## Annual Report for work on "Cooled Ion Frequency Standard"

**PERFORMING ORGANIZATION NAME AND ADDRESS**
National Bureau of Standards
Time and Frequency Division 524
Boulder, Colorado 80303

**CONTRIBUTING OFFICE NAME AND ADDRESS**
Office of Naval Research
Physics Program Office. 800 N. Quincy
Arlington, VA 22217

### ABSTRACT
The purpose of this work is to develop techniques to overcome the fundamental limits of present frequency standards—the second and residual first-order Doppler shifts. To this end we study suitable frequency reference transitions in ions which are stored on electromagnetic traps and cooled by radiation pressure to < 1K.
Summary of Work on

"COOLED ION FREQUENCY STANDARD"

(FY 86)

submitted to

Office of Naval Research

ONR Contract No. N00014-85-F-0004

Project Leader:

D.J. Wineland

Frequency & Time Standards Group

524.11

National Bureau of Standards

Boulder, Colorado 80303

FTS: 320-5286

(303) 497-5286

Approved for public release; distribution unlimited.

Reproduction in whole or in part is permitted for any purpose of the United States Government
Contract Description

The purpose of this work is to develop techniques to overcome the fundamental limits of present methods for high resolution spectroscopy and frequency standards—the second and residual first-order Doppler shifts. To this end, we study suitable frequency reference transitions in ions which are stored in electromagnetic traps and cooled by radiation pressure to < 1K.

Scientific Problem

The scientific problems are (1) to suppress second order and residual first order Doppler shifts in atomic frequency standards in a fundamental way—by substantially reducing the kinetic energy of ions stored in electro-magnetic traps, (2) to study suitable reference transitions in ions that can be used as frequency standards, and (3) to study the problems generic to all stored ion frequency standards. The goal is to achieve at least a factor of 100 improvement in accuracy over the present best device, the Cesium beam frequency standard, which has an accuracy of approximately one part in $10^{13}$.

Scientific and Technical Approach

Laser cooling is employed on all experiments in order to suppress Doppler shifts. Temperatures < 0.1K are routinely achieved. To avoid light shifts on "clock" transitions we investigate "sympathetic cooling" where one ion species is laser cooled and by Coulomb collisions cools another ion species of spectroscopic interest. We will continue experiments on Mg$^+$ and Be$^+$ in order to study generic problems with traps. We are developing a separate experiment for Mg$^+$ ions. These experiments have the goal of realizing a frequency standard with $10^{-15}$ or better accuracy.
PROGRESS DURING LAST CONTRACT PERIOD

A. EXPERIMENTAL

1. Sympathetic cooling.

For the experiments on rf and microwave spectroscopy, especially for an ion microwave frequency standard, we think a significant advance can be made by use of a process called sympathetic cooling. In sympathetic cooling, two ion species are stored in the trap simultaneously. (Say $^9\text{Be}^+$ and $^{201}\text{Hg}^+$). One species which is easily laser cooled ($^9\text{Be}^+$, because of the high laser powers easily generated) sympathetically cools the other ion species via Coulomb collisions. This technique has two important advantages over the direct laser cooling method (employed in our first $^9\text{Be}^+$ clock experiment): (1) Laser cooling can be supplied by a laser which is easy to make -- like the one for $^9\text{Be}^+$. In the $\text{Hg}^+$ experiments we have determined that it will be very difficult to obtain much more 194 nm power for direct cooling -- the main limit is imperfections in the nonlinear crystals which causes thermal "blooming" of the beams. (2) Since the cooling laser will in general be non resonant with the energy levels of the "clock" ion, a.c. stark shifts from the cooling laser can be negligible ($<10^{-10}$). Therefore, the cooling laser can be left on continuously and the reheating encountered in the direct laser cooling experiments can be avoided. In addition, interrogation times of clock transitions can be made much longer -- linewidths of less than 1 millihertz do not seem unreasonable (interrogation times of greater than 500 s). Moreover, the temperature of the ions and therefore the second order Doppler shift will be much smaller and much more stable in this steady state situation and resulting frequency shifts should be easier to characterize than in the transient condition of the $^9\text{Be}^+$ clock.

