The JPL Trapped Mercury Ion Frequency Standard

J. D. Prestage, G. J. Dick, and L. Maleki

Communications Systems Research Section

In order to provide frequency standards for the DSN which are more stable than present-day hydrogen masers, a research task was established under the Advanced Systems Program of the TDA to develop a $^{199}Hg^+$ trapped ion frequency standard. This article describes the first closed-loop operation of this standard.

Mercury 199 ions are confined in an RF trap and are state-selected through the use of optical pumping with 194-nm UV light from a ^{202}Hg discharge lamp. Absorption of microwave radiation at the hyperfine frequency (40.5 GHz) is signaled by atomic fluorescence of the UV light. The frequency of a 40.5-GHz oscillator is locked to a 1.6-Hz-wide atomic absorption line of the trapped ions. The measured Allan variance of this locked oscillator is currently $\sigma_y(\tau) = 4.4 \times 10^{-12}/\sqrt{\tau}$ for $20 < \tau < 320$ seconds, which is better stability than the best commercial cesium standards by almost a factor of 2. This initial result was achieved without magnetic shielding and without regulation of ion number.

I. Introduction

Recently, there has been much activity directed toward the development of trapped ion frequency standards. This has occurred because ions confined in an RF quadrupole trap are subjected to very small perturbations of their atomic energy levels and to weak forces which equalize any population differences among the ground-state hyperfine levels. Potentially, the largest source of frequency fluctuation for such a standard stems from the motion of the atoms within the trap via the second-order doppler or relativistic time dilation effect. To minimize this perturbation, heavy ions are preferable to light ions, since for a given energy a heavy ion will have a smaller velocity. For this reason and for other reasons discussed later in this article, $^{199}Hg^+$ ions have been used for most trapped ion frequency standard work.

Only two parameters are needed to describe the short-term stability of a passive atomic frequency standard. One is the line Q, which equals $f/\Delta f$ where f is the resonant frequency of the reference atom and Δf is the width of the atomic resonance. For $^{199}Hg^+$, f is 40.5 GHz and Δf is as small as 0.1 Hz; thus, trapped ion standards have line Q's which are orders of magnitude higher than other microwave atomic frequency standards.

The other parameter that determines stability is the signal-to-noise ratio (SNR) achieved in measuring the atomic resonance. The short-term stability is inversely proportional to the product of Q and SNR. At present, four groups are developing trapped Hg+ ion clocks: Hewlett-Packard in Palo Alto, California [1]; the National Bureau of Standards in Boulder, Colorado [2]; l'Universite Paris-Sud in Orsay, France [3]; and JPL/NASA. It should be noted that F. G. Major first proposed the use of trapped $^{199}Hg^+$ ions as a frequency standard while working for NASA at the Goddard Space Flight Center in 1969 [4].

II. Ion Trapping

The electrode structure and time-varying voltages used to trap ions are shown in Fig. 1. The electric potential inside the trap when no ions are present is

$$V_T = \frac{(U_0 + V_0 \cos \Omega t)(r^2 - 2z^2)}{\xi^2}$$

where, for the present work, $\Omega = (2\pi)\ 500$ kHz, $U_0 = 20$ V, $V_0 = 660$ V, $\xi^2 = (r_0^2 + 2z_0^2)$, and r_0 = the inside radius of ring electrode = 1.9 cm = $\sqrt{2}\,z_0$.

A charged particle moving in the inhomogeneous oscillatory electric field of the ion trap feels a net force (averaged over one cycle of Ω) toward the region of the weaker field provided that the amplitude of its motion at frequency Ω is small compared with its distance from the center of the trap. The motion under these conditions is a combination of a fast oscillation at frequency Ω (micromotion) and a slower frequency ω, as shown in Fig. 2.

