RESULTS OF TWO YEARS OF HYDROGEN MASER CLOCK OPERATION AT THE U.S. NAVAL OBSERVATORY AND ONGOING RESEARCH AT THE HARVARD-SMITHSONIAN CENTER FOR ASTROPHYSICS

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ABSTRACT

Two SAO VLG-11 hydrogen masers have been in service at the U.S. Naval Observatory in September 1983. The systematic frequency drifts have been measured against the U.S.N.O. time scale and their causes identified. The drift is now less than 1×10^{-15} per day.

By operating masers at cryogenic temperatures with wall coatings of CF_4 frozen in place, we have determined that the conventional coatings of the maser hydrogen storage bulbs have 3 times the surface area than is projected geometrically. There are excellent prospects for extending the storage time by a factor of 3 and for reducing the wall shift. Present efforts are underway to realize an atomic hydrogen maser to operate at 0.4K with storage volume coatings of superfluid helium 4. The stability expected of the device is in the 10^{-18} region for intervals beyond 1 hour.

1.0 INTRODUCTION

In the development of atomic clocks two properties are of paramount interest: stability and accuracy. It is clear that accurate measurements cannot be achieved without adequate stability and that the numbers expressing the level of stability are far ahead of those describing accuracy. In this paper we describe the stability behavior, since 1983, of highly stable atomic hydrogen masers at the U.S. Naval Observatory, (USNO) Washington, DC, in relation to the U.S. Naval Observatory Coordinated Universal Time (UTC) time scale derived from an ensemble of about 25 cesium clocks

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carefully monitored against primary cesium standards in various National Standards Laboratories. We will describe work in progress to improve the performance of hydrogen masers and speculate, very conservatively, on the future.

2.0 PRESENT STATUS OF VLG MASER PERFORMANCE

First let us look at where we now stand in regard to frequency stability. Figure 1 shows the latest data in a statistical sense. This is the Allan variance, which is the 1σ probability of the magnitude of frequency change from one interval, τ seconds long, to the next interval of equal duration.

Two noise processes set the limits to stability. First is the additive white noise power, kT per unit bandwidth that accompanies the power from the oscillating H atoms. This spectral distribution gives a τ^{-1} slope in the Allan variance.[1] Second is the effect of thermal noise power within the atomic resonance line width (normally about 1 Hz), which gives a $\tau^{-\frac{1}{2}}$ slope in the Allan variance. The flattening out and rise in the curve after about 10^4 sec is chiefly due to systematic effects such as linear drift of frequency with time.

To measure long-term systematic effects we obviously need a good long-term reference. The best system we know of for long-term time stability is the USNO UTC time scale. Two SAO VLG-11-series masers have been in operation at the USNO since September 1983 and continuous data have been obtained since that date. Figure (2) shows the frequency departure of the masers at the USNO since they were installed. The output frequency of the masers in Hz is represented by the vertical scale. The downward going dotted lines show the frequency shifts that occurred when the cavity resonance frequency of the masers was retuned. The overall drift in frequency since the masers were installed in September 1983 is $\frac{\Delta f}{f} = 2.5 \times 10^{-12}$. The frequency adjustments made to the cavity resonators are shown in Figure (3).

Frequency adjustments have been made using the standard flux-tuning procedure for hydrogen masers. In this procedure the cavity frequency is adjusted so that it has a null frequency pulling effect on the ensemble of oscillating hydrogen atoms.[2,3,4] The frequency pulling effect in its simplest form, excluding the effect of interatomic collisions, can be described as

$$\Delta f_{out} = \frac{Q \text{ cavity}}{Q \text{ atoms}} \Delta f \text{ cavity}$$

The Q of the atoms $= \frac{2\pi f}{2\gamma_2}$ is the ratio of the oscillation frequency to their total rate of loss of phase coherence (T_2 process) owing to the combined effects of removal from the