We have recently realized this technique using $^9\text{Be}^+$ and $^{198}\text{Hg}^+$ ions. The results are that the basic technique is valid; by laser cooling $^9\text{Be}^+$ ions, the $^{198}\text{Hg}^+$ ions were cooled to a temperature less than 1K. In addition, the shapes of the resulting two species plasma were measured and found to compare fairly well with theory. One interesting aspect of such plasma is that the
two species separate due to the differences in centrifugal forces as indicated in Figure 1.

![Diagram showing two species separation](image)

Be$^+$ alone  \[ \rightarrow \]  Hg$^+$ + Be$^+$

(a)  \[ \rightarrow \]  (b)  \[ \rightarrow \]  (c)

Fig. 1. Experimental and theoretical shapes of ion plasmas. (a): $^8$Be$^+$ ion plasma alone. (b): Same $^8$Be$^+$ ions as in (a), but with $^{198}$Hg$^+$ ions added. (c): Theoretical shape generated for the same experimental conditions as in (b).

2. Single $^{198}$Hg$^+$ ion laser cooling.

We have stored single $^{198}$Hg$^+$ ions in the rf trap shown in Fig. 2.

![Diagram showing rf trap](image)

Fig. 2. Schematic drawing showing cross section view of rf trap electrodes for $^{198}$Hg$^+$. The electrodes are figures of revolution about the z-axis and are made from molybdenum.
Typical operating conditions were an oscillating voltage of about 500 V applied between ring and endcaps at 20 MHz and a static potential of about 10 V between ring and endcaps in order to give an overall approximately spherical well. The 194 nm fluorescence light (see Fig. 3) scattered from the ion was imaged onto a plate about 20 cm away from the trap.

![Diagram](image)

**Fig. 3.** $^{198}$Hg$^+$ level structure showing the 194 nm cooling transition and the $^3D_{5/2}$ level which can be excited via the 2 photon transition or a single photon quadrupole transition.

This plate had a 50 µm diameter aperture at the position of the ion’s image so that the fluorescence from the ion could pass through the plate and be captured by a PMT. This technique avoided the background fluorescence from stray scattered light to a high degree. If the cooling laser frequency was swept from low to high values while observing the fluorescence light, curves like those in Fig. 4 resulted.

The radiation from the 194 nm cooling laser had a power of about 4 µW focused to a waist $\omega_0 = 10$ µm at the position of the ion. The curve is asymmetric as expected. This is because when the laser is tuned below the ion’s resonant frequency, cooling takes place and the ion is well localized and has minimal Doppler broadening, therefore the scatter rate is high. As soon as the laser is tuned beyond the ion’s resonant frequency, the ion rapidly heats causing an abrupt decrease in fluorescence because the laser beam/ion spatial overlap is
reduced and the Doppler broadening becomes significant. Hence, to a good approximation, we see only the low frequency side of the ion resonance.

Fig. 4. Photon fluorescent counts (vertical axis) vs. 194 nm laser tuning (horizontal axis) for a single $^{199}$Hg$^+$ ion.

We have also observed the 2 photon $S \rightarrow D$ transition on a single, cold ion. (Last year we observed this on a cloud of room temperature ions). Effort is currently underway to observe the $S \rightarrow D$ transition via quadrupole excitation; this would remove the a.c. Stark shift inherent in the 2 photon excitation. Work is also underway to investigate this transition as a possible optical frequency standard since the lifetime of the D state (0.1 s) gives an intrinsic $Q$ of $10^{16}$. Moreover, we expect to be able to reduce all systematic effects (including the second order Doppler shift) below 1 part in $10^{16}$.