The action of the RF field in trapping ions is described by the electric pseudopotential energy [5]:

$$\Psi = \frac{e^2\,V_0^2\,(r^2 + 4z^2)}{m\Omega^2 \xi^4}$$

where m and e are the ion's mass and charge, respectively. The dc potential energy

$$\phi_{dc} = \frac{eU_0\,(r^2 - 2z^2)}{\xi^2}$$

is added to the pseudopotential, giving the total potential energy for an ion in the trap:

$$\phi_T = \frac{m\omega_r^2 r^2}{2} + \frac{m\omega_z^2 z^2}{2}$$

where

$$\omega_r^2 = \frac{2e^2 V_0^2}{m^2 \Omega^2 \xi^4} + \frac{2eU_0}{m\xi^2}$$

and

$$\omega_z^2 = \frac{8e^2 V_0^2}{m^2 \Omega^2 \xi^4} - \frac{4eU_0}{m\xi^2}$$

Under the conditions listed earlier, $\cong 19$ electron volts of kinetic energy is required for a ^{199}Hg$^+$ ion at the trap center to reach one of the trapping electrodes.

Our trap is inside a vacuum chamber with a pressure of $\cong 1$–2×10^{-8} torr. By heating a powder of isotopically enriched mercuric oxide (HgO) to about 100°C, a vapor of neutral ^{199}Hg fills the vacuum chamber to about 10^{-8} torr partial pressure. Electrons from an LaB$_6$ single-crystal filament are injected into the trap ($\cong 30$ μA, 300 V), ionizing some of the neutral ^{199}Hg inside the trap electrode structure.

The resulting ion cloud is much hotter than the room-temperature neutral vapor in part because ionization of the vapor takes place throughout the trap. It has been found experimentally that the average kinetic energy of the ions is about 10 percent of the well depth. The resulting 2 eV of kinetic energy would produce a fractional second-order doppler shift of about 10^{-11}. To reduce this shift, the vacuum system is filled to about 10^{-6} torr of ^{4}He. The ^{199}Hg$^+$ ions collide with these room-temperature helium atoms and are cooled to just above room temperature.

III. Magnetic Levels and State Selection

The magnetic structure of the ground-state hyperfine levels of ^{199}Hg$^+$ is shown in Fig. 3. The energy difference between the $(F = 0, m_F = 0)$ and $(F = 1, m_F = 0)$ levels is used to define the standard frequency, approximately $f_{Hg^+} = 40.507347997$ GHz. The measured frequency, f, depends quadratically on the magnetic field at the position of the ion cloud, $f = f_{Hg^+} + 97B^2$ (Hz/G^2). For comparison, the field dependence for hydrogen atoms is $f = f_H + 2750B^2$ (Hz/G^2).

The ions are state selected by use of optical pumping with light from a ^{202}Hg discharge lamp. The energy levels of ^{202}Hg$^+$ and ^{199}Hg$^+$ are compared in Fig. 4. Ultraviolet light of wavelength 194.2 nm ($\cong 6.4$ eV) from the ^{202}Hg lamp, when collected and focused onto the ^{199}Hg$^+$ ions, will excite the transition $^2S_{1/2}\ (F = 1, m_F) \rightarrow {}^2P_{1/2}$.

The $^2P_{1/2}$ state decays after 2 ns lifetime to either the $^2S_{1/2}\ (F = 0, m_F = 0)$ or the $^2S_{1/2}\ (F = 1, m_F)$ state, thereby scattering a 194-nm photon. Since the transition $^2S_{1/2}\ (F = 0, m_F = 0) \rightarrow {}^2P_{1/2}$ is not resonant with the light from the ^{202}Hg lamp, the ions are pumped out of the $^2S_{1/2}\ (F = 1, m_F)$ states into the $^2S_{1/2}\ (F = 0, m_F = 0)$ state, at which time

the ions stop scattering UV light. A flux of about 3×10^{12} photons per second per square centimeter passing through the ion cloud will depopulate the $^2S_{1/2}$ ($F = 1, m_F$) levels in about 1/2 second. An oscillating magnetic field (strength 10^{-6} G) at a frequency of 40.507347997 GHz will transfer the atoms from the $^2S_{1/2}$ ($F = 0$, $m_F = 0$) state to the $^2S_{1/2}$ ($F = 1$, $m_F = 0$) state in about 1 second. The ions will then scatter UV light until they are pumped back into the nonfluorescing $^2S_{1/2}$ ($F = 0, m_F = 0$) state.

