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SINGLE ION OPTICAL FREQUENCY STANDARD*

J. C. Bergquist, Wayne M. Itano, D. J. Wineland, F. Diedrich†,
F. Elsner††, and M. G. Raizen

National Institute of Standards & Technology
325 Broadway
Boulder, CO 80303

Abstract

Experimental research at NIST toward the realization of an optical frequency standard of high accuracy is briefly reviewed. Our studies have concentrated on single, laser-cooled ions since they offer several attractive features toward the achievement of high accuracy (better than 1 part in 10^{17}). These features include long storage times which eliminate transit-time broadening, confinement that is nearly nonperturbative to the internal level structure of the ion, laser-cooling that reduces motional shifts to small values, and "electron-shelving" whereby transitions to long-lived states can be detected with unit probability. We have studied spectroscopically the electric-quadrupole allowed transition at 282 nm ($\sim 1 \times 10^{15}$ Hz) in a single, laser-cooled $^{199}Hg^+$ ion stored in an rf Paul trap with extremely high resolution. The measured linewidth is limited presently by the spectral purity of the laser to about 80 Hz. Possible improvements and future directions will be discussed.

Introduction

A single atom "at rest in space" has been promoted as an ideal system for spectroscopic measurements.[1] Certainly, a motionless atom that is free of any perturbations must be considered as an ideal reference for a frequency (and time) standard with high accuracy. A close approximation to this system is a single, laser-cooled ion in a miniature radio-frequency (rf) Paul trap. It appears realistic that a single, trapped ion could be free from perturbations due to collisions, Doppler shifts and electric or magnetic fields to the order of 10^{-18}.[1,2] However, a reference based on one atom is limited to a signal-to-noise ratio of unity (at best) for a single measurement cycle.[2-4] Therefore, the stability of a frequency standard is compromised by using single ions to achieve high accuracy. The loss of stability caused by using a single ion can be partially recovered by locking to a narrow transition with a resonance frequency that lies in the optical region of the spectrum. The stability of an oscillator locked to N atoms using the Ramsey method of interrogation (assuming 100% detection efficiency) is given by [5]

$$\sigma_y(\tau) = (\tau N T_R \omega_0^2)^{-½} \quad (\tau > T_R) \tag{1}$$

where τ is the averaging time, T_R is the Ramsey interrogation period and ω_0 is the transition frequency in radians per second. If the atomic transition is in the middle of the visible spectrum ($\omega_0/2\pi \simeq 5 \times 10^{14}$) and the Ramsey period is 1 s, the stability for a single atom could be as good as $3.2 \times 10^{-16}\tau^{-½}$. The potential high accuracy and stability of a single-ion optical frequency standard make it attractive as a clock. An important problem, however, is the difficulty in generating time from optical frequencies. The technology to connect microwave frequencies to visible frequencies has been demonstrated,[6] but it is cumbersome, laser intensive and narrowband. Schemes for broadband division from the optical region to the microwave region[7,8] and for simpler multiplication have been proposed and are being investigated.[8] These would help realize the full benefits of a single ion, optical frequency standard.

Experiment

Presently at NIST we are studying the $5d^{10}6s\ ^2S_{½}(F=0,m_F=0) \rightarrow 5d^96s^2\ ^2D_{5/2}\ (F=2,m_F=0)$ electric-quadrupole transition ($\omega_0/2\pi \simeq 1 \times 10^{15}$ Hz) in $^{199}Hg^+$ as an optical frequency standard.[3,5,9,10] There are other ions with credentials to compete as an optical frequency standard[1,11] and several groups throughout the international community are investigating some of these choices. A ^{199}Hg atom is ionized and trapped in the harmonic pseudopotential well created by an rf potential applied between the electrodes of a miniature Paul trap (endcap separation $2z_0 \simeq 660\ \mu m$). The amplitude of the rf potential (at frequency $\Omega/2\pi \simeq 21$ MHz) could be varied up to 1.2 kV. The ion is laser-cooled to a few millikelvins by a few microwatts of radiation from two 194 nm sources.