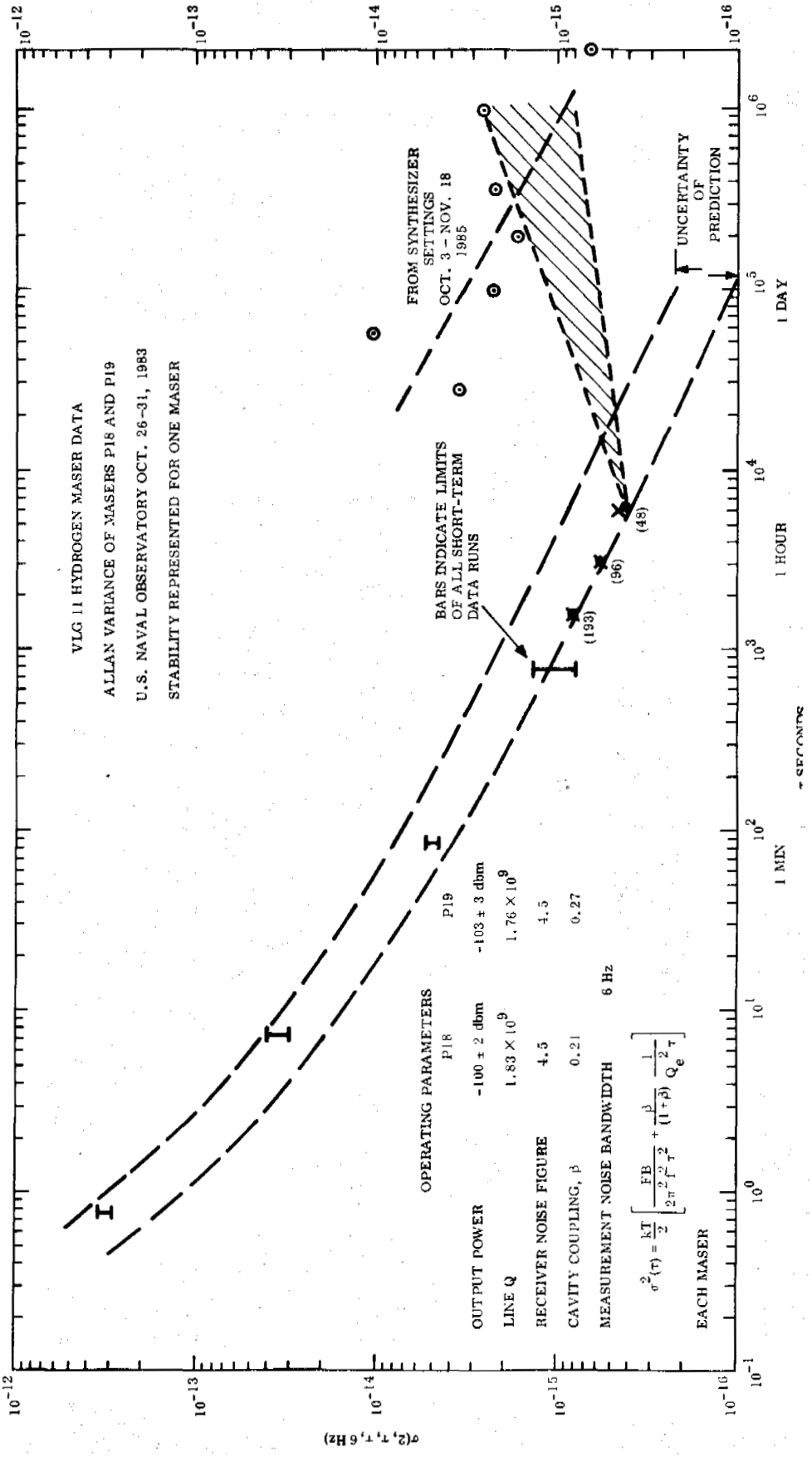


Figure 1. Allan Variance Data taken from SAO VLG-11 masers at the U.S. Naval Observatory including long-term estimates made from synthesizer settings.

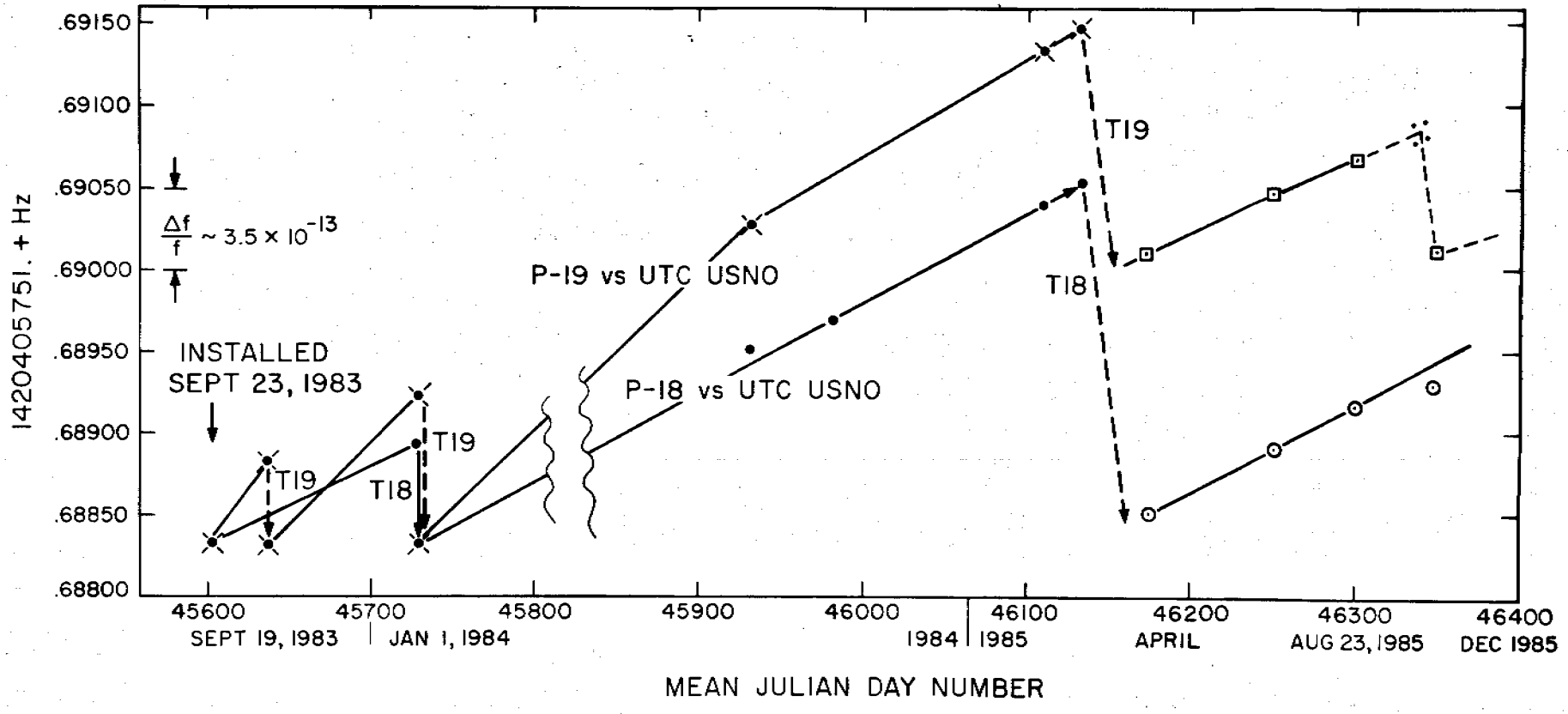


Figure 2. History of frequency drift and resettings of the SAO VLG-11 masers at the U.S. Naval Observatory, September 1983- December 1985.