IV. UV Optical System

Figure 5 shows the optical system used to collect and focus UV light from the ^{202}Hg lamp onto the ion cloud. The lamp is excited with 15–20 watts of RF power (160 MHz), creating a very bright discharge in the quartz cell containing the ^{202}Hg vapor with about 30 millitorr of argon buffer gas. The useful light from the ^{202}Hg lamp is from the 194.2-nm transition in ^{202}Hg$^+$, as shown in Fig. 4.

However, the brightest wavelengths produced in the lamp are from transitions in the neutral mercury atom. Any light detected at wavelengths other than 194 nm will degrade the SNR of the measured atomic resonance. There are three ways we suppress the detection of light with wavelengths different from 194 nm:

(1) The photomultiplier tube (PMT) is sensitive only to light that has wavelengths between 160 and 320 nm with peak sensitivities of 12 to 15 percent at 210 nm. The brightest line coming from the lamp in this bandwidth is 254 nm and is 200 times brighter than the 194-nm line.

(2) The ellipsoidal collection mirror has a thin-film dielectric coating which maximizes reflection at 194 nm while keeping the reflectivity at 254 nm at 10 percent, with lower reflectivities for longer wavelengths. The entrance window to the trap region is coated to be 98 percent reflective at 254 nm while being 90 percent transmitting at 194 nm.

(3) All light collected by the detection optics—stray scattered light plus fluorescent light from the atoms—is filtered with a 194-nm bandpass filter with a peak transmission of 30 to 40 percent and a bandwidth of 45 nm. The solid angle subtended by the collection optics around the trap center is 5 percent of the total 4π solid angle. The total collection efficiency of this detection system is equal to the solid angle multiplied by the bandpass filter loss multiplied by the PMT sensitivity, or $0.05 \times 0.35 \times 0.12 = 2 \times 10^{-3}$.

The atomic fluorescence and stray scattered light can be seen in Fig. 6. The scan is triggered by the start of the electron pulse, which forms the ions and causes some increase in detected light. After about 1 second the electron pulse is switched off, the ions are pumped into the $^2S_{1/2}$ ($F = 0$, $m_F = 0$) level, and only stray light is collected. At 2 seconds, the 40.5-GHz radiation is switched on, transferring some of the atoms into the $^2S_{1/2}$ ($F = 1, m_F = 0$) state, where they scatter light as discussed earlier. Finally, at about 3 seconds, the microwaves are switched off and the atomic fluorescence dies away with a time constant of about 1/5 second.

V. Closed-Loop Operation

The sequence of operations used to carry out a measurement of the $^2S_{1/2}$ ($F = 0$, $m_F = 0$) $\rightarrow$ $^2S_{1/2}$ ($F = 1$, $m_F = 0$) frequency is shown in Fig. 7. By repeating this sequence as the frequency of the microwaves is stepped in 0.2-Hz increments, we measure the resonance curve shown in Fig. 8. For the 0.5-second square microwave pulse used in this measurement, the smallest linewidth theoretically attainable is 1.6 Hz, which corresponds to $Q = 2.5 \times 10^{10}$.

Locking the 40.5-GHz oscillator to the 1.6-Hz-wide resonance line is done by stepping the oscillator 0.8 Hz to either side of the resonance and adjusting the center frequency of this ±0.8-Hz step to null the difference in fluorescence rates. More precisely, suppose the center frequency of the 40.5-GHz oscillator, F_i, is within one linewidth of the resonance. Three measurements of the fluorescence are made on alternate sides of F_i: C1 at $F_i + 0.8$ Hz, C2 at $F_i - 0.8$ Hz, and C3 at $F_i + 0.8$ Hz. The oscillator center frequency is then changed to

$$F_{i+1} = F_i + \frac{(0.8 \text{ Hz/T}) (C1 + C3 - 2C2)}{2(\text{SIGNAL})}$$

where SIGNAL is the height of the fluorescence above background and T is the loop time constant in units of measurement cycle time. The "second difference" (C1 + C3 – 2C2) is used because it is insensitive to linear drifts in lamp intensity. If the first difference (C1 – C2) had been used to change F_i, a linear drift in lamp intensity would have forced a linear drift in the 40.5-GHz oscillator's frequency away from the atomic line center. The sequence of center frequencies obtained in this closed-loop operation is shown in Fig. 9. The measurement shown lasted just over 2 hours. A single measurement cycle—consisting of an electron pulse to load ions in the trap, a waiting period while ions are optically pumped, lamp off while microwaves drive the transition, and lamp on and counter on to monitor fluorescence—lasts about 2.5 seconds. The loop time constant, T, is 5 measurement cycles, i.e., about 12.5 seconds.

