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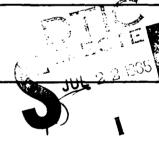
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20 ABSTRACT (Continue on reverse side if necessary and identify by block number)

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 85)

ONR Contract No. NCOO14-85-F-0004

Project Leader:

D. J. Wineland

Frequency & Time Standards Group
524.11

National Bureau of Standards
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#### Contract Description

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 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 ion 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  $\leq 0.1 \text{K}$  are routinely achieved. To avoid light shifts on "clock" transitions we will 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  $^{201}\text{Hg}^+$  ions. This experiment has the goal of realizing a microwave frequency standard with  $10^{-15}$  accuracy.

#### A. Hg<sup>+</sup> studies

#### 1. 198Hg<sup>+</sup> Penning trap experiments

We have written and submitted for publication a report on our measurement of the  ${\rm Hg}^+$   ${\rm g}_J$  factor. This was an important step in the project because of the necessity of "mixing" the Zeeman energy sublevels in the ground state of  $^{201}{\rm Hg}^+$ . (For the  $^{201}{\rm Hg}^+$  "clock" experiment) A previous measurement by another group of  ${\rm g}_J({\rm Hg}^+)$  was not accurate enough and apparently in error by a significant amount.

- 2. Laser cooling of <sup>198</sup>Hg<sup>+</sup> in Penning trap. We have observed a small amount of laser cooling from our 194 nm laser beam. Unfortunately with the small amount of 194 nm power available (≤ 5 μW typical) this laser cooling is not adequate for future "clock" and other spectroscopic experiments. This problem appears to have two practical solutions: (1) Increase 194 nm laser power and improve vacuum in Penning trap (to help prevent rethermalization to ambient temperature by background gas), (2) employ "sympathetic cooling" as described below. We have chosen the latter option because of its inherent advantage over the direct laser cooling method.
- 3. Observation of the  ${}^2S_{1/2} + {}^2D_{5/2}$  "optical clock" transition in  $\frac{198 \, \mathrm{Hg}^+}{}$ . We have constructed a miniature rf trap (Fig. 1) in which we have stored  ${}^{198}\mathrm{Hg}^+$ . By tuning the 194 nm source to the  ${}^2S_{1/2} + {}^2P_{1/2}$  transition (Fig. 2), we can "see" the ions by collecting the 194 nm scattered fluorescence light in a phototube

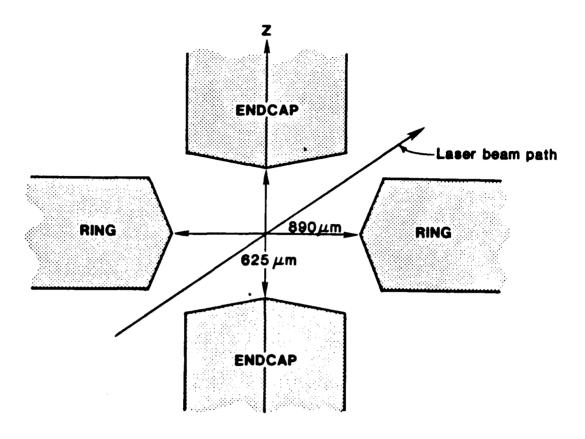


Fig. 1. Schematic showing cross section view of rf trap electrodes for  $^{198}{\rm Hg}^+$ . The electrodes are figures of revolution about the z-axis and are made from molybdenum.

(About 200 ions are stored and thermalized to room temperature using about  $10^{-3}$  Pa of He background gas). The  $^2S_{1/2}$  +  $^2D_{5/2}$  transition is driven via Doppler free two photon spectroscopy ( $\lambda$  = 563 nm); when the transition is made, the number of ions in the  $^2S_{1/2}$  ground state is reduced and therefore the intensity of 194 nm fluorescent light is reduced (Fig. 2). The linewidth in these first measurements was primarily limited by the laser. Future work will be devoted to reducing this width—it should eventually be possible to observe this transition with a width of only 1 Hz! (Q -  $10^{15}$ ) limited by the radiative lifetime. In the course of the above work, optical AM sidebands were observed in the

2-photon spectrum (Fig. 2). This is the first time such sidebands (either AM or FM) have been observed in optical spectra.