3. Ion Plasma Studies. (Penning Trap)

a. Ion "Cloud" dynamics. The importance of this problem for high resolution spectroscopy is that the primary systematic error for a cloud of ions is expected to be the uncertainty in second order Doppler shift due to the rotation of the ion cloud. Therefore a detailed analysis of this motion is necessary to account for this effect. (We can usually cool the cyclotron and axial velocities to negligible values.) For our one species ion plasmas,
we are usually in the regime where the Debye length of the plasma is small compared to its dimensions; in this case, for ions in thermal equilibrium, we expect a "cloud" of constant density and no shear in the rotation velocity. (If there is shear, frictional heating would occur which would tend to drive the cloud to thermal equilibrium). We can search for shear by measuring the rotational velocity induced first order Doppler shift of an optical transition as a function of laser beam position perpendicular to the rotation (B field) axis. This is shown in Figure 5; to the extent that the slope of the plot is a straight line, there is no shear in the cloud. To within measurement precision, this is the case.

An additional result of our theory would indicate that the ion clouds are uniformly charged spheroids of revolution (about the z or B field axis) and that there is a definite relationship between the aspect ratio of the plasma and the rotation frequency, independent of how the ion cloud is formed (by
laser cooling). Measurements are currently underway to test this theory and so far, agreement is good within our measurement precision of about 10%.

b. Laser induced plasma oscillations. In most of our experiments on laser cooling with Penning traps, the laser simultaneously cools the cyclotron motion and compresses the ion cloud (reduces the magnetron radii). For certain conditions of laser detuning and spatial position, instabilities in the plasma occurs as indicated in the oscillatory fluorescence rate vs. time as indicated in Figure 6. We have observed similar behavior in all the

![Image](image)

Fig. 6 Oscillatory fluorescence level vs. time from a cloud of $^8$Be$^+$ ions.

ion clouds stored in Penning traps. Clearly, in any high resolution optical pumping/double resonance experiment, this is a situation to be avoided since the signal would appear noisy. In the past we have used operating conditions away from such obvious instabilities but more recently we have discovered that even when the regular oscillations are absent, the fluorescence spectrum has $1/f$ noise below about 5 to 10 Hz. Therefore, although not as severe, this $1/f$ noise will limit the signal to noise in resonance experiments and therefore we must investigate this problem further. (See theory section).

c. Liquid and solid plasmas. As an extension of previous work by us, we have reached higher values of $\Gamma$ in our single species plasmas. $\Gamma$ is a parameter which is approximately equal to the ratio of the Coulomb energy between nearest neighbor ions to the kinetic energy per ion. Specifically, $\Gamma = q^2/ak_bT$ where $q$ = ion charge, $k_b$ = Boltzmann's constant, $T$ = temperature and $a$ = mean spacing between ions, i.e. $4\pi a^3/3 = n_0$, where $n_0$ is the ion density. For $\Gamma > 2$ the plasmas should show liquid properties; for $\Gamma < 150$ they should
become solids. Previously we had observed $\Gamma = 10$, recently we have observed $\Gamma = 100$ (Determined by measuring density and temperature). These higher values of $\Gamma$ were realized by using a "diagonal" cooling beam (one which entered and exited the trap in the gaps between ring and endcaps). In the future we hope to observe liquid and solid properties via Bragg scattering (see below).

4. Apparatus construction. Two new trap systems are nearing completion—one for the superconducting magnet (described in the next paragraph) and one for a special $^{24}\text{Mg}^+$ atom state switching experiment described in the proposed work.