15

The Allan variance derived from the sequence of frequencies $\{F_i\}$ is shown in Fig. 10. The reference frequency for the 40.5-GHz oscillator is provided by a hydrogen maser (SAO-21 in the Frequency Standards Laboratory). For times that are long compared to the loop time constant, the Allan variance falls as $4.4 \times 10^{-12}/\sqrt{\tau}$. This short-term stability is nearly a factor of 2 better than that of the best commercial cesium standards ($8.5 \times 10^{-12}/\sqrt{\tau}$). These initial results were obtained without shielding the 0.8 G ambient magnetic field in the trap. The residual field dependence at 0.8 G is 157 Hz/G. A frequency stability of 2×10^{-13} at this field sensitivity requires magnetic field fluctuations smaller than 60 μG over the 320 seconds required to reduce statistical error to 2×10^{-13}. For this reason, we have not pushed the Allan variance data beyond 320 seconds in this first test.

VI. Conclusion and Summary

In its first closed-loop operation, the frequency stability of the trapped ^{199}Hg$^+$ frequency standard has been measured to be

$$\sigma_y(\tau) = \frac{4.4 \times 10^{-12}}{\sqrt{\tau}}$$

for $20 < \tau < 320$ seconds.

Many improvements are under way to increase the short- and long-term stability of this standard, including increased fluorescence collection efficiency, shielding the ambient magnetic field, and designs for traps which could store one hundred times the present ion number.

Acknowledgments

References

[1] L. S. Cutler, R. P. Giffard, P. J. Wheeler, and G. M. Winkler, "Initial Operational Experience with a Mercury Ion Storage Frequency Standard," in *Proceedings of the 41st Annual Symposium on Frequency Control*, pp. 12–19, May 1987.

[2] D. J. Wineland, W. M. Itano, J. C. Bergquist, and F. L. Walls, "Proposed Stored ^{201}Hg$^+$ Ion Frequency Standards," in *Proceedings of the 35th Annual Symposium on Frequency Control*, pp. 602–611, May 1981.

[3] M. Jardino, M. Desaintfuscien, and F. Plumelle, "Prospects for a Mercury Ion Frequency Standard," *Journal de Physique*, vol. C8, p. 327, 1981.

[4] F. G. Major, "Remarks on the Application of RF Spectroscopy of Stored Ions to Frequency Standards," in *Proceedings of the FAA Conference on Utilization of Time and Frequency in Collision Avoidance Systems*, pp. 125–126, 1968.

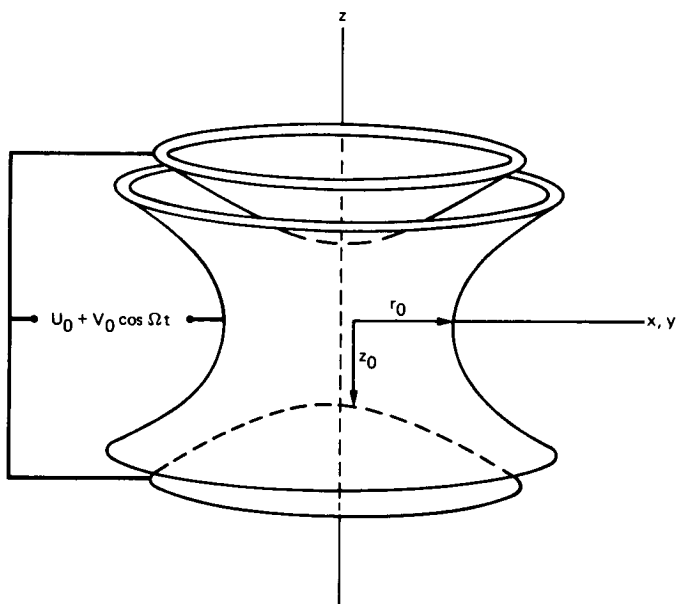[5] H. G. Dehmelt, "Radio Frequency Spectroscopy of Stored Ions," *Adv. At. Mol. Phys.*, vol. 3, pp. 153–154, 1967.