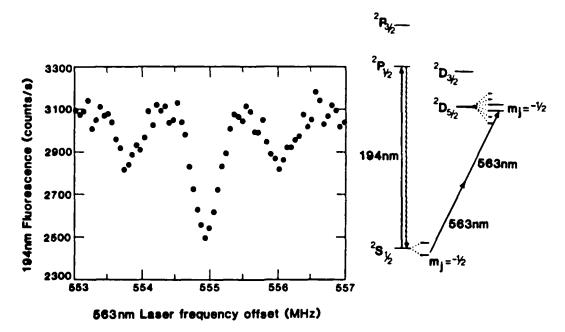


Fig. 2. Two-photon  $^2\mathrm{S}_{1/2}$   $^2\mathrm{D}_{5/2}$  transition in  $^{198}\mathrm{Hg}^+$ . AM sidebands caused by the harmonic secular motion of the ions are visible in this scan. The frequency scan is 4 MHz at the fundamental laser frequency ( $\lambda$  = 563 nm). The depth of the central component is about 25% of full scale. The integration time is 2 s/point. In the inset is a simplified energy level diagram of  $^{198}\mathrm{Hg}^+$ , depicting the levels of interest.

4. Measurement of  $\mathrm{Hg}^+$   $5\mathrm{d}^9$   $6\mathrm{s}^2$   $^2\mathrm{D}_{5/2}$  state lifetime. Using pulsed 2-photon excitation, the lifetime of this state was measured by observing the return of the 194 nm fluoresence to steady state. These measurements agreed with theory (R. Garstang, 1962) and are therefore a good check of the assumptions made in the theoretical calculation on these high Z, relativistic systems.

#### B. <sup>9</sup>Be<sup>+</sup> experiments

- 1. Laser cooled atomic clock. We have completed work on the first frequency standard based on laser cooled atoms. This work which was reported in Phys. Rev. Lett. in April, concentrates on detailed measurements made of systematic effects in this system. Two key features are: (1) an inaccuracy essentially equal to the U.S. best Cesium atomic clock with a clear direction for improvement and (2) this system facilitates studies of generic stored ion frequency standard systematic effects since laser cooling is easier to achieve than in Hg<sup>+</sup>.
- 2. "Strongly coupled" liquid and solid plasmas. Since the previous grant renewal request we have published our first report on the strongly coupled nature of our atomic ion non-neutral plasma [Phys. Rev. Lett. 53, 348 (1984)]. This work is interesting in itself because novel states of matter (liquid and solid plasmas) can be studied but is also very important for the frequency standard problem. This is because the largest uncertainty in any systematic frequency shift in the <sup>9</sup>Be<sup>+</sup> clock (or one based on Hg<sup>+</sup> or another ion) is due to the uncertainty in the ion's kinetic energy and therefore an uncertainty in the second order Doppler frequency shift. Therefore we must study in detail the ion dynamics.
- 3. Laser Cooling experiments. By analyzing the <sup>9</sup>Be<sup>+</sup> clock data, we have determined that when the cooling laser is turned off, the ions are heated by: (1) background gas collisions and (2) a plasma heating process which may be "resonant particle transport". (See Fig. 3) The latter process is one discussed by

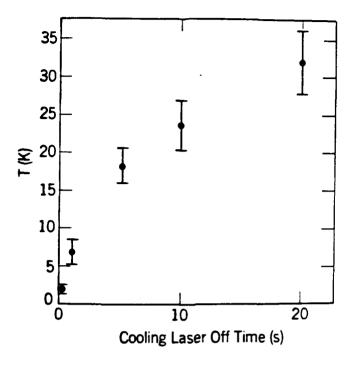


Fig. 3. Temperature, T, of the cyclotron and axial motions as a function of the length of time that the cooling laser is off measured for  ${}^9\text{Be}^+$  ions in a Penning trap.

