A 303-MHz Frequency Standard Based on Trapped Be⁺ Ions

J. J. Bollinger, D. J. Heinzen, W. M. Itano, S. L. Gilbert, and D. J. Wineland

Abstract—A 303-MHz hyperfine transition in the ground state of ${}^9\mathrm{Be}^+$ was used as a basis of a frequency standard. The ions were stored in a Penning ion trap. Linewidths as narrow as 900 $\mu\mathrm{Hz}$ were obtained. The frequency stability was measured to be better than $3\times10^{-12}\tau^{-1/2}$. The inaccuracy in the second-order Doppler shift was reduced to 5 parts in 10^{15} by laser cooling. An apparent pressure shift with an unexpectedly large value was discovered which limits the accuracy of the current experiment to approximately 1 part in 10^{13} .

I. INTRODUCTON

TON TRAPS provide the confinement with low perturbations Inecessary for improved frequency standards. For a microwave (or RF) frequency standard, typically many ions are stored in a trap in order to increase the signal-to-noise ratio and obtain good frequency stability. With many ions in a trap, the secondorder Doppler shift is one of the largest systematic contributions to the inaccuracy of the standard [11-[6]. In frequency standards which aim at high accuracy, the technique of laser cooling can be used to reduce the second-order Doppler shift to an acceptable level [1]-[3]. In this paper we investigated the use of Be + ions stored in a Penning trap to make a microwave (or RF) frequency standard with high accuracy. Be + is technically easy to trap and laser cool and, therefore, is a good candidate for investigating the use of stored ions as a microwave frequency standard. Other ions with even higher potential line Q's are discussed in [3].

Fig. 1 shows the energy level structure of the ground state of ${}^9\mathrm{Be}^+$ as a function of the trap magnetic field B. At B=0.8194 T, the transition between levels 1 and 2, called the clock transition, depends only quadratically on magnetic field fluctuations and is, therefore, a suitable transition for a frequency standard. In this experiment, an oscillator is locked to this clock transition. The basic idea of this experiment has been described previously [7]-[10]. A description of the current experiment [10] is given below.

II. EXPERIMENTAL TECHNIQUE

Between 5000 and 10 000 $^9\mathrm{Be}^+$ ions and 50 000 to 150 000 $^{26}\mathrm{Mg}^+$ ions were stored simultaneously in a cylindrical Penning trap [11] with $B \simeq 0.8194$ T under conditions of high vacuum ($\simeq 10^{-8}$ Pa). In order to minimize second-order Doppler shifts of the clock transition, the $^9\mathrm{Be}^+$ ions were cooled to less than 250 mK by the following method. The $^{26}\mathrm{Mg}^+$ ions were directly laser cooled and compressed by a narrow-band (~ 1 MHz) ra-

Manuscript received June 13, 1990; revised October 4, 1990. This work was supported by the Office of Naval Research and by the Air Force Office of Scientific Research.

The authors are with the National Institute of Standards and Technology, Boulder, CO 80303.

IEEE Log Number 9042018.

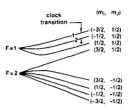


Fig. 1. Hyperfine energy levels (not drawn to scale) of the ${}^{9}\text{Be}^{+}$ 2s ${}^{2}\text{S}_{1/2}$ ground state as a function of magnetic field. At B=0.8194 T the 303-MHz clock transition is independent of magnetic field to first order.

diation source at 280 nm. The ${}^9\mathrm{Be}^+$ ions were then sympathetically cooled [12] by their Coulomb interaction with the cold Mg⁺ ions. A narrow-band 313-nm radiation source was used to optically pump and detect the ${}^9\mathrm{Be}^+$ ions [13], [14]. Lenses were used to image the ${}^9\mathrm{Be}^+$ fluorescence onto the photocathode of a photon-counting imaging detector. The overall detection efficiency was $\sim 2 \times 10^{-4}$. With the 313-nm source tuned to the $2s^2S_{1/2}(m_I=3/2,m_J=1/2)$ to $2p^2P_{3/2}(3/2,3/2)$ transition, 94% of the ${}^9\mathrm{Be}^+$ ions were optically pumped into the $2s^2S_{1/2}(3/2,1/2)$ ground state. The 313-nm source was then turned off to avoid further optical pumping and ac Stark shifts. The sympathetic cooling of the ${}^9\mathrm{Be}^+$ ions by the Mg⁺ ions provided a steady cooling source independent of the 313-nm radiation and therefore permitted the use of long transition times.