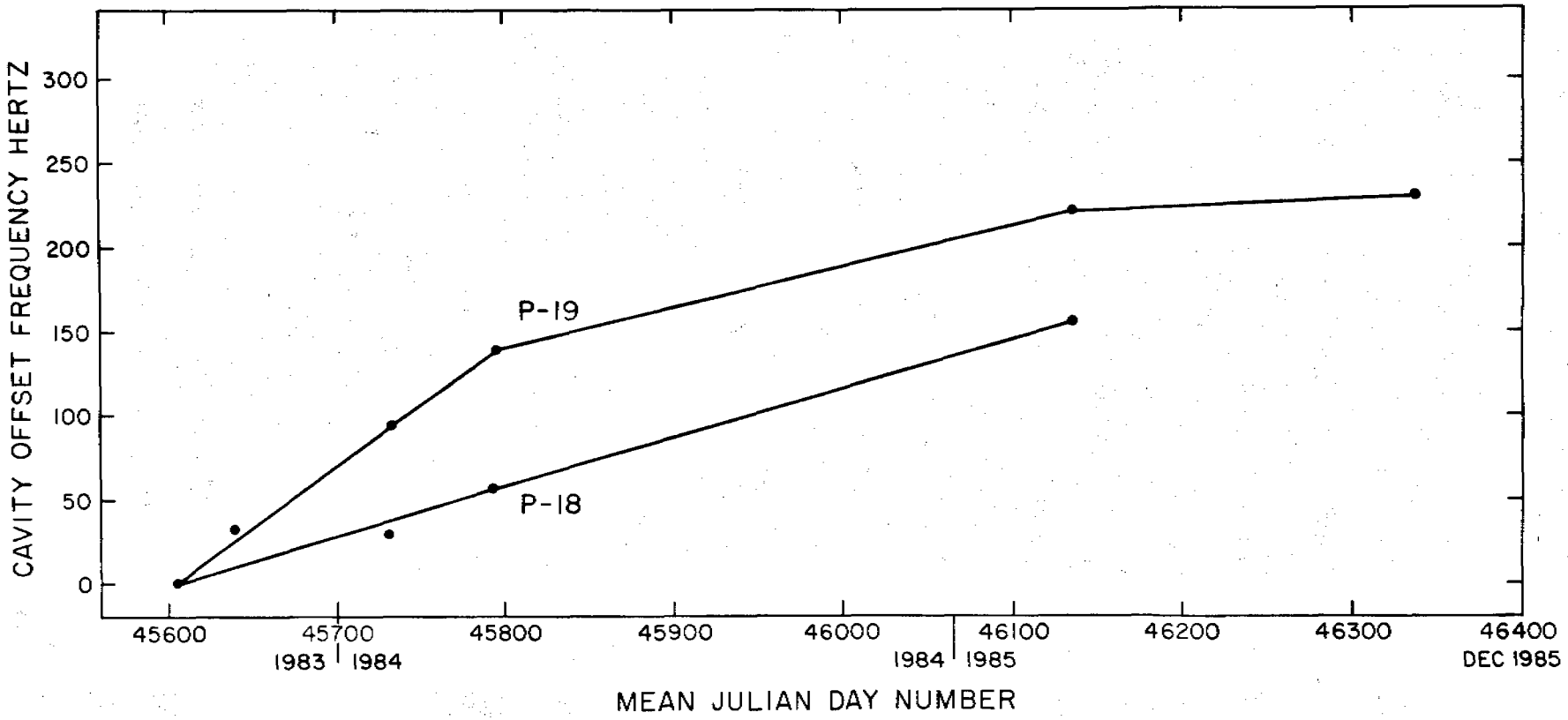


Figure 3. History of cavity frequency adjustments made to SAO VLG-11 masers at the U.S. Naval Observatory September 1983-December 1985.

RF interaction. 1) by emergence from the storage bulb, and 2) by loss of phase coherence, a) from collisions with the wall surfaces and b) by interatomic spin-exchange collisions. To tune the maser we modulate the Q of the atoms by varying the atomic hydrogen flux and adjust the cavity frequency so that no shift in the output frequency, Δf_{out}, is observed.

Maser P-18 has been retuned three times since September 28, 1983, and its frequency has been reset to within 2 parts in 10^{13} of UTC-USNO, the Coordinated Universal Time as maintained by the U.S. Naval Observatory.

In the case for P-19 there was a readjustment of the magnetic field settings on or about MJD 45,800; this resulted in an offset of P-19's frequency of about 1.3 parts in 10^{12} faster as shown by subsequent retuning operations on MJD 46,159 and 46,340, where no further shift was observed.

The cavity offset data, shown in Figure (3), exhibit monotonic increases in cavity resonance frequency that have been found on all other VLG-11-series maser cavity resonators and appear to result from very slow and steady settling of the optically polished surfaces at the mechanical joints between the two end covers and the cylinder, which are clamped together with a force of about 1300 Newtons. These parts are made of CER-VIT, a glass-ceramic material that has excellent mechanical stability and a very low coefficient of linear expansion, $|\alpha| < 5 \times 10^{-7}/^{\circ}C$. The total cavity frequency shift to date is about 230 Hz and represents a total excursion of about 0.2 microns (2000Å) in the position of the end covers. The rate of settling since MJD 45,800 is about 2.8Å per day. Some evidence of levelling out of this rate is shown by P-19's behavior. This follows our experience with other masers where, after about 3 years, very little cavity drift is seen. Figure (4) shows the daily rate of drift of the masers with respect to UTC-USNO and to each other. The rapid decrease in drift rate right after installation is the result of the warm-up of the masers and their thermal stabilization.

The two masers operating at the USNO provide redundancy by being operated so as to provide a synchronized output signals. This phase coherence is obtained by automatically adjusting the synthesizer frequency of one maser so that a phase difference of less than 10 picoseconds is observed in 1.2 GHz signals synthesized by each maser system.[5] Adjustments are made in steps of 10 microhertz and by recording these and the times at which they are made, we can obtain a measure of the long-term relative stability. Because the steps are each 7.1×10^{-15} in $\frac{\Delta f}{f}$, only long-term averages will reflect the maser stability. The data at 10^5-10^6 second region of Figure (1) are made from synthesizer settings and the shaded area is a good estimate of the maser's statistical behavior for time intervals beyond 10^4 seconds.