plasma physicists as being an important loss mechanisms in tandem-mirror fusion devices. There is evidence that it is the same mechanism which causes heating in our Penning traps. One way resonant particle transport is mediated is by misalignment between the trap's magnetic and electric axis. Consequently, we have tried to improve this alignment and have seen about a factor of two reduction in this heating. (We may now be limited by imperfections in the trap electrodes themselves, and we are therefore building a new apparatus with axial symmetry a primary requirement. (See below))

9Be<sup>+</sup> ions. This work, which has been published in Phys. Rev.
Lett. only involved an evaluation and interpretation of <sup>9</sup>Be<sup>+</sup>
clock frequency stability data. It tests one of the foundations of Einstein's theory of General Relativity called "local Lorentz

invariance" at a level 300 times more sensitive than the previous best results provided in separate experiments by Hughes et al. and Drever in 1961.

#### C. Theoretical Studies.

- 1. Studies of trap designs using computer solutions. The trap of Fig. 1 is noteworthy because although the inner surfaces of the trap are machined with simple conical cuts, the trap dimensions and angles were chosen (from computer analysis) to make the fourth and sixth order anharmonic contributions to the potential vanish. This trap therefore approximates very closely one with hyperbolic surfaces. A new trap is in the final design stages. In this trap, axial symmetry (including trap electric and magnetic axis alignment) is emphasized since axial symmetry may play a dominant role in ion heating. Perhaps somewhat surprising is that the best trap for all experiments (except those on mass spectroscopy where absolute harmonicity, or quadratic potentials, are desired) appears to be a simple cylinder with azimuthal cuts made a appropriate places to make the basic Penning trap. Part of the reason for this is that we want the electrodes as far away as possible from the ions in order to avoid imperfections in the trap potential due to contact potential variations etc. In a superconducting magnet geometry, a simple cylinder seems to best satisfy this requirement.
- 2. Angular momentum of trapped atomic particles. We have made a theoretical (and experimental) investigation of angular momentum in the stored ions and come to the conclusion that angular momentum (and its conservation) is very important in Penning traps but much less important (or difficult to make conserved) in rf or neutral particle traps. This work has been submitted for publication in Jour. Opt. Soc. Am. B.
- 3. Spatial anisotropy experiment. (See Sect. III.B.4 above) A significant part of this study involved a theoretical interpretation of the results. Although the tasic theoretical

framework has been established by the general relativists, numerical estimates were required for the particular system ( $^9\mathrm{Be}^+$  nucleus) involved in our experiments.

4. Sympathetic cooling. In the  ${}^{9}\text{Be}^{+}$  clock, it was necessary to turn the cooling laser off while driving the "clock" transition in order to avoid a.c. Stark shifts of the clock energy levels. Because of this, the ions reheated while the laser was off (Fig. 3) causing a second order Doppler frequency shift of about -4 x  $10^{-13}$ . It is the limit in our ability to measure this shift that limits the uncertainty in the experiment to a about 1 x  $10^{-13}$ . We are attacking this problem in two ways. (1) By using better trap designs. As noted previously, we believe that adherence to axial symmetry will reduce this heating. (2) "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 (9Be\*, because of the high laser powers easily generated) sympathetically cools the other ion species via Coulomb collisions. This has been demonstrated previously at NBS, Boulder with the three isotopes of Mg<sup>+</sup>. More recently we have observed that BeH ions are held at the center of trap by laser cooled Be. This technique has two important advantages over the direct laser cooling method (employed in the  $^9\mathrm{Be}^+$  clock experiment): (1) Laser cooling can be supplied by a laser which is easy to make--like the one for Be+. In the 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 ( $<<10^{-15}$ ). 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 theoretically modeled the two species ion thermal distribution functions. The difficult part of this project was a computer solution to an electrostatics problem in order to solve for the spatial distribution of the ions. An important feature is the result that the two ion species should centrifugally separate. This separation is beneficial in that the a.c. Stark shift from the cooling laser is further reduced; it is potentially harmful since it reduces the thermal contact between the ions. Theoretically, this separation does not appear to be strong enough to invalidate sympathetic cooling but it must be tested experimentally.