Fig. 1 diagram labels:

z

$U_0 + V_0 \cos \Omega t$

r_0

x, y

z_0

Fig. 1. Electrode structure and voltages used in this work

Fig. 3:

ENERGY, GHz

F = 1

40.507347997 GHz + 97 B^2 (Hz/G^2)

F = 0

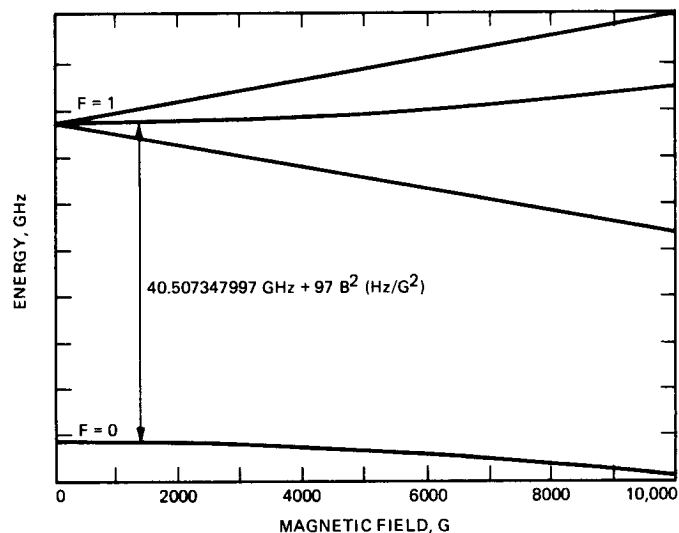0   2000   4000   6000   8000   10,000

MAGNETIC FIELD, G

Fig. 3. Energy levels of the ground state of ^{199}Hg$^+$ versus the magnetic field, showing the first-order field-independent transition used in this work
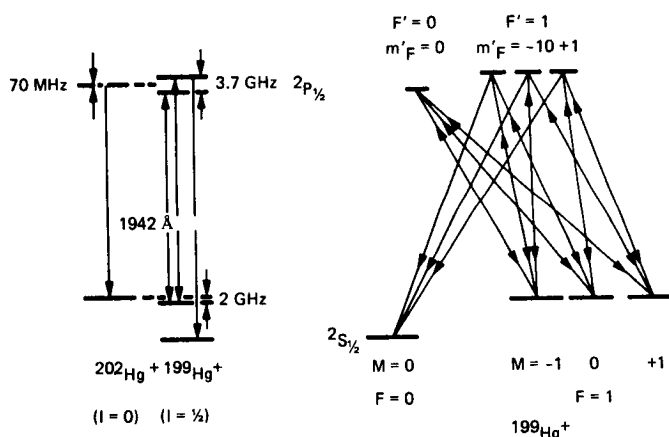
Fig. 2:

POSITION

MASS 199
TRAP FREQUENCY 500 kHz
RF VOLTAGE 650 V
DC VOLTAGE 50 V

TIME

Fig. 2. One-dimensional motion for ion in combined RF and dc trapping fields

Fig. 4:

70 MHz   3.7 GHz $2_{P_{1/2}}$

1942 Å

2 GHz

^{202}Hg + ^{199}Hg$^+$

(I = 0)   (I = ½)