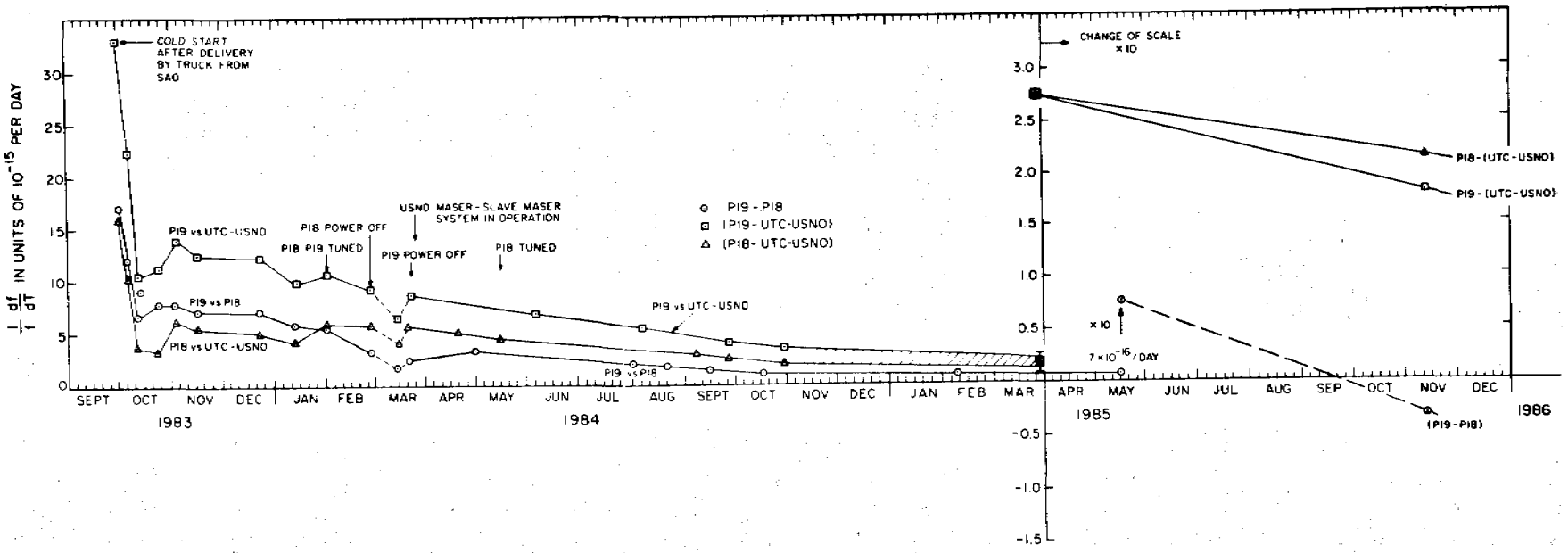


Figure 4. Daily rate of frequency drift of the SAO VLG-11 masers at the U.S. Naval Observatory, September 1983 - December 1985.

In summary, then, we can say that the stability of the VLG-11 masers at the U.S.N.O. is as given in Figure (1) and that the day-to-day frequency uncertainty is about 2×10^{-15}. We believe that the drift is predictable to within about 5×10^{-15} over periods of several days to a week in duration.

3.0 IMPROVEMENTS TO STABILITY

3.1 Production Of A Beam Of Atoms In A Single Hyperfine Sublevel

The conventional hydrogen maser employs a state selection system that focuses both upper hyperfine magnetic sublevels of the F=1 state of atomic hydrogen into the maser storage bulb. Half of these are unwanted and cause unnecessary spin exchange line broadening, and under certain conditions of magnetic inhomogeneity can cause frequency shifts. The benefits of obtaining a population of atoms purely in the F=1, m_F=0 state have been discussed and demonstrated by Lacey and Vessot,[6] Audoin, et.al.[7] and Urabe et.al.[8] with varying degrees of success. Our approach is shown schematically in Figure (5) showing the hyperfine energy structure of the atoms as a function of magnetic field at various stages in the beam. The vertical axis is greatly magnified to show deflections. We believe the focussing system provides a more effective atomic hydrogen state-selection filter than those previously used. Atoms emerge from the hexapole on the left with their two upper magnetic hyperfine sublevels m_F=1,0 focussed into a beam. The section labelled A.F.P. (for adiabatic fast passage) [9] contains an axial d.c. magnetic field that varies spatially along the beam axis. Within the region of d.c. magnetic field gradient we apply a transverse oscillating magnetic field at the Larmor frequency of an atom situated at the center of the d.c. coil. The result is that as the atom passes through this system its axis of quantization is reversed in direction and the m_F=+1 state becomes the m_F=-1 state; the m_F=0 state is undisturbed. We then pass the beam through a second magnet, roughly twice the length of the first, which permits only atoms in the m_F=0 state to reach the bulb.

The system hardware is shown in Figure (6). At the left is the atomic hydrogen dissociator, the first magnet, the coil system, the second magnet, and, at the right, the atomic hydrogen storage bulb. By operating the hydrogen maser with the same flux of atoms into the bulb as before, but in a single state, instead of a beam with two states, we anticipate a decrease in the spin-exchange quenching rate of 50% and a corresponding doubling of the power output. While this will improve the stability shown in Figure (1) by a factor of $2^{-\frac{1}{2}}$, its main purpose is to remove the frequency

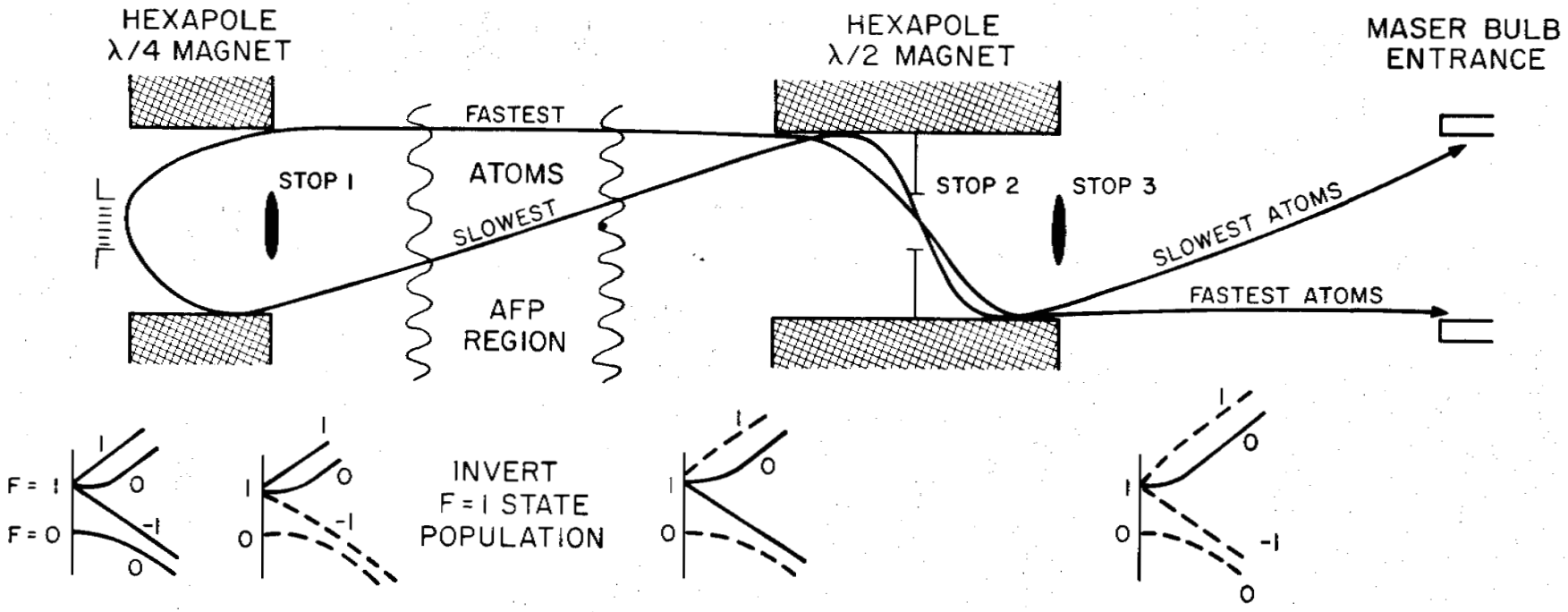


Figure 5. Magnetic focussing scheme for isolating the F=1, m_F=0 hyperfine state in the atomic hydrogen beam entering the maser storage bulb.

