Laser-Cooled-Atomic Frequency Standard

J. J. Bollinger, J. D. Prestage, Wayne M. Itano, and D. J. Wineland
Time and Frequency Division, National Bureau of Standards, Boulder, Colorado 80303
(Received 5 December 1984)

The first frequency standard based on laser-cooled atoms is reported. Beryllium atomic ions were stored in a Penning trap and cooled by radiation pressure from a laser. The frequency of the $^9Be^+$ $(M_I,M_J)=(-\frac{3}{2},+\frac{1}{2})\leftrightarrow(-\frac{1}{2},+\frac{1}{2})$ ground-state hyperfine transition at its magnetic-field–independent point was determined to be 303 016 377.265 070(57) Hz. The accuracy of a frequency standard referenced to this transition was comparable to the best frequency standards, which are based on cesium atomic beams.

PACS numbers: 32.30.Bv, 06.30.Ft, 35.70.Fk

The principle of laser cooling was introduced[1,2] as a way of suppressing both first- and second-order Doppler shifts in high-resolution spectroscopy. Aside from the interest in the physics of the cooling process itself, two of the primary goals of this technique are more accurate atomic spectroscopy and improved frequency standards. This has been noted in the experimental and theoretical reports on trapped ions,[3-6] cooled atomic beams,[7-9] and trapped neutrals.[9-13] As a step toward these goals we report an accurate measurement of $^9Be^+$ ground-state hyperfine structure and present results on the first frequency (and time) standard based on laser-cooled atoms. Such experiments provide improved measurements of hyperfine constants and g-factor ratios.[5] In addition, they could lead to measurements of previously undetected physical quantities and improvements in previous experimental tests. Examples include the measurement of deviations of atomic hyperfine structure from that predicted by the Breit-Rabi formula due, for example, to nuclear diamagnetism[14] and various tests of gravitational interactions such as tests of spatial anisotropy (Hughes-Drever–type experiments).[15]

In the experiment reported here, $^9Be^+$ ions were confined in high vacuum ($<10^{-7}$ Pa) by the static magnetic and electric fields of a Penning trap.[5] The long confinement times and benign environment of the Penning trap are beneficial for high-resolution spectroscopy.[5] The three axially symmetric electrodes of this trap provide an electrostatic potential of the form $\phi_T=A(2z^2-r^2)$, where r and z are cylindrical coordinates. For $A>0$, positive ions experience a harmonic restoring force along $\hat{\mathbf{z}}$. A uniform magnetic field parallel to $\hat{\mathbf{z}}$ provides confinement in the radial direction. Ion radial motion is a superposition of circular cyclotron motion and a circular $\mathbf{E}\times\mathbf{B}$ drift "magnetron" motion about the axis of the trap.

An essential feature of this experiment was the reduction of the source of the largest systematic uncertainty, the second-order Doppler shift, by reduction of the ion kinetic energy with radiation pressure from a laser.[1-5] Cooling of the axial and cyclotron motions of a trapped ion by a laser beam tuned slightly lower in frequency than a strongly allowed resonance transition can be understood by considering the following one-dimensional model. Suppose that the ions are constrained to move along the z axis in a harmonic well and assume that a laser is directed at the ions along $\hat{\mathbf{z}}$. An ion absorbs and reemits photons predominantly when its velocity is directed against the laser beam, because the light frequency in the ion's frame is Doppler shifted into resonance. When averaged over all angles of reemission, the ion's momentum is reduced by h/λ per scattered photon, where λ is the laser wavelength. The "cooling" laser also couples with the magnetron motion and can be used to reduce the radius of the ion magnetron orbit and therefore compress the radius of the ion "cloud."[3,16]

Specifically, in this experiment a narrow-band (<4 MHz) radiation source (power $\simeq 20\ \mu$W) tuned to the low-frequency side of the

$$2s\,^2S_{1/2}(M_I=-\tfrac{3}{2},M_J=-\tfrac{1}{2})\rightarrow 2p\,^2P_{3/2}(-\tfrac{3}{2},-\tfrac{3}{2})$$

($\lambda=313$ nm) transition of $^9Be^+$ was used to cool and spatially compress the ions and optically pump them into the $(-\frac{3}{2},-\frac{1}{2})$ ground state.[16] The 313-nm source was obtained by frequency doubling of the output of a single-mode cw dye laser. The resonance fluorescence induced by this cooling laser was used to detect the ions.[16,17] The size, density, and temperature of the ion clouds were determined by use of a similar radiation source as a probe.[17] Typical clouds ranged from a few hundred to 2000 ions with cloud diameters from 300 to 500 μm and densities of about 3×10^7 ions/cm^3. Cyclotron and axial temperatures of less than 100 mK and effective magnetron temperatures of less than 2 K were obtained with the cooling laser applied continuously. Ion storage times were many hours without the laser applied; with the laser applied, ion loss is negligible.