The necessity of using two radiation sources to laser-cool $^{199}\mathrm{Hg}^+$ is caused by its hyperfine structure. A mercury isotope with hyperfine structure is required in order to have a first-order field-independent clock transition near $B = 0$. The frequency of one source is tuned slightly below the resonance of the strongly allowed transition from the $5d^{10}6s\ ^2S_{1/2}(F=1)$ level to the $5d^{10}6p\ ^2P_{1/2}(F=0)$ level (see Fig. 1). Since transitions from the $F=0$ excited level to the $F=0$ ground level are forbidden, the $^2S_{1/2}(F=1)$ and the $^2P_{1/2}(F=0)$ levels nearly comprise a two-level system. However weak off-resonance excitation in the Lorentzian tail of the $^2S_{1/2}(F=1) \rightarrow {}^2P_{1/2}(F=1)$ transition causes pumping into the $F=0$ ground level through the $F=1$ excited level. The second 194 nm source is tuned to the $^2S_{1/2}(F=0)$ to $^2P_{1/2}(F=1)$ transition in order to optically pump the ion back to the $F=1$ hyperfine level in the ground state. In this way, the ion can be cooled to near the Doppler cooling limit of 1.7 mK by scattering photons at 194 nm. Transitions to other levels are detected by measuring the fluorescence count rate from the 194 nm photons that are scattered by the ion and collected in a solid angle of about $5 \times 10^{-3} \times 4\pi$ sr. For $^{199}\mathrm{Hg}^+$, we achieve a peak count rate of about 25,000/s with a detector efficiency of 10%.

The narrow $^2S_{1/2}(F=0,m_F=0)$ - $^2D_{5/2}(F=2,m_F=0)$ clock transition is coherently driven by radiation at 282 nm obtained by frequency doubling the radiation from a cw dye laser that is stabilized to a high finesse cavity.[9,10] The frequency of the fundamental radiation at 563 nm could be offset from the cavity resonance and tuned through the ion resonance by means of an acousto-optic modulator. The 282 nm radiation and the 194 nm radiation are turned off and on sequentially in order to prevent broadening and shifts of the narrow S-D transition. Optical-optical double resonance[1,3,12] (electron shelving[1,13]) is used to detect each transition to the metastable D state as a function of the frequency of the 282 nm laser. At the beginning of each measurement, the ion is prepared in the $F=0$ hyperfine state in the ground level by blocking the 194 nm source tuned to the $^2S_{1/2}(F=0)$-$^2P_{1/2}$ $(F=1)$ transition for a period of 5 ms. During this time the 194 nm source tuned to the $^2S_{1/2}(F=1)$ - $^2P_{1/2}$ $(F=0)$ transition optically pumps the ions into the $F=0$ ground state. After this period, both sources at 194 nm are blocked and the 282 nm light is permitted to radiate the ion for a period that was varied up to 15 ms. The frequency of the 282 nm radiation was tuned to resonance or near resonance with the $^2S_{1/2}(F=0,m_F=0)$ - $^2D_{5/2}(F=2,m_F=0)$ transition. At the end of the probe period, the 282 nm radiation was turned off and both 194 nm sources were turned back on. If 194 nm fluorescence was detected no transition to the D state was recorded; if no fluorescence was detected, a transition to the D state was recorded. The data was digitized, 1 for fluorescence and 0 for no fluorescence, and then averaged with the previous results at this frequency. Then the frequency of the 282 nm radiation was stepped and the measurement cycle repeated. With the high fluorescence rate at 194 nm, it was possible to detect each transition with nearly no ambiguity in 10 ms.

Since there were long term drifts in the frequency of the narrow-band 282 nm radiation, we locked its frequency to the narrow S-D transition with an attack time of a few seconds.[10] To do this, we began each measurement cycle by stepping the 282 nm radiation to near the half power point on each side of the resonance N times (N varied from 8 to 32). At each step, we probed for 5 ms and then looked for any transition with the electron-shelving technique. We averaged the N results from each side of the resonance line, took the difference and corrected the frequency of the 282 nm laser. In this way, variations in the frequency of the 282 nm laser for time periods exceeding a few seconds were reduced.