The clock transition was detected by the following method. After the 313-nm source was turned off, the ions in the (3/2,1/2) state were transferred to the (1/2, 1/2) state and then to the (-1/2, 1/2) state by two successive RF π pulses. Each pulse was 0.2-s long and resonant with the appropriate transition frequency (around 321 and 311 MHz, respectively). The clock transition was then driven by Ramsey's method of separated oscillatory fields with RF pulses of about 1-s duration and a free-precession time on the order of 100 s. Free-precession periods as long as 550 s were also used. This procedure transferred some of the ions from the (-1/2, 1/2) state to the (-3/2, 1/2) state. Those ions remaining in the (-1/2, 1/2)state were then transferred back to the (3/2, 1/2) state by reversing the order of the two RF π pulses. The 313-nm source was then turned back on, and the population of ions in the (-3/2, 1/2) state was registered as a decrease in the ${}^{9}\text{Be}^{+}$ fluorescence, relative to the steady-state fluorescence, during the first second that the 313-nm source was on. The time for optical pumping from the (-3/2, 1/2) state to the (3/2, 1/2)state was approximately 10 s. After each measurement the system was reinitialized by leaving the 313-nm laser on for about 25 s.

III. RESULTS

Fig. 2 shows the Ramsey signal obtained with a 550-s free precession period. The $900-\mu Hz$ linewidth gives a line Q of 3.4

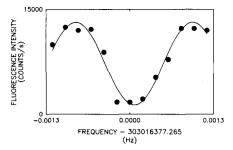


Fig. 2. Ramsey signal of the clock transition for a 550-s free precession period. The data are the result of one sweep (that is, one measurement per point). The sweep width is 2.4 mHz and the frequency interval between points is 0.2 mHz. The dots are experimental and the curve is a least-squares fit.

× 10¹¹. Ramsey signals obtained with a 100-s free precession period were used to servo the frequency of a synthesized RF source [7]-[10]. Measurements were taken near the frequencies corresponding to the half minimum points on both sides of the center frequency. The difference in the measured signal strengths on either side of the line center was used by a computer to servo the average frequency of the synthesizer to the clock transition frequency. Most runs were taken with a commercial cesium beam clock (fractional frequency stability $\sigma_{\nu}(\tau)$ $\approx 6 \times 10^{-12} \tau^{-1/2}$ for measurement time τ in seconds) as the reference oscillator for the synthesizer, but a few runs were taken with a passive hydrogen maser ($\sigma_v(\tau) \approx 2 - 3 \times 10^{-12}$ $\tau^{-1/2}$) as the reference oscillator. The stability of the ${}^{9}\text{Be}^{+}$ clock was measured to be better than $3 \times 10^{-12} \tau^{-1/2}$ for 10^3 s $\leq \tau$ $\leq 10^4$ s, which is within a factor of 4 of the theoretical limiting stability for the number of ions used [3]. For $\tau \ge 10^6$ s the frequency stability was apparently limited by the pressure shift discussed below to $\sim 3 \times 10^{-14}$. The largest contribution to the second order Doppler shift was due to the $\vec{E} \times \vec{B}$ rotation of the ions about the axis of the trap. The rotation frequency and ion cloud radius were measured by using a weak laser beam to probe the ion cloud [13]. The fractional second-order Doppler shift was calculated to be $(-1.2 \pm 0.5) \times 10^{-14}$.