At a magnetic field of about 0.8194 T [ground state $(-\frac{3}{2},-\frac{1}{2})\rightarrow(-\frac{3}{2},\frac{1}{2})$ electron spin-flip frequency of 23 914.01 MHz], the $(-\frac{3}{2},\frac{1}{2})\rightarrow(-\frac{1}{2},\frac{1}{2})$ ground-state hyperfine transition, ν_1 (see Fig. 1),

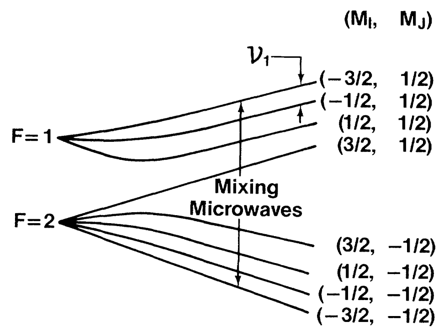

FIG. 1. Hyperfine structure (not drawn to scale) of the $^9Be^+$ $2s\,^2S_{1/2}$ ground state as a function of magnetic field. ν_1 is independent of magnetic field to first order at $B=0.8194$ T.

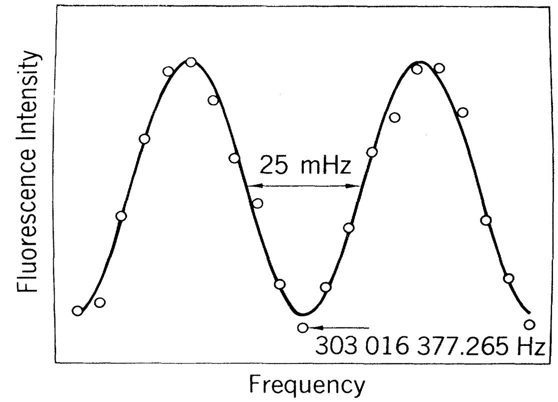

FIG. 2. Signal obtained on the ν_1 field-independent transition for $(t,T)=(0.5$ s, 19 s). The sweep width was 100 mHz and the frequency interval between points was 5 mHz. The dots are experimental and are the average of ten sweeps; the curve is a least-squares fit.

depends only quadratically on the magnetic field deviation, δB, as $\delta\nu_1/\nu_1 = -0.017(\delta B/B)^2$. The ν_1 transition was detected by optical-microwave-rf triple resonance. Microwave radiation tuned to the electron spin-flip resonance transferred half of the ion population from the optically pumped $(-\frac{3}{2},-\frac{1}{2})$ state to the $(-\frac{3}{2},+\frac{1}{2})$ state. Some of the $(-\frac{3}{2},+\frac{1}{2})$ state population was transferred to the $(-\frac{1}{2},+\frac{1}{2})$ state by application of rf near the 303-MHz ν_1 transition frequency. As a result of the microwave mixing this resulted in an additional decrease in the $(-\frac{3}{2},-\frac{1}{2})$ state population and therefore a decrease in the observed fluorescence.

Ramsey's method of separated oscillatory fields[18] was used to interrogate ν_1. An rf pulse of duration t was applied, followed by a free precession interval of duration T and a second rf pulse of duration t coherent with the first pulse. Data were taken with $T=10$ s and $t=0.1$, 0.5, and 2.0 s, and with $T=19$ s and $t=0.5$ s. Typically the cooling laser and mixing microwaves were on for a period of 3 s, during which the $^9Be^+$ ions were prepared in the $(-\frac{3}{2},-\frac{1}{2})$ and $(-\frac{3}{2},+\frac{1}{2})$ states. The cooling laser and mixing microwaves were then turned off in order to avoid light and ac Zeeman shifts during the interrogation period. After the interrogation period, the laser and microwaves were turned back on, and the signal was obtained from the fluorescence count rate during the first 0.3 s of this time interval. Figure 2 shows the signal obtained for the $(t,T)=(0.5$ s, 19 s) interrogation. The linewidth $\Delta\nu_1 = 25$ mHz gives a $Q \equiv \nu_1/\Delta\nu_1$ of 1.2×10^{10} on the 303-MHz ν_1 transition frequency.