In Fig. 1, we show a spectrum obtained of the resonance of the narrow S-D transition. For this figure 138 consecutive scans were made where the 282 nm probe period was 15 ms, and the step size was 15 Hz at 563 nm (30 Hz at 282 nm). The resonance shows a clearly resolved triplet with the linewidth of each component less than 40 Hz (< 80 Hz at 282 nm). We believe that this triplet structure is caused by Rabi power broadening. The 282 nm radiation is focussed on the ion to a spot size of about 25 μm; therefore, on resonance, a laser flux of fewer than 10^6 photons/s will saturate the transition. Below the data is a theoretical lineshape calculated for an ion at rest, for no broadening due to collisions or laser bandwidth, for a pulse length of 15 ns and for sufficient power to give a 3.5 π-pulse. Qualitatively the figures compare well. The fluctuations from measurement cycle to measurement cycle in the quantum-occupation number of the ion in the harmonic well of the trap cause variations in the transition probability of the ions. This, and the finite linewidth of the laser, likely cause the general broadening and weakening of the signal. We plan to study the lineshape and the effects of power broadening in more detail in future experiments.

We have discussed so far the possibility of improving the frequency stability in a single-ion frequency standard by increasing the frequency of the clock transition. From Eq. 1, we see that it is also possible to improve the stability of the frequency device as the square root of the number of trapped ions. However, for two or more laser-cooled ions in a quadrupole rf trap, the Coulomb force between ions repels them apart and away from trap center. If the ions do not reside at the center of the trap, they experience a force from the rf trapping potential whose magnitude increases with the distance away from trap center. This force causes motion of the ion at the trapping field frequency. Thus the ions are no longer at rest in space and the second-order Doppler shift due to the motion of ions at the rf frequency (termed "micromotion") grows

