

## TIME AND FREQUENCY STANDARDS BASED ON CHARGED PARTICLE TRAPPING

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Summary

Microwave or optical frequency standards based on internal resonance transitions of ions confined in electromagnetic traps have the fundamental advantages of long observation times and small perturbations. These advantages are somewhat offset by low signal to noise ratios. Work at NBS has concentrated on microwave hyperfine transitions of atomic ions stored in Penning-type ion traps. The use of narrowband, tunable light sources for state selection and detection and for reducing the average kinetic energy of the ions (laser cooling) is an important feature of this work. Results to date include the fluorescence detection and cooling to about 50 mK of a single Mg ion and the observation of a 0.012 Hz linewidth on a 300 MHz  $^{25}\text{Mg}^+$  hyperfine transition. A frequency standard based on  $^{201}\text{Hg}^+$  ions is under development. Related work, mostly based on RF-type ion traps, is underway at several other labs.

Introduction

At present, the most accurate (reproducible) frequency standards are based on microwave transitions of atoms or molecules. The stability of a frequency standard increases with increased  $Q$  (transition frequency divided by linewidth) and increased signal to noise ratio. The reproducibility depends upon control of environmental factors. Standards based on narrow optical transitions have the advantage of higher  $Q$  for a given interaction time, for cases where the linewidth is limited by interaction time. However, the use of such a frequency standard to generate precise time, one of the chief applications of frequency standards, is very difficult with current technology. The main difficulty is dividing an optical frequency down to the RF region. Also, high-stability optical sources are not easy to produce.

An atomic frequency standard can be either active or passive in nature. In an active device, such as a self-oscillating hydrogen maser, excited

atoms decay, emitting radiation with a stable frequency. In a passive device, such as a cesium atomic beam, the atomic resonance frequency is probed by radiation derived from an oscillator whose frequency is controlled in a feedback loop so that the frequency of the radiation matches that of the atoms. All of the proposed frequency standards based on stored ions to be discussed in this paper are passive devices.

Ions can be confined for long periods (as long as days) under ultrahigh vacuum conditions in ion traps by electric and magnetic fields. For frequency standard applications, stored ions have the combined advantages of long interaction times (hence narrow resonance lines), because both the storage and relaxation times can be long, and small perturbations to transition frequencies. Atoms in atomic beams also have small perturbations, but the interaction time is limited to the flight time through the apparatus ( $\leq 0.01$  s). Atoms can be stored without relaxation in buffer gases or coated cells for times up to about 1 s, but the transition frequencies are significantly perturbed by collisions. The fundamental disadvantage of ion traps is the low signal to noise ratio, due to the small number of ions that can be stored (typically  $10^6$  or less in a trap of cm dimensions).

Several laboratories have worked on developing a frequency standard based on the 40.5 GHz ground state hyperfine splitting of  $^{199}\text{Hg}^+$  ions stored in a trap of the RF quadrupole (Paul) type.<sup>1-3</sup> State selection and detection is by optical pumping. Resonance light from a lamp containing  $^{202}\text{Hg}^+$  pumps ions from the  $F=1$  level to the  $F=0$  level. Resonant microwave radiation repopulates the  $F=1$  level and is detected by an increase in the resonance fluorescence intensity. Resonance linewidths of about 1 Hz have been observed.<sup>4</sup> At present, the main accuracy limitation is the second-order Doppler (time dilation) shift, which is relatively high (about  $10^{-11}$ ) because the average ion kinetic energy is a few eV. In a similar experiment on trapped  $^{171}\text{Yb}^+$  ions, Blatt et al.<sup>5</sup> have observed a 0.06 Hz linewidth on a 12.6 GHz hyperfine transition, corresponding to a  $Q$

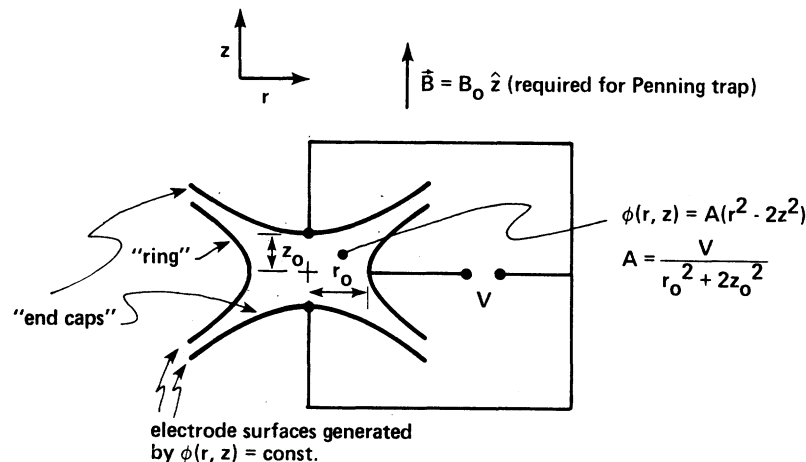


Figure 1. Electrode configuration for RF or Penning trap.