A synthesized rf source near 303 MHz was used to probe ν_1. A passive hydrogen maser [fractional frequency stability[19] $\sigma_y(\tau)=1.5\times10^{-12}\tau^{-1/2}$ for measurement time τ in seconds; frequency drift $<3\times10^{-16}$/d] was used as the external reference for this source. A computer alternately stepped the rf frequency by $\pm\Delta\nu_1/2$ about a calculated frequency f_i. After each rf frequency step, a measurement of the signal was made and a new frequency f_{i+1} was obtained about which the next rf step was made. f_{i+1} was obtained through a digital servo system which steered the frequencies $\{f_i\}$ to the ν_1 transition frequency in a way that was independent of a linear drift in the total count rate.[20–22]

Data were taken without interruption for a period of approximately 2 h. After this, the run was stopped and the magnetic field measured and reset if necessary. The average frequency and $\sigma_y(\tau)$ were calculated from the frequencies $\{f_i\}$. For each run, $\sigma_y(\tau)$ was fitted to a $\tau^{-1/2}$ dependence for τ greater than the servo attack time (~ 100 s). A total of sixty runs were used in the final determination of the ν_1 transition frequency. For the 29 runs with a (0.5 s, 19 s) Ramsey interrogation, $\sigma_y(\tau)$ ranged from $1.3\times10^{-11}\tau^{-1/2}$ to $4\times10^{-11}\tau^{-1/2}$. The statistical uncertainty of the average frequency of each run was estimated from the fitted $\sigma_y(\tau)$ at τ equal to the total measurement time of the run. A weighted-average frequency for each group of identical (t,T) interrogations was calculated.

In order to determine the frequency offset due to the second-order Doppler or time dilation effect, the cyclotron and axial temperatures of the ions were measured (via optical Doppler broadening) as a function of the time that the cooling laser was turned off by a technique similar to that discussed in Ref. 17. Figure 3 shows the results of these measurements on typical ion clouds. The heating shown in Fig. 3 produced a second-order Doppler shift, $-\langle v^2/2c^2\rangle\nu_1$, of $-107(27)$ μHz for the (0.5 s, 19 s) interrogation. The contribution of the magnetron rotation of the cloud to the second-order Doppler shift was almost an order of magnitude less. The weighted-average frequency of each (t,T) interrogation was corrected for the total second-order Doppler shift. The weighted average of the resulting frequencies provided (if further sys-

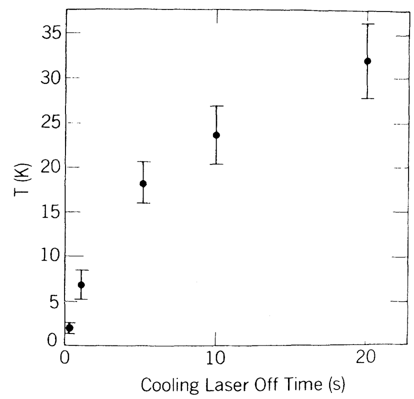

FIG. 3. Temperature, T, of the cyclotron and axial motions as a function of the length of time that the cooling laser is off.

tematic corrections are neglected) a 33-μHz (1.1×10^{-13}) one-standard-deviation determination of the ν_1 transition frequency relative to the passive hydrogen maser reference.

Table I lists the estimated systematic errors. The 3 parts in 10^6 magnetic field fluctuations produced a small offset in the measured ν_1 frequency through the quadratic magnetic field dependence of ν_1. Because ν_1 is a $\Delta m_F = \pm 1$ transition, there was a small shift due to the Earth's rotation. This shift arises because only one rotating component of the applied (linearly polarized) rf field was responsible for inducing the ν_1 transition. The ions may be viewed as being in an inertial frame with the laboratory (including the trap and applied rf field) rotating around the ions once per sidereal day. This produced a shift equal to $f_R \cos\beta$, where f_R is the rotation frequency of the Earth and β is the angle between the magnetic field and the Earth's rotation axis. Microwave leakage during the interrogation period could have produced a frequency shift in ν_1 as large as 1 part in 10^{14}. By stepping the rf synthesizer so that the sides of the ninth side lobes [in the (0.5 s, 19 s) interrogation], rather than the central Ramsey lobe, were sampled, a frequency shift in ν_1 due to a background slope was measured to be less than 5×10^{-15}. We estimated the frequency shift due to the blackbody electromagnetic field at 300 K to be $-3(3) \times 10^{-16}$.[23] Any frequency shift due to a coherence in the hyperfine states that survived the repumping period was eliminated by randomization of the phase of the rf before each interrogation period. Fractional shifts of the ground-state hyperfine frequency of $^{137}Ba^+$ (Vetter, Stuke, and Weber[24]) and $^{199}Hg^+$ (Cutler, Giffard, and McGuire[25]) due to He collisions have been measured to be about 5×10^{-11}/Pa and 4×10^{-11}/Pa, respectively. On the basis of this, a frequency shift in ν_1 due to background neutral collisions with a vacuum better than 10^{-7} Pa was estimated to be much less than 1 part in 10^{15}. We have not considered

TABLE I. Estimated systematic errors. The size of the effect is the fraction of the transition frequency ν_1.