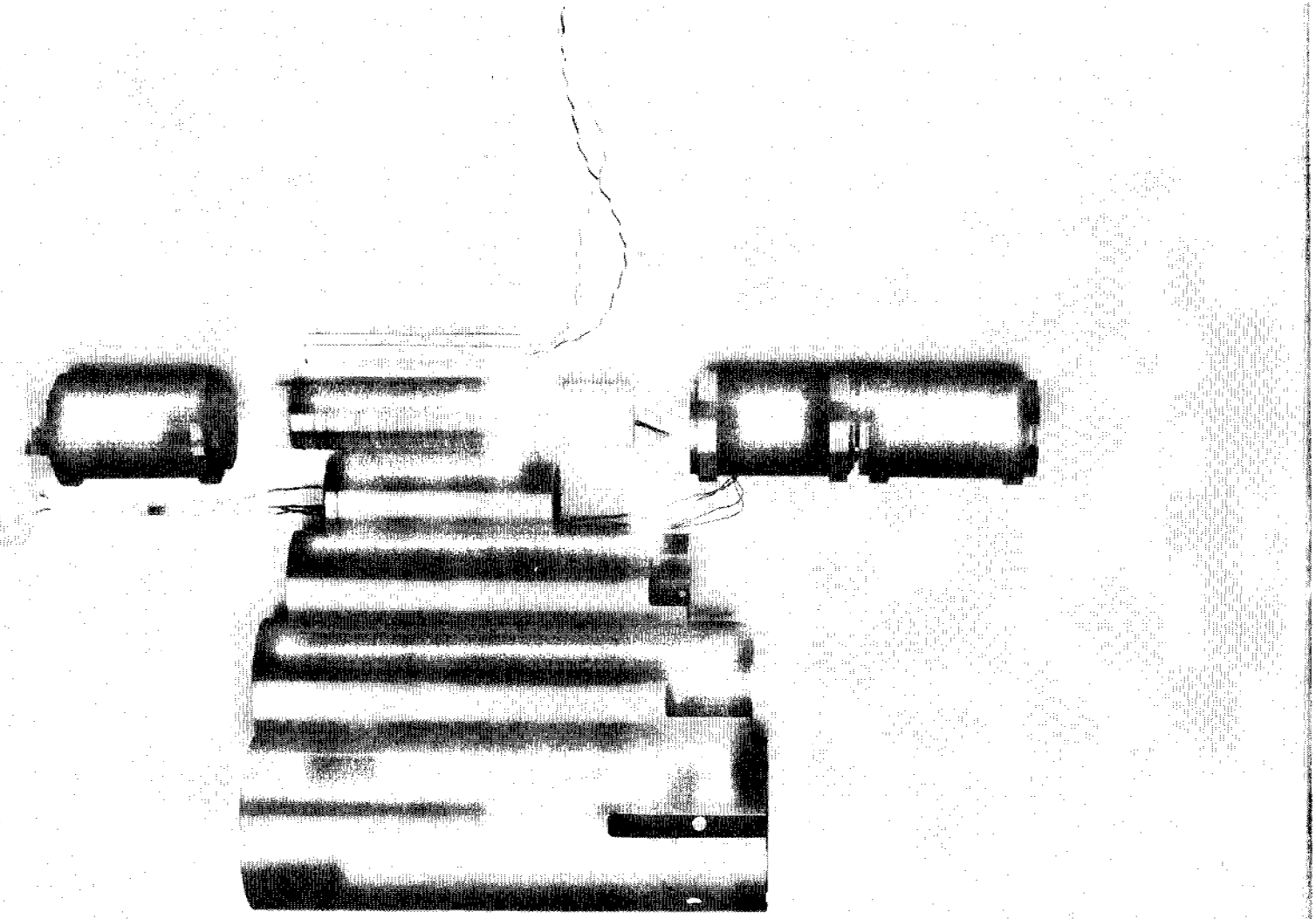


Figure 6. Photo of hardware used to implement the adiabatic fast passage technique for improved atomic hyperfine state selection.

shift resulting from spin-exchange collisions among atoms distributed asymmetrically in the ± 1 states about the m_F=0 state. This effect, discovered by S. Crampton,[10] at present is the chief source of systematic frequency shifts owing to changes in the external magnetic environment.

3.2 Maser Wall Coatings At Low Temperatures

Another aspect we are working on is related to the capability of the surfaces of the storage volume to reflect the impinging hydrogen atoms without perturbing their atomic state and, more stringently, without disturbing the phase of the oscillating magnetic dipole moment resulting from the spin-spin interaction of the proton and electron.

In our recent work[11] on low temperature hydrogen masers with storage volume surfaces of CF_4 frozen-in-place from the gaseous phase we have made measurements of the interaction energy from wall collision frequency shift data made over temperatures down to about 40°K. We found, as expected, that the interaction energy for Teflon, which is a fluorocarbon polymer, and for CF_4 were quite similar, as seen in the slopes of the plots shown in Figure (7).

The theoretical basis for this representation of the data is described in a paper by Hardy and Morrow[12] that describes the frequency shift owing to the wall collision effects in terms of the time spent on the surface during the wall interaction, which is governed by the atom-to-surface binding energy and the hyperfine frequency shift while in a surface energy state.