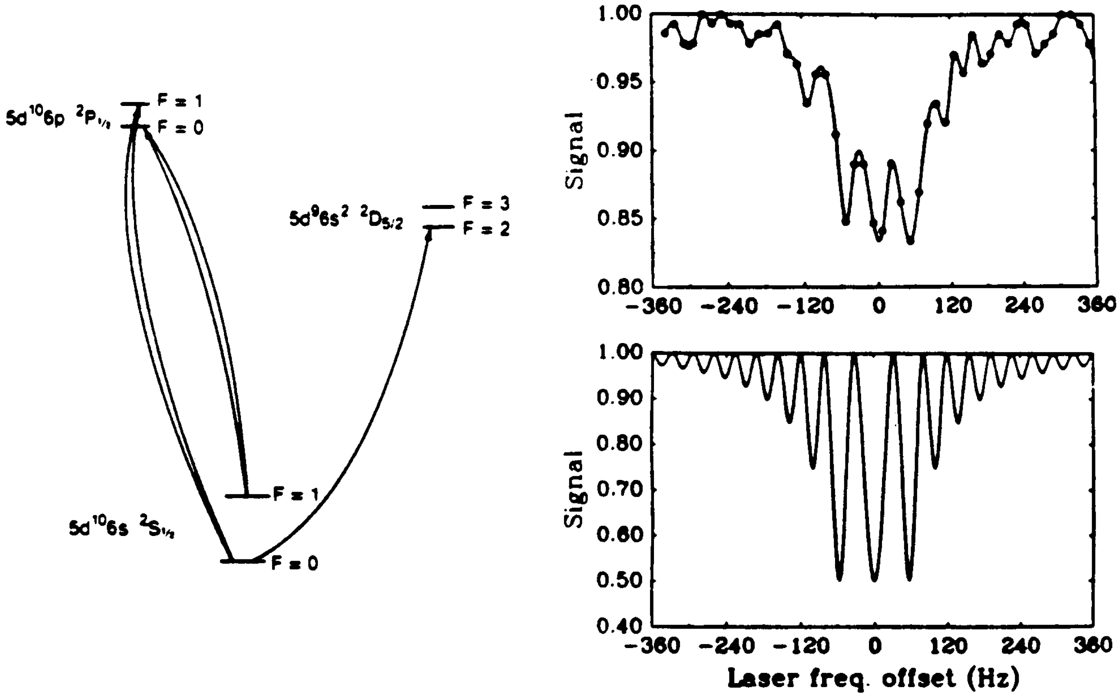

Figure 1. On the left is a simplified energy level diagram for $^{199}Hg^+$ at zero magnetic field. The quantized signal of the power-broadened lineshape obtained by scanning through the Doppler-free resonance of the $^2S_{1/2}(F=0,m_F=0)$ - $^2D_{5/2}(F=2,m_F=0)$ transition in a single laser-cooled $^{199}Hg^+$ ion is shown in the upper figure on the right. The frequency of a narrowband laser at 563 nm is doubled and long-term stabilized by locking to the S-D transition in the ion. The frequency of the laser is then stepped through the resonance in 15 Hz increments (30 Hz increments at 282 nm) for 138 consecutive sweeps. The lower-right figure shows the lineshape calculated for conditions similar to the experimental conditions for the upper figure, except that the ion is assumed to have zero temperature and the laser is assumed to have zero linewidth.

substantially larger than 2×10^{-18}.[2]

Another type of trap, the linear rf trap,[5,14-18] may allow the possibility to trap many ions while at the same time keeping the amplitude of their micromotion small. Ideally, the energy in the micromotion should be approximately equal to the energy in the laser-cooled secular motion of the ion. If so, then, even with many cold ions, it will be possible to achieve small second-order Doppler shifts ($< 2 \times 10^{-18}$). In fact, the fractional second-order Doppler shift should be somewhat less in a linear rf trap than in a quadrupole rf trap[5,16] because of the absence of rf micromotion along the symmetry axis of the linear trap. A schematic of a linear rf trap is shown in Fig. 2. An rf potential is applied to a pair of opposing rods while the other pair is held at ground potential. This produces a time averaged force which confines ions in the radial direction. An electrostatic potential applied to the end sections of the electrodes confines the ions along the trap axis. In a linear trap the rf fields approach 0 along a line (ideally along the trap axis) rather than only at a point as in the quadrupole rf trap.

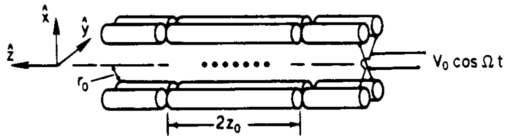

Figure 2. An example of a possible electrode configuration for a linear rf trap. In this configuration, if the length of the electrodes is much greater than the linear extent of the ions, the rf potential at the position of the ions has no z component. Thus, the energy in the micromotion of ions that are trapped along the axis of symmetry (the z-axis) can be on the order of the secular motion energy.

Use of a linear rf trap would allow us to store many laser-cooled ions ($N \simeq 50$), each nearly at rest in a benign environment and each acting as an independent clock.[5,14,15] Hence, the limitation to accuracy caused by second-order Doppler shifts in a frequency standard based on a linear array of trapped ions should be equivalent to a single ion frequency standard and the stability should be better by the square root of the number of stored ions. Thus, a linear trap frequency standard becomes attractive even for microwave frequencies. For example, from Eq. 1, for $T_R = 100$ s, $N = 50$ and $\omega_0/2\pi = 40.5$ GHz (the ground state hyperfine splitting in $^{199}\mathrm{Hg}^+$) the frequency stability would be $\sigma_y(\tau) = 5.5 \times 10^{-14}\tau^{-1/2}$. We have begun our studies of both a microwave and optical frequency standard based on a linear trap with up to approximately 30 laser-cooled ions. We have observed crystallization of the cold ions along the trap axis, and observed the 40.5 GHz microwave transition with a fractional resolution of about 6×10^{-12}. Future possibilities include using the nearly motionless string of ions in tests of various principles in fundamental physics such as interference experiments and cavity QED.

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†Present address: Gsänger Optoelektronik GmbH, Robert-Koch-Straße 1a, D-8033 Planegg 1, Germany
††Present address: Universität Hamburg, D-2000 Hamburg, Germany

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