

VARIABLE VOLUME MASER TECHNIQUES

Victor S. Reinhardt
NASA/Goddard Space Flight Center
Greenbelt, Maryland 20771

INTRODUCTION

The hydrogen maser achieves its extremely good frequency stability principally by confining the masing atoms for long periods in a teflon coated storage bulb.¹ Because of this confinement, the frequency of the hydrogen maser is shifted due to collisions of the atoms with the walls of the storage bulb. This is shown in Figure 1. The wall shift is given by:

$$f = \frac{\phi v}{2\pi} \cdot \frac{A}{4V}$$

where ϕ is the average phase shift per collision, v is the average atomic speed, and $(4V/A)$ is the mean free path between wall collisions, λ .

In order to use the hydrogen maser as a primary frequency standard, one must correct for the wall shift. The obvious method to do this is to vary λ by operating hydrogen masers with different size storage bulbs, and to make frequency comparisons between the masers. This has been done many times², but has not produced good results because ϕ has not reproduced well from bulb to bulb.

In 1969, Douglas Brenner proposed another approach.³ If one were to make a maser with a flexible storage bulb, one could vary λ but keep ϕ constant. This would allow for the correction of the wall shift without the uncertainties associated with changing bulbs. The technique is outlined in Figure 2. This method has been a fruitful one, leading to several devices and increasing degrees of success. This paper will discuss this variable volume technique and the strengths and weaknesses of the devices which have been based on this technique.

THE VARIABLE VOLUME TECHNIQUE

The first variable volume maser was built by Douglas Brenner.⁴ He used several types of flexible bulbs with a variety of wall coatings. One of his teflon bulbs is shown in Figure 3. As shown in Figures 1 and 2, Brenner's technique relies on one's knowledge of B , the ratio of the bulb volumes, and the fact that both the value of ϕ and the value of f_0 , the corrected frequency, remain constant

during the change of volume. With careful measurement, B can be determined to the 0.1% level.^{4,5} How this effects the error, however, is strongly dependent on the size of B . As B approaches unity the uncertainty in determining f_0 goes to infinity for given errors in measuring B , f_1 , and f_2 . The size of B in Brenner's device is limited severely by the fact that the filling factor¹ is degraded in the compressed volume. Brenner achieved values of 1.18 to 1.37 for B .

The value of ϕ is effected by both changes in the atomic density in the bulb and changes in the surface properties of the bulb. If the relative rate at which atoms strike the bulb surface change when one changes the volume, ϕ may change. In a bulb with good communication between sections, this effect can be made negligably small.^{4,5,6} Changes in the surface properties of the teflon during the measurement process can be induced by the changes in stress that occur when the bulb volume is changed. Brenner discussed this problem⁴ and estimated that the stress effect would change ϕ by 0.25%. The major weakness of his device, however, is in the uncertainty caused by lack of knowledge of the effects of stress on his storage bulbs.

Changes in f_0 during the measurement process come from shifts in the maser frequency other than the wall shift: the doppler shift, magnetic shifts and spin exchange shifts. It has been determined that to one part in 10^{14} of the hyperfine frequency the atomic velocities are thermalized to the bulb temperature.⁷ One therefore can correct for the doppler shift by measuring the bulb temperature.

The principal magnetic shift can be corrected for by measuring the zeeman frequency.¹ Magnetic inhomogeneity shifts⁸ can cause errors as large as parts in 10^{12} . These shifts are a function of the average inhomogeneity over the storage bulb and so change when the storage bulb volume is changed. Inhomogeneity shifts can be corrected for,^{8,9} but unfortunately Brenner was unaware of these shifts when he performed his measurements, so he did not correct for them.

Spin exchange shifts come from collisions of the radiating hydrogen atoms with other hydrogen atoms and with other paramagnetic gases.^{1,8,10,11} The principal part of the hydrogen spin exchange shift is corrected out with flux tuning.¹ An anomalous part is not.¹¹ This shift is 4×10^{-4} of the non-hydrogen spin exchange part of the atomic linewidth.^{8,11} In a variable volume maser, except for contributions from magnetic inhomogeneities and from background gases, the non-spin exchange linewidth is proportional to the inverse bulb volume just as the wall shift is,¹² so most of the anomalous spin exchange shift is corrected for by the same process which measures the wall shift. The background

contribution to the linewidth can be kept small by keeping the partial pressure of paramagnetic gases small,⁸ and the magnetic linewidth can be kept small by reducing the size of magnetic inhomogeneities. One should therefore be able to keep uncertainties due to the anomalous spin exchange shift below the 10^{-14} level.