of  $2 \times 10^{11}$ , the highest yet obtained in microwave spectroscopy. Various proposals have been made for optical frequency standards based on narrow transitions in atomic ions,<sup>7,9</sup> but none of them have yet been experimentally realized.

### Experimental Methods and Results

The stored ion experiments at the National Bureau of Standards (NBS)<sup>10-15</sup> have all used Penning traps. In a Penning trap, an outline of which is shown in Fig. 1, ions are confined by a quadrupole electric potential and a uniform magnetic field. Since only static fields are used, cooling the ions to reduce the second-order Doppler shift is much easier than in an RF trap, which uses oscillating inhomogeneous electric fields. Confinement of an ion in a Penning trap is unstable, because collisions with residual gas molecules increase the radial extent of the orbit. In practice, however, ions can be confined for days even without laser cooling (to be explained below), and indefinitely with laser cooling.

Laser cooling (also called optical sideband cooling or radiation pressure 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. The effects of frequency detuning, orientation, and intensity profile of the laser beam on laser cooling of an ion in a Penning trap have been calculated.<sup>18</sup>

Laser cooling of  $\text{Mg}^{+10,11,13}$  and  $\text{Be}^{+15}$  ions has been achieved, using the strongly allowed first resonance lines. For both ions, the light sources were the second harmonics, generated in nonlinear crystals, of CW dye lasers. It is easier to reach very low temperatures with low ion densities, because of space charge induced motion. Single ions can be detected by fluorescence, as shown in Fig. 2. The four plateaus are due to the presence in the trap of three, two, one, and zero  $^{24}\text{Mg}$  ions, which were neutralized, one by one, by  $^{25}\text{Mg}$  atoms coming from an oven.<sup>15</sup> Optical Doppler broadening measure-

ments on a single ion indicated that the effective temperature for motion parallel to the laser beam was about 50 mK.<sup>13</sup> Laser cooling and detection of a single trapped ion has also been reported by Neuhauser et al.<sup>17,18</sup>

Long relaxation times for hyperfine and Zeeman sublevels make possible a very sensitive double resonance technique.<sup>12</sup> In some cases, the laser polarization and frequency can be adjusted so that most of the ions are transferred to the sublevel which is coupled most strongly to the excited state and which scatters photons at a high rate. A transition from this level induced, for example, by microwaves, results in an interruption of the photon scattering until the ion is pumped back to the original sublevel by weak, off-resonance transitions. The number of photons not scattered during this period can be very large, in fact greater than  $10^6$ , so that the microwave transition can be detected with nearly 100% quantum efficiency, even though only a small fraction of scattered photons are counted. This is important for maximizing the signal to noise ratio in a frequency standard.

Figure 3 shows a hyperfine resonance obtained on a small cloud of laser cooled  $^{25}\text{Mg}^{+}$  ions.<sup>14</sup> The oscillatory lineshape results from the use of the Ramsey separated oscillatory field technique, applied in the time domain. Two coherent 1.02 s RF pulses separated by 41.4 s were used to drive the transition. This resonance demonstrates the long relaxation times possible with stored ions. Line broadening due to magnetic field variations was eliminated by operating the trap near a magnetic field at which the derivative of the transition frequency with respect to field is zero.<sup>9</sup> Figure 4 shows a similar hyperfine resonance of  $^9\text{Be}^{+}$ . Two 2 s RF pulses separated by 4 s were applied.<sup>19</sup>

### Future Work

Details of a specific proposal for a microwave frequency standard based on a hyperfine transition of  $^{201}\text{Hg}^{+}$  ions stored in a Penning trap have been published previously.<sup>20</sup> The main advantage of Hg is the high frequency of the transition (26 GHz). The potential accuracy is estimated to be about 1 part in  $10^{15}$ . (At present, the most accurate frequency standards are laboratory  $\text{Cs}$  atomic beams, with an accuracy of 1 part in  $10^{13}$  to  $10^{14}$ .) For laser cooling and optical pumping of Hg, a narrowband, tunable, CW 194 nm source is required. Such a source has recently been developed at NBS.<sup>21</sup> The second harmonic of an argon ion 515 nm laser, generated in an ADP (ammonium dihydrogen phosphate) crystal, is sum frequency mixed with a 792 nm dye