The expression

$$-T\Delta\phi(T) \text{ vs. } \frac{A}{V}\frac{V_g}{A_g}\frac{h}{k}2\pi\Delta_s e^{\ Eb/kT}$$

relates the phase shift per collision $\Delta\phi$, as determined from the geometrical volume to areas ratio, V_g/A_g, obtained from the storage volume geometry to temperature, T, and energy of interaction Eb. Here h is Planck's constant, k is Boltzmann's constant and Δ_s is the hyperfine frequency shift of atomic hydrogen when it is bound to a surface. The value of $\Delta\phi$ is calculated from measurements of the wall collision frequency shift, $\Delta\nu_w$, using the expression

$$\Delta\phi_w = \frac{1}{2\pi}\frac{\Delta\nu_w}{f_c}$$

where f_c is the wall collision rate as determined geometrically from

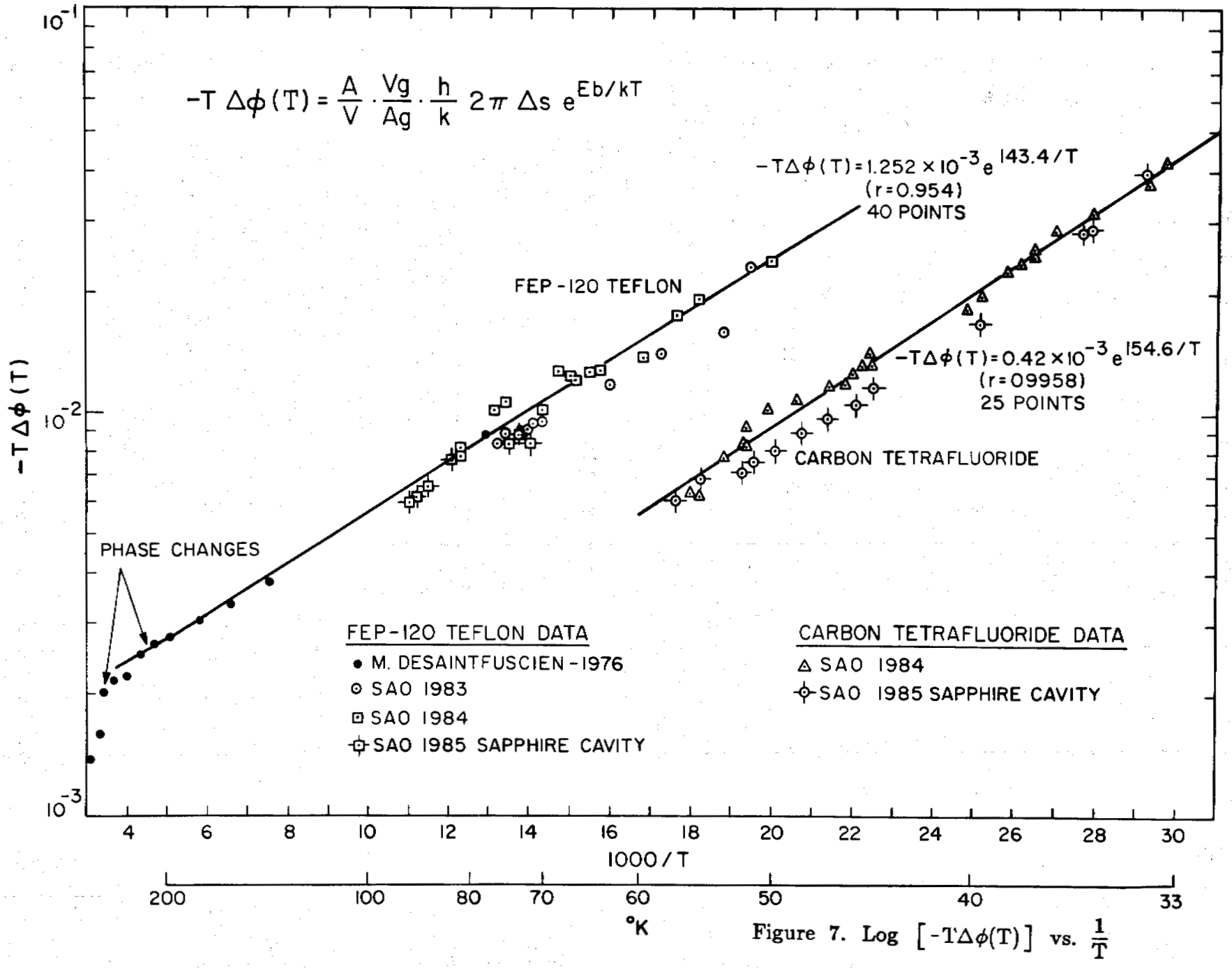


Figure 7. Log $\left[-T\Delta\phi(T)\right]$ vs. $\frac{1}{T}$

$$f_c = \frac{\text{average velocity}}{\text{mean free path}} = \frac{\bar{v}}{4V_g/A_g}.$$

Here $\bar{v} = \left(\frac{8kT}{\pi m}\right)^{\frac{1}{2}}$

The great surprise to us was that the wall shift was about four times less than we had anticipated. From these results and other experimental evidence obtained from x-ray photoelectron spectroscopy studies of our Teflon surfaces made at the U.S. Naval Research Laboratory[13], we conclude that the Teflon is very rough in texture and porous to atomic hydrogen. The collision rate of hydrogen on Teflon appears to be about four times the rate predicted from the cavity dimensions. We visualize that by freezing CF_4 over the existing Teflon we fill the crevices and gaps and by smoothing the surface we reduce the collision rate. We are looking into improving our process for coating bulbs.

We foresee a factor of 4 improvement on the line Q of the maser if we can realize at room temperature the surface texture we have at low temperature with CF_4. This means that the $\tau^{\frac{1}{2}}$ curve, which now goes down to a few parts in 10^{-16} in $\frac{\Delta f}{f}$, should get into the high 10^{-17} range.

3.3 A Hydrogen Maser Operating At 0.5K

The most exciting prospect we have for making a breakthrough to get oscillator stability in the 10^{-18} range is to operate at very low temperatures, where there are many advantages. The atom moves more slowly, thus making fewer collisions with the walls. The cross section for spin exchange is reduced by a factor of about 200 when we cool atomic hydrogen to about $4^{o}K$; this enables a substantially larger number of atoms to interact with the rf field without perturbing each other. The superconducting qualities of various materials can be used to good advantage to control magnetic field and temperature. The properties of materials are far more stable when temperatures approach absolute zero.