F' = 0   F' = 1
$m'_F = 0$ $m'_F = -10 +1$

$2_{S_{1/2}}$ M = 0   M = -1   0   +1

F = 0   F = 1

^{199}Hg$^+$

Fig. 4. Ground and lowest optically excited states of ^{202}Hg$^+$ and ^{199}Hg$^+$ (left); a depiction of the manner in which light from ^{202}Hg$^+$ pumps ^{199}Hg$^+$ ions into the $^2S_{1/2}$ ($F = 0, m_F = 0$) level (right)
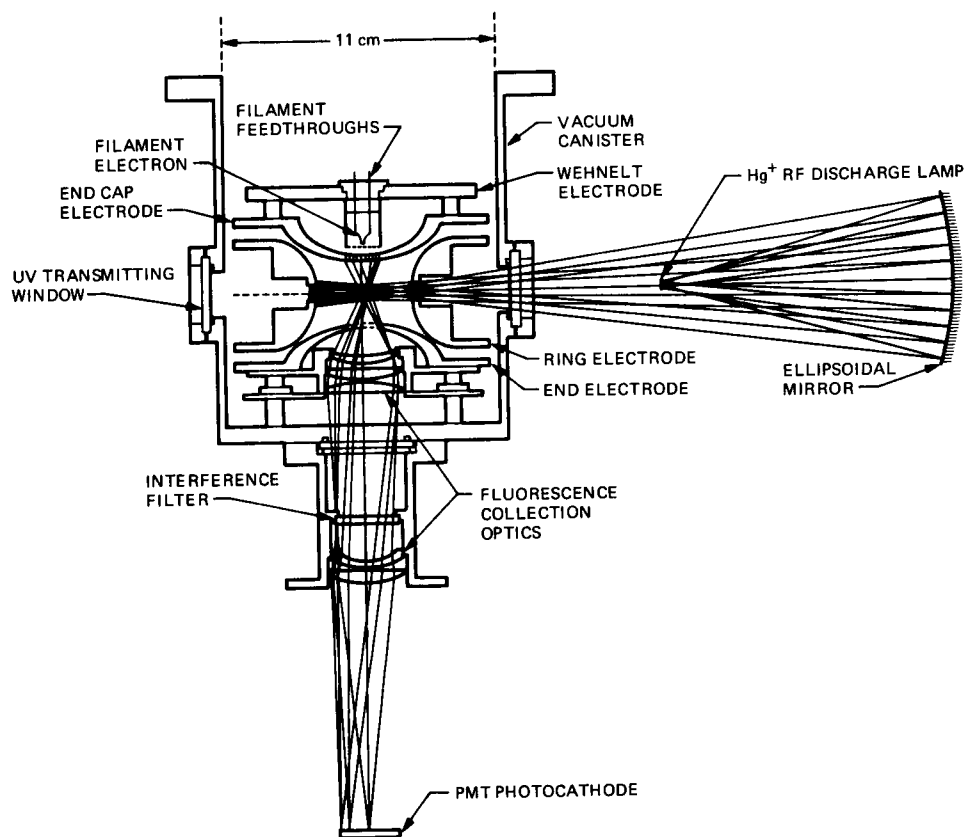
Fig. 5. UV light of 194 nm is scattered by the trapped ^{199}Hg$^+$ ions after the absorption of microwave radiation near 40.5 GHz
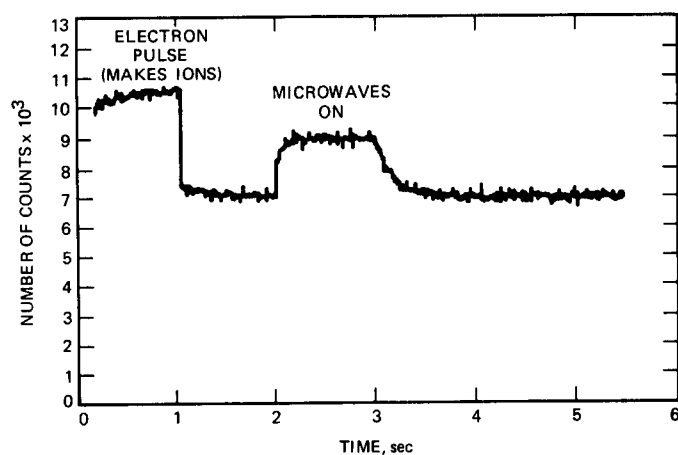