B. THEORETICAL

1. Trap design. The special purpose computer program for trap design has been used on the rf trap (Fig. 2) and on two new traps. The most sophisticated one to date is shown in figure 7. By adjusting the aspect ratios and dimensions of the cuts in the traps, the fourth and sixth order terms in the potential are nulled (to within machining errors) which makes the motional resonant frequencies harmonic. A first paper on this computer solution has been accepted by Physical Review and a paper applying this in various cases is being prepared. This is useful for performing mass spectroscopy and more importantly for driving unwanted ions from the trap. For the trap of Fig. 7, the main emphasis is on axial symmetry. At this point, we think that all the unresolved problems in our ion clouds: limit on density, heating, and instabilities are caused by spurious angular momentum being coupled into the ions from the outside. Since we have shown that the effect of background gas collisions is negligible, we are fairly sure this angular momentum is coupled via trap asymmetries. Therefore particular attention has been paid to making the trap highly axially symmetric. This required not only precise machining and alignment but also the need for a second loading trap. During loading, some neutral Mg or Be will inadvertently be plated onto the trap inner electrode surfaces in an asymmetric way. The contact potential variations due to this plating (up to about a volt) would be prohibitively large in the experimental trap necessitating two separate traps.
2. Laser Cooling. Our previous papers on the theory of laser cooling in both harmonic and Penning traps have basically been single particle theories in that Coulomb repulsion between the ions was neglected. In the rf trap, including the Coulomb repulsion gives no substantial difference from the single particle laser cooling theory but for the Penning trap the situation is more complicated. This is primarily because to cool the axial and cyclotron degrees of freedom we must extract energy (this is the usual case) but to "cool" the magnetron degree of freedom (i.e., reduce the magnetron velocity), we must add energy since most of the energy in this degree of freedom is potential energy. It is not surprising that trouble can occur in the special situation where we want to heat and cool at the same time. Recently, we have developed the theory for steady state (assuming a nonlinear angular momentum input which we think is due to trap asymmetries) and have just initiated measurements which seem to confirm this theory. Efforts are continuing to theoretically solve the dynamical problem and explain the oscillation behavior (Fig. 6). This theory is much more complicated and highly nonlinear since the laser cooling rate, torque input from the laser, cloud angular momentum, cloud density, shape, and size and the axial temperature and the cyclotron temperature are all nonlinearly interrelated. It is tempting to conjecture
that the periodic oscillations (Fig. 6) give rise to the 1/f noise through deterministic chaos but not enough is known yet about the oscillation.

3. **Sympathetic cooling.** Self consistent computer solutions have been generated for the two species ion plasma. One solution is shown in Figure 1. Other cases are being investigated, for example the case of simultaneous antiproton/electron storage. This case is particularly interesting in light of the number of people trying to cool, accumulate and store antiprotons for various experiments. Common to all of the groups is the idea to use electrons to sympathetically cool the antiprotons in a Penning trap.

4. **Nuclear diamagnetism via shifts to atomic hyperfine structure.** A strong magnetic field can distort both the electron cloud and the nucleus of an atom, giving rise to small energy shifts in addition to those predicted by the Breit-Rabi formula. Larson suggested that the very small frequency shift due to nuclear distortion (nuclear diamagnetism) may be measurable in an atomic system. We have calculated the size of this effect on the $|\Delta m_I| = 1$ ground state frequencies of several atoms and ions. We find that in general this shift is anomalously large (>50 mHz at 60 KG) in $^{201}$Hg$^+$. A shift of this size would be easily observable. Shifts in $^{26}$Mg$^+$ are considerably smaller (almost 3 orders of magnitude) but we anticipate being able to measure these shifts to about 10% precision.

5. **Telegraphic atomic fluorescence.** Efforts are currently underway to observe "telegraphic atomic fluorescence" in a three level system (in our case the single Hg$^+$ ion). This experimental possibility was originally discussed by Cook and Kimble and has been the subject of several other theoretical papers in the last year (see below, in proposed work). We have recently worked out the theory for observing telegraphic fluorescence via spontaneous Raman scattering (a six level system).

Cook and Kimble discussed the statistics of fluorescence emitted by a single three level atom (ground state "0", excited states "1" and "2"). Fluorescence from the 0-1 "strong" transition reveals the discrete nature of the 0-2 "weak" excitation (level 2 is assumed to radiatively decay slowly back to level 0).
In an alternative system, which is governed by the same rate equations as in Cook and Kimble’s analysis, and should display the same fluorescence statistics, level 2 is excited from and "decays" back to level 0 via spontaneous Raman scattering. Consider a $^{24}\text{Mg}^+$ ion in a magnetic field $(B>1\text{ kG})^2$. Here "0" = $(3s^2S_{1/2},M_J=-1/2)$, "1" = $(3p^2P_{3/2},M_J=-3/2)$ and "2" = $(3s^2S_{1/2},M_J=1/2)$. This system has the advantage that for a laser tuned near the 0-1 transition frequency, the ratio of the mean fluorescence "on" to "off" times is nearly independent of laser power and frequency fluctuations. This has been verified by examining the density matrix equations for the 6 level problem both analytically and on a computer.