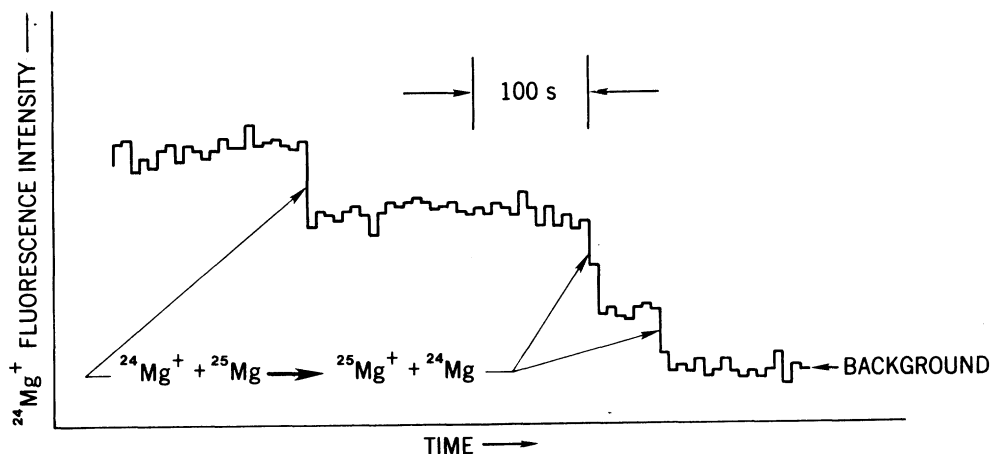


Figure 2. Fluorescence from a small cloud of  $^{24}\text{Mg}^{+}$  ions. The three large steps are due to the loss of individual ions.

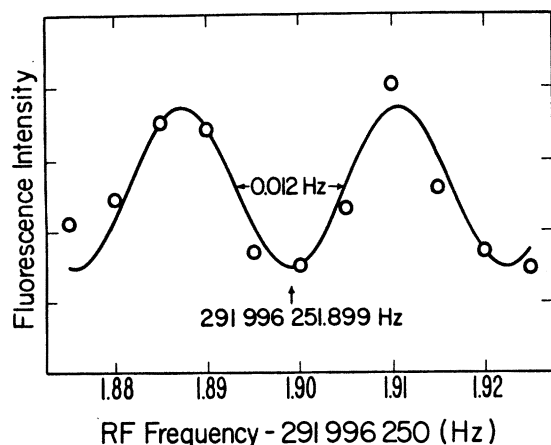


Figure 3.  $^{25}\text{Mg}^+$  hyperfine resonance.

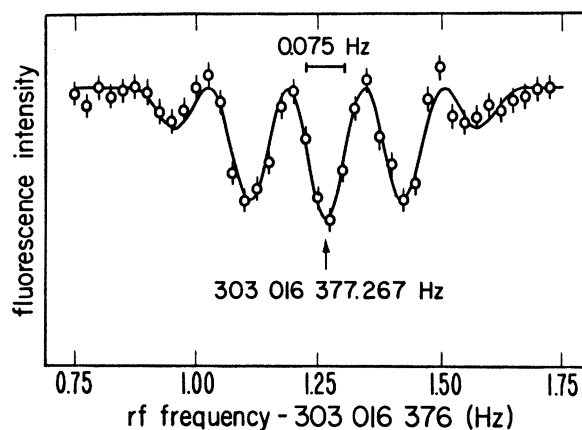


Figure 4.  $^9\text{Be}^+$  hyperfine resonance.

laser in a KB5 (potassium pentaborate) crystal. The output at 194 nm is about 2  $\mu\text{W}$ , which should be enough for preliminary experiments.

#### Acknowledgments

We wish to thank the Air Force Office of Scientific Research and the Office of Naval Research for their support of this work.