From the point of view of the signal quality we see that the thermal noise per unit bandwidth kT can be drastically reduced. This works directly on the fundamental limit of maser stability given by

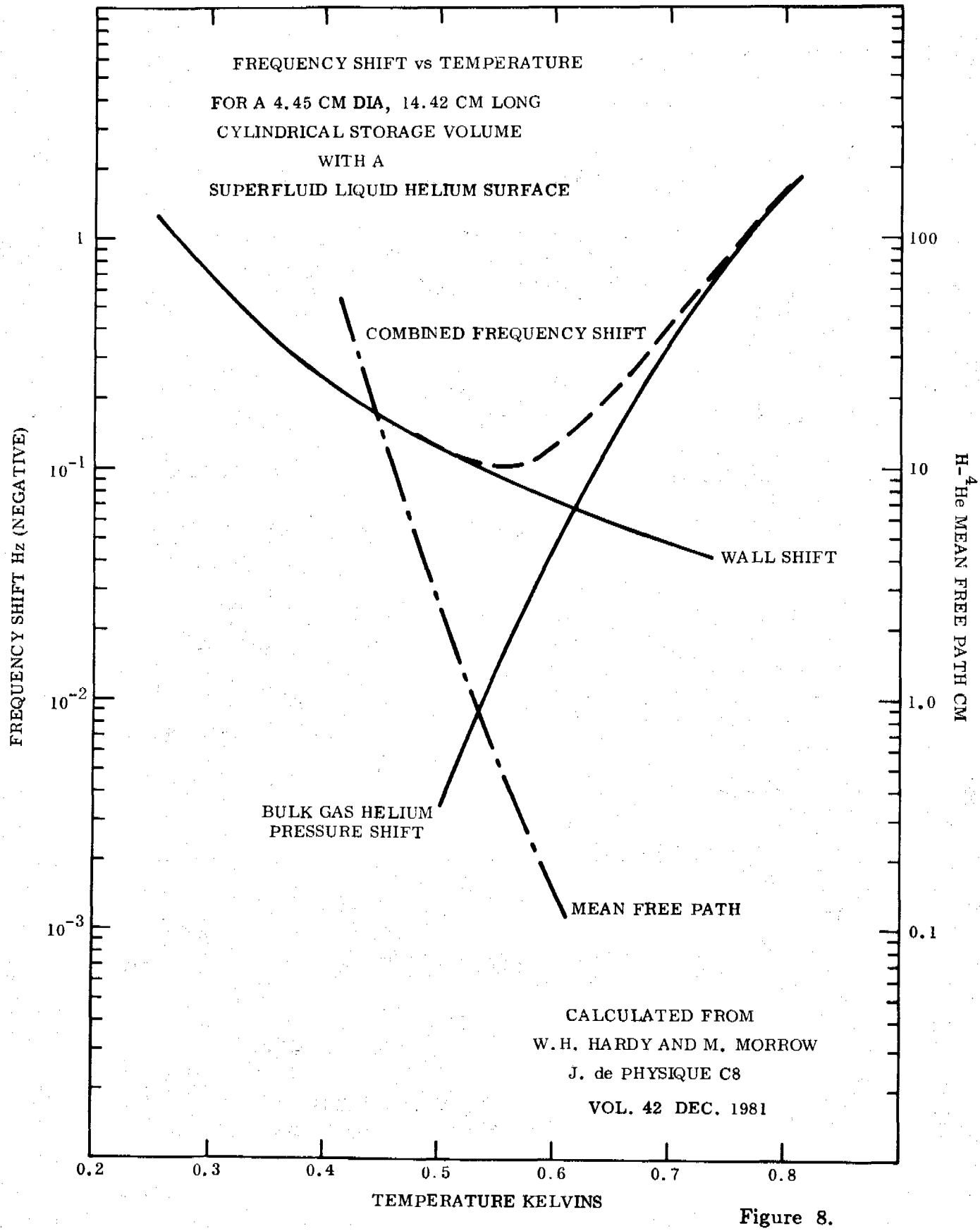
$$\frac{\Delta f}{f_o} = \frac{1}{Q}\left(\frac{kT}{P_b \tau}\right)^{\frac{1}{2}}$$

and on the more practical limit of signal to noise in the electronics system

$$\frac{\Delta f}{f_o} = \frac{1}{2\pi f_o \tau}\left(\frac{FkT}{P_o}\right),$$

where Q is the effective line Q of the atomic resonance, P_b is the power delivered to the receiver electronics, T is the physical temperature of the system, and F is the system's noise figure. Obviously, this whole low temperature scenario depends on the behavior of wall surfaces at low temperatures. We found that CF_4 works down to about 30K but, as has been shown in Figure (7), it has a very large coefficient of frequency shift with temperature and, while interesting as a way of studying wall surfaces, it is not a good coating for making a high stability oscillator.

However, a breakthrough in wall coatings has come from a totally different direction, the theoretical and experimental work in stabilizing a high density atomic hydrogen gas in high magnetic fields at temperatures low enough for Bose-Einstein condensation to occur. In 1980 Silvera and Walraven[14] reported that wall surfaces coated with superfluid ^{4}He would allow storage of polarized atomic hydrogen under magnetic field compression at densities greater than 1.8 x $10^{14}/cm^3$ at 0.27°K for times greater than 500 seconds. In 1980 Hardy, Morrow, and Jochemsen[15] reported magnetic studies showing that, besides being able to store atoms at densities appropriate for maser oscillation for about 6 months, there is a fortunate temperature behavior in the use of superfluid helium wall coatings. The H-He bulk gas collision hyperfine frequency shift, which dominates at higher temperatures owing to the rapid rise in vapor pressure of helium with temperature, and the H-He surface collision shift, which increases as temperature is lowered, when combined have a temperature at which there is a null in $\frac{\Delta f}{\Delta T}$. The behavior of these effects is illustrated in Figure (8) for a cylindrical volume 4.5 cm in diameter and 14.4 cm long. We see that there is a saddle point, which we estimate to have second-order frequency curvature with temperature $f(T) = 4T^2$. To maintain fraction frequency stability $\frac{\Delta f}{f} \sim 10^{-18}$ implies that ΔT must be held about 20μK, and this is well within the capability of modern low temperature technology[16]. Our expected stability behavior is shown in Figure (9).


FREQUENCY SHIFT vs TEMPERATURE
FOR A 4.45 CM DIA, 14.42 CM LONG
CYLINDRICAL STORAGE VOLUME
WITH A
SUPERFLUID LIQUID HELIUM SURFACE
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BULK GAS HELIUM
PRESSURE SHIFT
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Figure 8.