6. Temperature measurements via Raman scattering. The present lower limit on temperature is limited by our ability to measure Doppler broadening on the linewidth of the strongly allowed first resonance transitions. For temperatures below 10 mK, the Doppler broadening is a small fraction of the natural width. This problem might be alleviated by driving a stimulated Raman transition between two nearly degenerate levels in the electronic ground state of the ions. In $^{24}\text{Mg}^+$, for example, the $^2S_{1/2}(M_J = -1/2) \rightarrow ^2P_{3/2}(M_J = -1/2) \rightarrow ^2S_{1/2}(M_J = +1/2)$ transition could be driven by using two laser beams separated by the $M_J = -1/2$ and $M_J = +1/2$ ground-state frequency difference. The effects of the laser linewidth would be suppressed by generating the two laser lines with a phase modulator (technique used by S. Ezekiel, MIT); the linewidth of the overall transition would be limited by the ground-state lifetime and thus could be extremely narrow. The intensity of the motional sidebands would depend on $\mathbf{v} \cdot (\mathbf{E}_1 - \mathbf{E}_2)$, where $\mathbf{E}_1$ and $\mathbf{E}_2$ are the wave vectors of the two laser beams and $\mathbf{v}$ is the ion velocity. Thus the angle between the beams could be chosen to optimize the temperature information.

We have theoretically modeled this problem for $^9\text{Be}^+$ ions. In this case the Raman transition would be between two ground state hyperfine sublevels. An experiment was initiated to observe this effect but has been postponed due to the early departure of the post-doc working on this project.

7. Atomic clock theory. We have tried to do a careful design study for an atomic clock based on our present state of knowledge of the system of ions stored in a Penning trap. This study, which will be published in the
The proceedings of the Conference on Precision Electromagnetic Measurements (CPEM, June, '86), includes a detailed comparison of microwave clocks based on $^{201}\text{Hg}^+$, $^{28}\text{Mg}^+$, and $^9\text{Be}^+$. The main result of this study is that we think it would be advantageous to make the next step toward a rf/microwave clock by making one based on $^{26}\text{Mg}^+$ (or $^9\text{Be}^+$) rather than $^{201}\text{Hg}^+$ as we had originally planned. As discussed previously, the single reason for using $^{201}\text{Hg}^+$ vs. $^{28}\text{Mg}^+$ or $^9\text{Be}^+$ is that the "clock" hyperfine transition frequency is about 100 times higher than in $\text{Mg}^+$ or $\text{Be}^+$. All other things being equal—eg. linewidth and signal to noise ratio, the Q of the $\text{Hg}^+$ clock would be 100 times higher and the stability (not necessarily the accuracy) would be 100 times better.

With the main criterion being $10^{-15}$ accuracy, we have determined that the S/N is substantially less in the $\text{Hg}^+$ case and therefore we do not get the factor of 100 in stability. More realistically, a factor of 20 is possible. In addition, with the available 194 nm optical pumping power (about 5 $\mu$W limited by imperfections of available crystals) the optical pumping times are prohibitively long (perhaps an hour) making this system much more difficult to work with. (We could of course make a 194 nm build up cavity but this increases the complexity). There are several other practical factors including pressure problems with neutral $\text{Hg}$ and the necessity to mix the hyperfine ground states (6 microwave oscillators required) which make the $\text{Hg}^+$ system much more difficult to work with.