We have also theoretically estimated the expected light shifts when the cooling laser spatially overlaps the "clock" ion. For the  $^{201}\mathrm{Hg}^+$  case, shifts below  $10^{-15}$  appear very reasonable for expected practical conditions.

#### D. New Apparatus

- 1. 9Be\* apparatus We have modified the 9Be\* clock apparatus to include mirrors which will allow us to direct the popular laser beam along the trap diagonal (similar to what is shown in Fig. ) so that all three degrees of freedom are directly laser for all Previously, only the cyclotron and magnether degrees of freedom were directly laser cooled; the axial motion tended to the trated by photon recoil. This may allow significantly lower temperatures to be achieved.
- 2. Hg\*/Be\* combination trap The previous Hg\* Penning trap has been modified to allow simultaneous storage of Be\* and Hg\* ions--to test sympathetic cooling. Experimental set-up is now being completed.

3. Be<sup>+</sup>/Mg<sup>+</sup> combination trap Design is nearly complete on a trap for simultaneous storage of Be<sup>+</sup> and Hg<sup>+</sup> ions in the high field superconducting magnet. In this trap, particular attention is paid to trap axial symmetry. (See Sects. III.B.3 and III.C.1 above).

#### OFFICE OF NAVAL RESEARCH

PUBLICATIONS / PATENTS / PRESENTATIONS / HONORS REPORT

for

1 October 1984 through 30 September 1985

for

Contract N00014-85-F-0004
Proj. Task Area: RR011-03-01
Work Unit: NR 407-004

"COOLED ION FREQUENCY STANDARD"

Principal Investigator

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#### PAPERS SUBMITTED TO REFEREED JOURNALS

(Not Yet Published)

- "Energy and Radiative Lifetime of the 5d<sup>9</sup> 6s<sup>2</sup> D<sub>5/2</sub> State in HgII by Doppler-Free Two Photon Laser Spectroscopy," J. C. Bergquist, D. J. Wineland, Wayne M. Itano, H. Hemmati, H. -U. Daniel, and G. Leuchs, to be submitted to Opt. Lett. or Phys. Rev. Lett.
- 2. "Measurements of the  $g_J$ -Factors of the 6s  $^2S_{1/2}$  and 6p  $^2P_{1/2}$  States in  $^{198}Hg^+$ ," Wayne M. Itano, J. C. Bergquist, and D. J. Wineland, submitted to J. Opt. Soc. Am.
- 3. "Optical Pumping of Stored Atomic Ions," D. J. Wineland, Wayne M. Itano, J. C. Bergquist, J. J. Bollinger, and J. D. Prestage, to be published by Annales de Physique in the proceedings of "Symposium Alfred Kastler" held in Paris, Jan., 1985.
- 4. "Frequency Standards Based on Stored Ions," D. J. Wineland, IEEE Special Issue on Radio Measurements and Standards.
- 5. "Angular Momentum of Trapped Atomic Particles," D. J. Wineland, J. J. Bollinger, Wayne M. Itano, and J. D. Prestage, submitted for J. Opt. Soc. Am. B, Special Issue on the Mechanical Effects of Light.

#### PAPERS PUBLISHED IN REFEREED JOURNALS

- 1. "Laser Cooled Atomic Frequency Standard," J. J. Bollinger, J. D. Prestage, Wayne M. Itano, and D. J. Wineland, Phys. Rev. Lett. <u>54</u>, 1000 (1985).
- 2. "Limits for Spatial Anisotropy by Use of Nuclear Spin Polarized <sup>9</sup>Be<sup>+</sup> Ions," J. D. Prestage, J. J. Bollinger, Wayne M. Itano, and D. J. Wineland, Phys. Rev. Lett. 54, 2387 (1985).
- 3. "Trapped Ions, Laser Cooling, and Better Clocks,", D. J. Wineland, Science 226, 395 (1984).
- 4. "Frequency Standard Research Using Stored Ions," D. J. Wineland, Wayne M. Itano, J. C. Bergquist, J. J. Bollinger, and H. Hemmati, Prog. Quant. Electron. 8, 139 (1984).
- 5. "Hyperfine Structure of the 2p  $^2P_{1/2}$  State in  $^9Be^+$ ," J. J. Bollinger, J. S. Wells, D. J. Wineland, and Wayne M. Itano, Phys. Rev. A  $\underline{31}$ , 2711 (1985).