Systematic effect	Size of effect
Second-order Doppler	$-38(9) \times 10^{-14}$
Magnetic field fluctuations	$-1.3(^{+1.3}_{-2.3}) \times 10^{-14}$
Rotation of Earth	$-1.8(0.2) \times 10^{-14}$
Microwave leakage	$\leqslant 1 \times 10^{-14}$
Background slopes	$\leqslant 5 \times 10^{-15}$
Servo offsets	$\leqslant 5 \times 10^{-15}$
Blackbody radiation shift	$-3(3) \times 10^{-16}$
Coherence between cycles	$< 1 \times 10^{-15}$
Background gas collisions	$< 1 \times 10^{-15}$
Stark shifts	$< 1 \times 10^{-15}$
First-order Doppler	$< 1 \times 10^{-15}$
ac Zeeman shifts	$< 1 \times 10^{-15}$
Total systematic offset	$-41.1(9.4) \times 10^{-14}$

any possible frequency shifts due to the presence of nearby conducting surfaces.[26] The systematic uncertainty of 9.4×10^{-14} is comparable to that of the U.S. cesium standard (8.5×10^{-14}).[27] The combined uncertainty due to the random error and the systematic errors of Table I was 34 μHz (1.1×10^{-13}). The final measured frequency of the ν_1 transition was

$$\nu_1 = 303\,016\,377.265\,070(57) \text{ Hz},$$

where the uncertainty includes the 1.5 parts in 10^{13} uncertainty of the passive hydrogen maser frequency relative to the SI second.

The accuracy of the present measurement was limited by the second-order Doppler shift due to the ion heating shown in Fig. 3. The slow heating near the end of the interrogation period was consistent with collisional heating by the room-temperature background gas. The rapid heating observed immediately after the cooling laser was turned off may be caused by asymmetry-induced transport.[28] With the cooling laser off, axial asymmetries of the trap can increase the total canonical angular momentum of the ions, resulting in an increase in the ion-cloud radius. As the ion cloud expands, electrostatic potential energy of the ions due to the space-charge and trap electric fields is converted into thermal energy of the ions. The observed heating can possibly be reduced by improvement of the trap axial symmetry, by use of ion clouds of lower density, and by improvement of the vacuum. The use of a second ion (e.g., $^{24}Mg^+$) to "sympathetically" cool[3,29] the $^9Be^+$ ions could help prevent the heating, whatever its cause. The $^{24}Mg^+$ cooling-laser beam could be applied continuously throughout the interrogation period and could keep the $^9Be^+$ ions cold by Coulomb coupling with the cold $^{24}Mg^+$ ions. Because of the centrifugal separation of the $^9Be^+$ and $^{24}Mg^+$ ions,[30] the overlap of the $^9Be^+$ ions with the $^{24}Mg^+$ cooling-laser beam could be made very small and result in an ac Stark shift on the ν_1 transition of less than 1 part in 10^{15}. Use of these techniques might result in more than an order of magnitude improvement in the performance of the present $^9Be^+$ frequency standard or one based on other ions.[5]

We gratefully acknowledge the support of the U.S. Air Force Office of Scientific Research and the U.S. Office of Naval Research. We thank F. L. Walls for information on the passive hydrogen maser, S. R. Stein for many helpful discussions, and J. C. Bergquist, H.-U. Daniel, and G. Leuchs for carefully reading the manuscript.

[1]T. W. Hänsch and A. L. Schawlow, Opt. Commun. **13**, 68 (1975).

[2]D. J. Wineland and H. G. Dehmelt, Bull. Am. Phys. Soc. **20**, 637 (1975).

[3]D. J. Wineland, R. E. Drullinger, and F. L. Walls, Phys. Rev. Lett. **40**, 1639 (1978).

[4]W. Neuhauser, M. Hohenstatt, P. Toschek, and H. G. Dehmelt, Phys. Rev. Lett. **41**, 233 (1978); H. G. Dehmelt, IEEE Trans. Instrum. Meas. **31**, 83 (1982).