#### References

1. F. G. Major and G. Werth, "High-Resolution Magnetic Hyperfine Resonance in Harmonically Bound Ground-State  $^{199}\text{Hg}$  Ions," *Phys. Rev. Lett.* **30**, 1155 (1973).
2. M. D. McGuire, R. Petsch, and G. Werth, "Precision Determination of the Ground-State Hyperfine Separation in  $^{199}\text{Hg}^+$  Using the Ion-Storage Technique," *Phys. Rev. A* **17**, 1999 (1978).
3. M. Jardino, M. Desaintfuscien, R. Barillet, J. Viennet, P. Petit, and C. Audoin, "Frequency Stability of a Mercury Ion Frequency Standard," *Appl. Phys.* **24**, 107 (1981).
4. L. S. Cutler, R. P. Gifford, and M. D. McGuire, "A Trapped Mercury  $^{199}\text{Hg}$  Ion Frequency Standard," *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) pp. 563-578.
5. L. Maleki, "Recent Developments and Proposed Schemes for Trapped Ion Frequency Standards," *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) pp. 593-607.
6. R. Blatt, H. Schnatz, and G. Werth, "Ultrahigh-Resolution Microwave Spectroscopy on Trapped  $^{171}\text{Yb}^+$  Ions," *Phys. Rev. Lett.* **48**, 1601 (1982).
7. H. G. Dehmelt, "Mono-Ion Oscillator as Potential Ultimate Laser Frequency Standard," *IEEE Trans. Instrum. Meas.* **IM-31**, 83 (1982).
8. F. Strumia, "Analysis of New Microwave and Optical Frequency Standards Based on Ion Storage," *Proc. 32nd Annual Symposium on Frequency Control*, U. S. Army Electronics Command, Fort Monmouth, N. J., 1978 (Electronic Industries Assoc., 2001 Eye St., N. W., Washington, D. C. 20006, 1978) pp. 444-452.
9. P. L. Bender, J. L. Hall, R. H. Garstang, F. M. J. Pichanick, W. W. Smith, R. L. Barger, and J. B. West, "Candidates for Two-Photon Absorption Optical Frequency Standards," *Bull. Am. Phys. Soc.* **21**, 599 (1976).
10. D. J. Wineland, R. E. Drullinger, and F. L. Walls, "Radiation-Pressure Cooling of Bound Resonant Absorbers," *Phys. Rev. Lett.* **40**, 1639 (1978).
11. R. E. Drullinger, D. J. Wineland, and J. C. Bergquist, "High-Resolution Optical Spectra of Laser Cooled Ions," *Appl. Phys.* **22**, 365 (1980).
12. D. J. Wineland, J. C. Bergquist, W. M. Itano, and R. E. Drullinger, "Double-Resonance and Optical-Pumping Experiments on Electromagnetically Confined, Laser-Cooled Ions," *Opt. Lett.* **5**, 245 (1980).
13. D. J. Wineland and W. M. Itano, "Spectroscopy of a Single  $\text{Mg}^+$  Ion," *Phys. Lett.* **82A**, 75 (1981).
14. W. M. Itano and D. J. Wineland, "Precision Measurement of the Ground-State Hyperfine Constant of  $^{25}\text{Mg}$ ," *Phys. Rev. A* **24**, 1364 (1981).
15. D. J. Wineland and W. M. Itano, "Spectroscopy of Laser Cooled Be Ions," *Bull. Am. Phys. Soc.* **27**, 471 (1982); J. J. Bollinger and D. J. Wineland, unpublished.
16. W. M. Itano and D. J. Wineland, "Laser Cooling of Ions Stored in Harmonic and Penning Traps," *Phys. Rev. A* **25**, 35 (1982).
17. W. Neuhauser, M. Hohenstatt, and P. Toschek, "Optical-Sideband Cooling of Visible Atom Cloud Confined in Parabolic Well," *Phys. Rev. Lett.* **41**, 233 (1978).
18. W. Neuhauser, M. Hohenstatt, and P. Toschek, "Localized Visible Ba Mono-Ion Oscillator," *Phys. Rev. A* **22**, 1137 (1980).
19. D. J. Wineland, W. M. Itano, and J. J. Bollinger, unpublished.
20. D. J. Wineland, W. M. Itano, J. C. Bergquist, and F. L. Walls, "Proposed Stored  $^{201}\text{Hg}$  Ion Frequency Standards," *Proc. 35th Annual Symposium on Frequency Control*, U. S. Army Electronics Command, Fort Monmouth, N. J., 1981 (Electronic Industries Assoc., 2001 Eye St., N. W., Washington, D. C., 20006, 1981) pp. 602-611.
21. H. Hemmati, J. C. Bergquist, and W. M. Itano, "Generation of CW 194 nm Radiation by Sum Frequency Mixing in an External Ring Cavity," unpublished.