text{Be}^+$  ions were used in this experiment because the supply of  $^9\text{Be}$  in the oven was almost exhausted.
13. H. Hemmati, J. C. Bergquist, and W. M. Itano, *Opt. Lett.* **8**, 73 (1983).