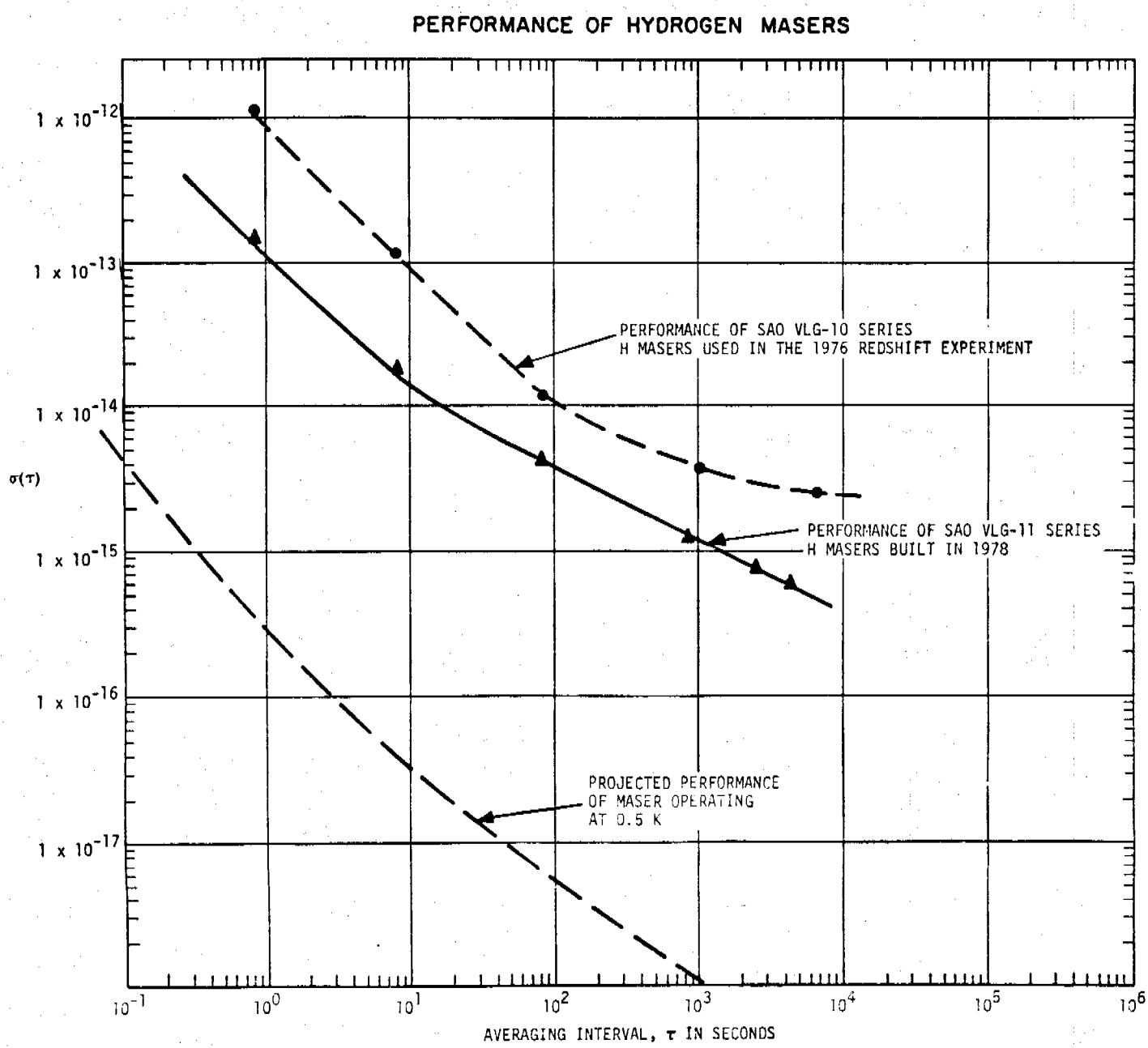

Figure 9. Projected performance of an atomic hydrogen maser operating near 0.5K.

4.0 CONCLUSIONS

Even though the conventional hydrogen maser can now probably be included in the category of maturing technology, there is still ample room for improvement through use of better wall coatings and by more efficient state selection techniques. The potential for a breakthrough in both stability and accuracy may be realized by very low temperature operation using wall surfaces of liquid helium. It may be time to think of what we can do with oscillators having fractional frequency stability well below 1×10^{-16}.

5.0 ACKNOWLEDGEMENTS

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6.0 REFERENCES

1. Vessot, R.f.C., "Frequency and Time Standards in Methods of Experimental Physics," Volume 12, Part C: Radio Observations, M.L. Weeks (Ed.), Academic Press, NY, 1976. pp198-227.

2. Kleppner, D., H.M.Goldenberg, and N.F. Ramsey, Phys. Rev. Lett. 126, 603 (1962)

3. Kleppner, D., H.C.Berg, S.B. Crampton, N.F. Ramsey, R.F.C. Vessot, H.E. Peters, and J. Vanier., "Hydrogen maser principles and techniques." Phys. Rev. Lett. 138 A972 (1965)

4. Vessot, R.F.C., "Proceedings of the Third Marcel Grossmann Meeting on General Relativity, Hu Ning ed. Science Press and North-Holland Publishing Co., pp 803-821 (1983).

5. Klepczynski, W.R., In "Satellite Transmissions," Proceedings of an International Symposium. Graz, Austria, 25 Sept. 1985. E.J. Rolfe, (ed.), Published by the European Space Agency, p 165.

6. Lacey, R.F. and R.F.C. Vessot. Proceedings of the 23rd Annual Symposium on Frequency Control, 6,7,8 May 1969. U.S. Army Electronics Command, Fort Monmouth, N.J., pp 279-283.

7. Audoin, C., M. DeSaintfuscien, P. Petit, and J.P. Schermann, IEEE Trans. on Instrumentation and Measurement. IM-17, 4, (1968), pp 351-353.

8. Urabe, S., K. Nakagin, Y. Ohta, M. Kobayashi and Y. Saburi, IEEE Trans. on Instrumentation and Measurement. IM-29, 4 (1980) pp 304-310.

9. Ramsey, N.F., Molecular Beams, Oxford University Press, 1956. pp 149.

10. Crampton, S.B., and H.T.M. Wang, Phys. Rev. Lett. A, 12 4, 1305, (1975).

11. Vessot, R.F.C., E.M. Mattison, E.A. Imbier, Z.C. Zhai, Proceedings of the 16th

Annual Precise Time and Time Interval Meeting, NASA Goddard Space Flight Center, Greenbelt, MD. Nov. 27-29, 1984., pp 357-374.

12. Hardy, W.N. and M. Morrow, Journal de Physique 42 (1981) C3-171.

13. Turner, N.H., Proceedings of the 9th Annual Precise Time and Time Interval Applications and Planning Meeting. NASA Goddard Space Flight Center, Greenbelt, MD, Nov. 29- Dec. 1, 1977 (NASA Technical Memorandum 78104)-11371-350.

14. Silvera, I.F. and J.T.M. Walraven, Phys. Rev. Lett. 44 (1980) 1964.

15. Morrow, M., R. Jochemsen, A.J. Berlinsky, and W.N. Hardy, Phys. Rev. Lett. 46 (1981) 195, also Phys. Rev. Lett. 47 (1981) 455E.

16. Lipa, J.A., Physica 107 B+C (1981) 343