#### BOOKS (AND SECTIONS THEREOF) SUBMITTED FOR PUBLICATION

- 2. "Two Photon Optical Spectroscopy of Trapped HgII," J. C. Bergquist,
  D. J. Wineland, W. M. Itano, H. Hemmati, H.-U. Daniel, and G. Leuchs,
  for <u>Laser Spectroscopy VII</u>, Proc. of Seventh Int. Conf. on Laser
  Spectroscopy, Maui, June, '85.

#### BOOKS (AND SECTIONS THEREOF) PUBLISHED

- "Spectroscopy of Stored Atomic Ions," D. J. Wineland, Wayne M. Itano,
   J. C. Bergquist, J. J. Bollinger and J. D. Prestage, in <u>Atomic Physics 9</u>, eds. R. S. Van Dyck, Jr. and E. N. Fortson (World Scientific, Singapore, 1984) p. 3.
- 2. "Progress Toward a Stored Ion Frequency Standard at the National Bureau of Standards," Wayne M. Itano, D. J. Wineland, J. C. Bergquist, F. L. Walls, in <u>Precision Measurement and Fundamental Constants II</u>, Eds: B. N. Taylor and W. D. Phillips, National Bur. Stands. (U.S.) Spec. Publ. 617 (1984), p. 93.
- 3. "Spectroscopy of Stored ions," D. J. Wineland, ibid, p. 83.

#### PATENTS FILED

None

#### PATENTS GRANTED

None

### INVITED PRESENTATIONS AT TOPICAL OR SCIENTIFIC/TECHNICAL SOCIETY CONFERENCES

- 1. "Generation of Strongly Coupled Ion Plasmas," J. J. Bollinger,
  Invited talk at 26th Annual Meeting of the Div. of Plasma Physics,
  Boston, MA, Oct. 29 Nov. 2, 1984.
- "Optical Pumping of Stored Atomic Ions," D. J. Wineland, Invited talk at "Symposium Alfred Kastler," Ecole Normale Superior, Paris, Jan. 1985.
- 3. "Strongly Coupled Nonneutral Ion Plasmas," J. J. Bollinger, Invited talk at the Institute of Theoretical Physics, Workshop on Strongly Coupled Plasmas, Santa Barbara, CA, Feb. 1985.
- 4. "Two Photon Optical Spectroscopy in HgII," J. C. Bergquist, Invited talk, Plenary Session, 39th Annual Freq. Control Symposium, Philadelphia, PA, May 1985.
- 5. "Laser Cooled, Trapped Ions," D. J. Wineland, M. A. Ferst Symposium honoring Prof. N. F. Ramsey. Georgia Tech. June '85.
- 6. "Two Photon Optical Spectroscopy of Trapped HgII," J. C. Bergquist, Seventh Int. Conf. on Laser Spectroscopy, Maui, Hawaii, June 1985.

#### OTHER INVITED TALKS

- "Laser Cooled Ions," D. J. Wineland, Colloquium, Rice Univ., Nov. 1984.
- 2. "Laser Cooled, Trapped Ions," D. J. Wineland, Max Plank Institute, Garching, W. Germany, Jan. 1985.
- 3. "Laser Cooled, Trapped Ions", D. J. Wineland, Universitat Hamburg, W. Germany, Jan. 1985.
- 4. "High Precision Spectra of Laser Cooled Ins," D. J. Wineland, Colloquium, Cal Tech, Feb. 1985.
- 5. "Laser Cooled Ions", D. J. Wineland, Colloquium, Univ. of Georgia, 1985.

#### HONORS / AWARDS / PRIZES

None

#### KEY PERSONNEL (FY 85)

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