Moreover, with sympathetic cooling, the performance of a $\text{Be}^+$ or $\text{Mg}^+$ clock would be substantially better than originally expected (primarily because of the expected narrower linewidths). We anticipate that the short term stability would be limited by available local oscillators (active hydrogen masers) when using $\text{Mg}^+$ or $\text{Be}^+$, so that the potential improvement available with $\text{Hg}^+$ could not be realized anyway (at the present time). This, coupled with the fact that the lasers, vacuum equipment and ancillary equipment are much easier to deal with in $\text{Hg}^+$ or $\text{Be}^+$ compel us to make our next clock experiment based on $\text{Mg}^+$ or $\text{Be}^+$. Ultimately, it still appears that the $\text{Hg}^+$ system would give better performance, but for the near future we can learn more about this system ($\text{Hg}^+$) by studying $\text{Mg}^+$ or $\text{Be}^+$.
8. Synchrotron frequency divider. If we are successful in the
development of an optical frequency standard, its use will be somewhat limited
since at present, devices do not exist to measure these frequencies in a phase
coherent way, i.e., count cycles of the radiation. The techniques of harmonic
mixing (originally developed in the low frequency region) appear technically
feasible but so far have not been applied to the visible spectrum. Moreover,
such schemes are very cumbersome due to the many lasers and mixing stages
required. Additionally, the tuning range is limited.

The synchrotron divider could potentially overcome these difficulties. Aside
from its possible practical applications, many interesting basic phenomena
could be studied including the relativistic anharmonic oscillator, and extreme
nonlinear interactions in a new regime. Presently, we are studying some of
the basic processes involved. A graduate student is currently investigating
the possibility of using positive feedback to stabilize the magnetron orbit of
the electron.
OFFICE OF NAVAL RESEARCH
PUBLICATIONS† / PATENTS / PRESENTATIONS / HONORS REPORT
for
1 October 1985 through 30 September 1986
for

Contract N00014-86-F-0004
Proj. Task Area: RRO11-03-01
Work Unit: NR 407-004

"COOLED ION FREQUENCY STANDARD"

Principal Investigator
David J. Wineland

Time and Frequency Division
National Bureau of Standards
Boulder, Colorado 80303
FTS - 320-5286
(303) 497-5286

Reproduction in whole, or in part, is permitted for any purpose of the United States Government.

*This document has been approved for public release and sale; its distribution is unlimited.

†Work reported in these publications was also supported in part by the National Bureau of Standards and The Air Force Office of Scientific Research.
PAPERS SUBMITTED TO REFEREED JOURNALS
(Not Yet Published)


PAPERS PUBLISHED IN REFEREED JOURNALS


BOOKS (AND SECTIONS THEREOF) SUBMITTED FOR PUBLICATION

BOOKS (AND SECTIONS THEREOF) PUBLISHED


PATENTS FILED

None

PATENTS GRANTED

None
INVITED PRESENTATIONS AT TOPICAL OR SCIENTIFIC/TECHNICAL
SOCIETY CONFERENCES

1. "Limits for Spatial Anisotropy by Use of Nuclear Spin Polarized $^9$Be$^+$ Ions", 1985 Gordon Conf. on Atomic Physics; J.D. Prestage.


3. "Frequency Standards Based on Stored Ions", Conference on Precision Electromagnetic Measurements," Gaithersburg, MD June '86; D.J. Wineland.

4. "Laser Cooling of a Single HgII Ion." IQEC '86 June '86; R.G. Hulet.


OTHER INVITED TALKS

7. "Laser Cooled Trapped Ions," Univ. of Nebraska, Colloquium Apr. '86; D.J. Wineland.

HONORS / AWARDS / PRIZES

2. NBS Condon Award (for written exposition) to D.J. Wineland, Dec. '85.
GRADUATE STUDENTS SUPPORTED UNDER
CONTRACT FOR YEAR ENDING 30 SEPTEMBER 1986

Carl S. Weimer (Colorado State Univ.) (Partial support)

POSTDOCTORALS SUPPORTED UNDER
CONTRACT FOR YEAR ENDING 30 SEPTEMBER 1986

John D. Prestage (partial support, finished Feb. 1986)

Lawrence C. Brewer (partial support)

Randall G. Hulet (NRC post-doc, ONR equipment support)

Sarah L. Gilbert (NRC post-doc, ONR equipment support)
END
10-86
DTIC