[5]D. J. Wineland, W. M. Itano, and R. S. Van Dyck, Jr., Adv. At. Mol. Phys. **19**, 135 (1983).

[6]R. Blatt, H. Schnatz, and G. Werth, Phys. Rev. Lett. **48**, 1601 (1982).

[7]V. I. Balykin, V. S. Letokhov, and V. I. Mushin, Pis'ma Zh. Eksp. Teor. Fiz. **29**, 614 (1979) [JETP Lett. **29**, 560 (1979)].

[8]W. D. Phillips and H. Metcalf, Phys. Rev. Lett. **48**, 596 (1982).

[9]*Laser Cooled and Trapped Atoms,* edited by W. D. Phillips, U.S. National Bureau of Standards Special Publication No. 653 (U.S. GPO, Washington, D.C., 1983), and Prog. Quantum Electron. **8**, 115 (1984).

[10]A. Ashkin and J. P. Gordon, Opt. Lett. **4**, 161 (1979).

[11]W. H. Wing, Phys. Rev. Lett. **45**, 631 (1980).

[12]D. E. Pritchard, Phys. Rev. Lett. **51**, 1336 (1983).

[13]J. Dalibard, S. Reynaud, and C. Cohen-Tannoudji, Opt. Commun. **47**, 395 (1983).

[14]D. J. Larson, *Hyperfine Interactions* (North-Holland, Amsterdam, 1978), Vol. 4, p. 73.

[15]C. M. Will, *Theory and Experiment in Gravitational Physics* (Cambridge Univ. Press, Cambridge, 1981), p. 30; J. D. Prestage, J. J. Bollinger, W. M. Itano, and D. J. Wineland, to be published.

[16]W. M. Itano and D. J. Wineland, Phys. Rev. A **24**, 1364 (1981).

[17]J. J. Bollinger and D. J. Wineland, Phys. Rev. Lett. **53**, 348 (1984).

[18]N. F. Ramsey, *Molecular Beams* (Oxford Univ. Press, London, 1956).

[19]$\sigma_y^2(\tau)=\langle[f_{i+1}(\tau)-f_i(\tau)]^2/2\bar{f}^2\rangle$, where $f_i(\tau)$ is the ith measurement of the average frequency over a measurement time interval τ, the brackets denote an average over all measurements i, $\bar{f}=\langle f_i\rangle$, and zero dead time is assumed between measurements. See J. A. Barnes *et al.*, IEEE Trans. Instrum. Meas. **20**, 105 (1971).

[20]M. Jardino, M. Desaintfuscien, R. Barillet, J. Viennet, P. Petit, and C. Audoin, Appl. Phys. **24**, 107 (1981).

[21]L. S. Cutler, R. P. Giffard, and M. D. McGuire, in *Proceedings of the Thirteenth Annual PTTI Applications and Planning Meeting, Washington, D.C., 1981* (NASA, Washington, D.C., 1982), NASA Publication No. 2220, p. 563.

[22]J. J. Bollinger, W. M. Itano, and D. J. Wineland, in *Proceedings of the Thirty-Seventh Symposium on Frequency Control, Philadelphia, 1983* (Systematics General Corp., Wall Township, N.J., 1983), p. 37.

[23]W. M. Itano, L. L. Lewis, and D. J. Wineland, Phys. Rev. A **25**, 1233 (1982).

[24]J. Vetter, M. Stuke, and E. W. Weber, Z. Phys. A **273**, 129 (1975).

[25]L. S. Cutler, R. P. Giffard, and M. D. McGuire, in *Proceedings of the Thirty-Seventh Annual Symposium on Frequency Control, Philadelphia, 1983* (Systematics General Corp., Wall Township, N.J., 1983), p. 32.

[26]M. Babiker and G. Barton, Proc. Roy. Soc. London, Ser. A **326**, 255 (1972); G. L. Guttrich and K. W. Billman, Phys. Lett. **24A**, 53 (1967).

[27]D. J. Wineland, D. W. Allan, D. J. Glaze, H. W. Hellwig, and S. Jarvis, IEEE Trans. Instrum. Meas. **25**, 453 (1976).

[28]C. F. Driscoll and J. H. Malmberg, Phys. Rev. Lett. **50**, 167 (1983); D. L. Eggleston, T. M. O'Neil, and J. H. Malmberg, Phys. Rev. Lett. **53**, 982 (1984).

[29]R. E. Drullinger, D. J. Wineland, and J. C. Bergquist, Appl. Phys. **22**, 365 (1980).

[30]T. M. O'Neil, Phys. Fluids **24**, 1447 (1981).