Fig. 6. Light scattered from the ions upon application of microwave radiation (40.5 GHz) is detected together with stray light from electrodes and windows
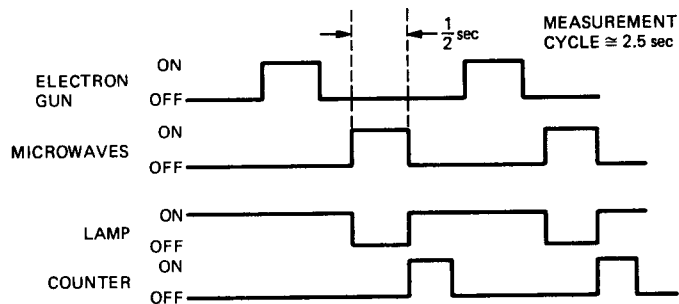

Fig. 7. Sequence of operations used to determine whether the 40.5 GHz is correctly tuned to the atomic resonance (two cycles are shown)
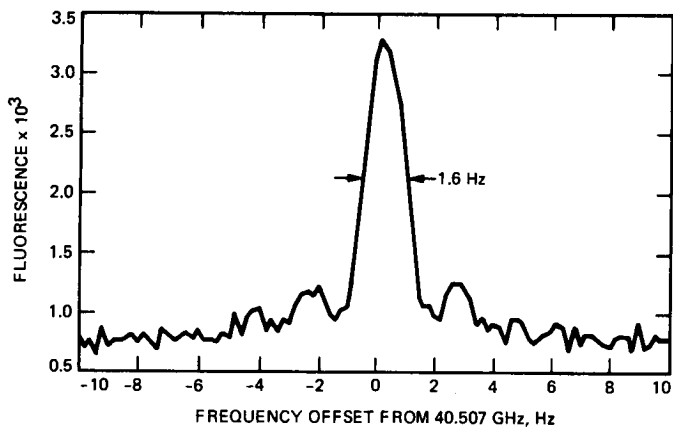
Fig. 8. Atomic fluorescence as the frequency of the 40.5-GHz oscillator is swept through the atomic resonance
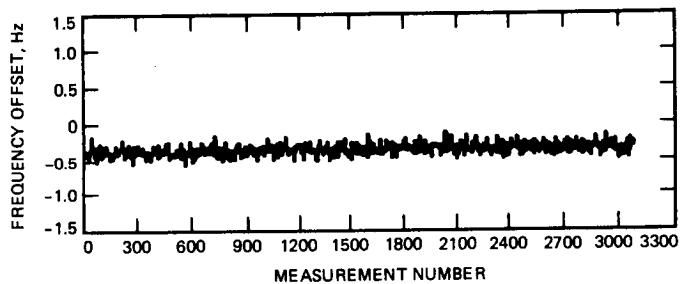

Fig. 9. Frequency deviation of the 40.5-GHz oscillator while servoed to the 1.6-Hz-wide atomic resonance (each measurement lasts 2.5 seconds)
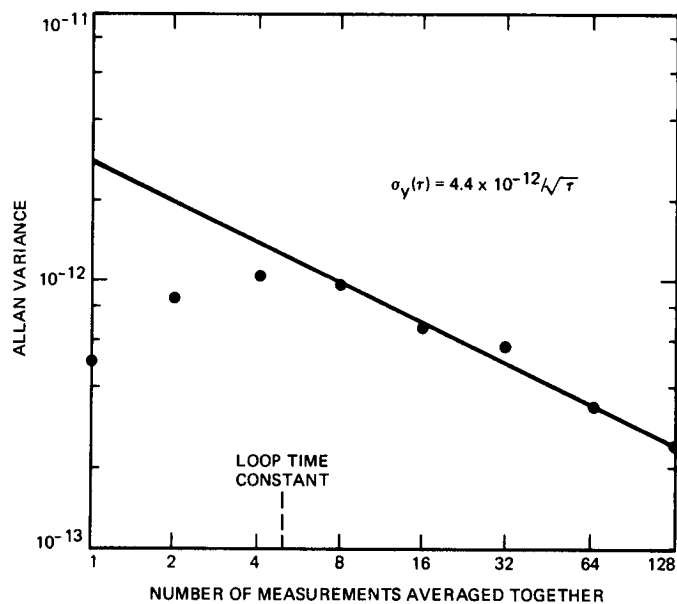

Fig 10. Allan variance of the sequence of center frequencies shown in Fig. 9; multiply by 2.5 seconds to convert the horizontal axis to time