An apparent pressure shift more than three orders of magnitude larger than expected was discovered when the background gas pressure was increased. The background gas pressure could be increased by moving the magnet of the sputter ion pump which evacuated the trap region so that it overlapped fewer pumping cells and reduced the pumping speed. (We checked to make sure the magnetic field at the site of the ions was not disturbed. In addition, the shape and apparent temperature of the ion cloud could be monitored immediately before and after the Ramsey interrogation period from the image of the ion fluorescence. The increased background gas had a negligible effect on the cloud shape and temperature.) The composition of the gas was not known since the pressure was measured with a Bayard-Alpert gauge. However, when the vacuum vessel containing the trap was initially evacuated, the dominant background gases were H2 and He. If the background gas was dominated by He, the fractional pressure shift was about $-3 \times 10^{-6} \, \text{Pa}^{-1}$; if the background was dominated by H2, the pressure shift was about $-9 \times 10^{-6} \text{ Pa}^{-1}$. Atomic ion hyperfine pressure shifts due to He have previously been measured in 137Ba+ [15] and in 199 Hg⁺ [16] to be 5 × 10⁻¹¹ Pa⁻¹ and 4 × 10⁻¹¹ Pa⁻¹, respectively. Vetter et al. [15] show that the long-range chargeinduced interaction between the Ba⁺ and the noble gas atoms gives one of the largest contributions to the pressure shift. Since this interaction depends primarily on the polarizability of the neutral atom we would expect that the pressure shift for He atoms on ${}^9\text{Be}^+$ ions would not be significantly different than that for Ba $^+$ or Hg $^+$. Similarly, since the polarizability of H₂ is midway between Ar and Ne, we might expect the pressure shift for H₂ on Be $^+$ to be near those for Ar and Ne on Ba $^+$, which were measured to be -6×10^{-9} Pa $^{-1}$ and -6×10^{-10} Pa $^{-1}$, respectively [15].

The apparent large discrepancy between our data and other measured pressure shifts is not understood at this time. One possible explanation is suggested by studies [17] of radiative association of C^+ with H_2 to form CH_2^+ . In the models of this process, it is assumed that the H_2 can stick to the C^+ for a long enough time to allow the C^+-H_2 complex to radiatively stabilize. This sticking is possible because the collision energy can be taken up by the internal degrees of freedom in the H_2 molecule. The sticking time can be orders of magnitude longer than the interaction time during a simple elastic collision. If a similar sticking mechanism is active in $H_2 - Be^+$ collisions, it may account for the apparent large pressure shift. We plan to check this hypothesis by measuring the pressure shifts of Be^+ on Ne and Ar, where the sticking would not be expected to occur, and on H_2 , where sticking might occur.

If the apparent collisional shift is real, the background pressure of 10^{-8} Pa in this trap will limit the accuracy of the Be⁺ clock to about 1 part in 10^{13} . We are continuing to search for other causes of systematic error such as electronic offsets. We feel the accuracy could be improved beyond 5 parts in 10^{15} in the future. It may be necessary to use liquid He cryopumping to reduce the background pressure.

ACKNOWLEDGMENT

The authors thank F. Moore and J. Wells for carefully reading the manuscript.

REFERENCES

- [1] D. J. Wineland, W. M. Itano, J. C. Bergquist, and F. L. Walls, "Proposed stored ²⁰¹Hg⁺ ion frequency standards," in *Proc. 35th Ann. Symp. Freq. Control*, Philadelphia, PA, pp. 602-611, 1981.
- [2] D. J. Wineland, W. M. Itano, J. C. Bergquist, J. J. Bollinger, F. Diedrich, and S. L. Gilbert, "High accuracy spectroscopy of stored ions," in *Proc. 4th Symp. Frequency Standards and Metrology*, (Ed. A. DeMarchi) Ancona, Italy, pp. 71-77, 1989.
- [3] D. J. Wineland et al., "Progress at NIST towards absolute frequency standards using stored ions," IEEE Trans. Ultrason. Ferroelec. Freq. Contr., vol. 37, pp. 515-523, 1990.
- [4] M. Jardino, F. Plumelle, M. Desaintfuscien, and J. L. Duchene,

- "Experimental determination of the energy of stored ions from the side-bands in their microwave spectrum," in *Proc. 38th Ann. Symp. Freq. Control*, pp. 431-434, 1984.
- [5] L. S. Cutler, C. A. Flory, R. P. Giffard, and M. D. McGuire, "Doppler effects due to thermal macromotion of ions in an RF quadrupole trap," Appl. Phys. B., vol. 39, pp. 251-259, 1986.
 [6] J. D. Prestage, G. J. Dick, and L. Maleki, "New ion trap for
- [6] J. D. Prestage, G. J. Dick, and L. Maleki, "New ion trap for frequency standard applications," J. Appl. Phys., vol. 66, pp. 1013-1017, 1989.
- [7] J. J. Bollinger, S. L. Gilbert, W. M. Itano, and D. J. Wineland, "Frequency standards utilizing Penning traps," in *Proc. 4th Symp. Frequency Standards and Metrology*, (Ed. A. DeMarchi) Ancona, Italy, pp. 319-325, 1989.
- [8] J. J. Bollinger, W. M. Itano, and D. J. Wineland, "Laser cooled Be⁺ Accurate Clock," in Proc. 37th Ann. Symp. Freq. Contr., pp. 37-41, 1983.
- [9] J. J. Bollinger, J. D. Prestage, W. M. Itano, and D. J. Wineland, "Laser-cooled-atomic frequency standard," *Phys. Rev. Lett.*, vol. 54, pp. 1000-1003, 1985.
- [10] J. J. Bollinger, D. J. Heinzen, W. M. Itano, S. L. Gilbert, and D. J. Wineland, "Test of the linearity of quantum mechanics by RF spectroscopy of the Be⁺ ground state," *Phys. Rev. Lett.*, vol. 63, pp. 1031-1034, 1989.

- [11] S. L. Gilbert, J. J. Bollinger, and D. J. Wineland, "Shell-structure phase of magnetically confined strongly coupled plasmas," Phys. Rev. Lett., vol. 60, pp. 2022-2025, 1988.
- [12] D. J. Larson, J. C. Bergquist, J. J. Bollinger, W. M. Itano, and D. J. Wineland, "Sympathetic cooling of trapped ions: A lasercooled two species nonneutral ion plasma," *Phys. Rev. Lett.*, vol. 57, pp. 70-73, 1986.
- [13] L. R. Brewer, J. D. Prestage, J. J. Bollinger, W. M. Itano, D. J. Larson, and D. J. Wineland, "Static properties of a non-neutral Be+-ion plasma," Phys. Rev. vol. A38, pp. 859-873, 1988.
- [14] W. M. Itano and D. J. Wineland, "Precision measurement of the ground-state hyperfine constant of ²⁵Mg⁺," *Phys. Rev.*, vol. A24, pp. 1364-1373, 1981.
- [15] J. Vetter, M. Stuke, and E. W. Weber, "Hyperfine density shifts of ¹³⁷Ba⁺ ions in noble gas buffers," Z. Phys., vol. A273, pp. 129-135, 1975.
- [16] L. S. Cutler, R. P. Giffard, and M. D. McGuire, "Mercury-199 trapped ion frequency standard," in *Proc. 37th Ann. Symp. Freq. Contr.*, pp. 32-36, 1983.
 [17] E. Herbst, "A reinvestigation of the rate of the C⁺ + H₂ radiative
- [17] E. Herbst, "A reinvestigation of the rate of the C⁺ + H₂ radiative association reaction," Astrophys. J., vol. 252, pp. 810-813, 1982, and references therein.