VARIABLE VOLUME DEVICES

To overcome the stress problems associated with the Brenner flexible bulb, Norman Ramsey proposed using a thin flexible teflon cone attached to a rigid cylinder as the variable volume storage bulb. This idea was implemented by Pierre Debely¹³ and is shown in Figure 4. The great advantage of this configuration is that a thin cone can be inverted in such a way that only the edges of the cone are stressed. This reduces the region of possible stress effects to a negligibly small area, eliminating the uncertainty due to surface stressing. The volume ratio achieved by Debely was limited to 1.3 because of difficulties in obtaining maser oscillation with the cone inverted. The accuracy of the Debely device was limited to 2.4×10^{-12} because of the small value of B, drifts in the reference maser, and areas in the cone which became exposed when the cone was inverted. The asymmetrical bulb also made Debely's device especially susceptible to magnetic inhomogeneity shifts which he did not consider.

To overcome the problems associated with the Debely device, Norman Ramsey proposed combining the flexible cone with the large storage box hydrogen maser.¹⁵ This was implemented by the author^{12,14} and is shown in Figure 5. In this device, the flexible cone is outside the microwave cavity, so the size of B is not limited by oscillation requirements. Magnetic inhomogeneity problems are also reduced. Because the device has a linewidth a factor of ten narrower than a conventional hydrogen maser, anomalous spin exchange effects are correspondingly reduced. The present configuration of the device achieved an accuracy limit of 2.4 parts in 10^{13} , but this was due to a large measurement uncertainty for B and a long term stability floor of a part in 10^{13} caused by the necessity of using an amplifier in the maser feedback loop. With passive maser techniques,¹⁶ an accuracy of a part in 10^{14} should be achievable.

Another device which shows much promise is the concertina hydrogen maser developed by Harry Peters.⁶ The device is shown in Figure 6. In this device, the variable volume storage bulb is a flexible bellows of teflon film. There are stress effects, but to first order they cancel due to the bellows configuration. This device also has the great advantage of allowing measurement over a continuous range of calibrated volumes. Stress effects will be determined by curve fitting the frequency shift data and by using bellows of differing thickness. The assymetrical arrangement of the storage bulb in the microwave cavity makes

this device very susceptible to magnetic inhomogeneity problems, but these can be measured⁹ and calibrated out for each volume setting. Preliminary measurements with the concertina maser have been consistent with hydrogen hyperfine measurements using the flexible bulb-large storage box maser,¹⁷ and with a recent compilation of wall shift measurements.¹⁸ A very interesting side result of this measurement is that at 40°C, ϕ for type L FEP film¹⁹ is a factor of four smaller than ϕ for FEP teflon sintered on a quartz storage bulb.

FURTHER DEVELOPMENTS

In 1970, Paul Zitzewitz and Norman Ramsey^{20,22} discovered that the wall shift goes through zero at approximately 100°C. This was later verified by Robert Vessot and Martin Levine.^{21,22} Based on this, a device using a variable volume storage bulb as a null detector for the zero wall shift point was proposed by Vessot and Levine.^{21,22} The great advantage of this device is that one need not know B accurately to calibrate out the wall shift, and because one actually operates with a zero wall shift, the device can be operated in an automated fashion.²²

One can generalize this idea to take advantage of dropping the necessity of measuring B without having to go to the zero wall shift temperature. This generalization is demonstrated in Figure 7. Since the value of ϕ changes with temperature, one need only change the temperature of a variable volume device while making measurements at the same two volumes. By extrapolating to the point where the curves for each temperature intersect, one can determine the zero wall shift point without actually reaching it. As discussed previously in the context of determining stress effects,⁶ if one makes measurements at at least three temperatures, by the scatter of the intersection points, one can estimate the errors due to assumptions of constant ϕ or due to changes in f_0 .

One need not even have a linear measure of the inverse volume or λ^{-1} for the method to work. Since the wall shift is homogeneous in ϕ , for any monotonic function $X(\lambda^{-1})$, the intersection point of a family of curves in ϕ can only occur at zero wall shift. Also because the wall shift is homogeneous in ϕ , one can make a single point transformation $\lambda^{-1}(X)$ where X is the measurement parameter such that the frequency change linear in λ^{-1} . One can therefore arbitrarily mark two points, X_1 and X_2 , on the ordinate axis of a graph and linearly extrapolate to $\lambda^{-1} = 0$ which is experimentally determined by the intersection of the straight lines.

Based on this method and the knowledge gained from previous devices, a new variable volume device is being developed at NASA's Goddard Space Flight Center. It is shown in Figure 8. The device will have a flexible cone variable

volume element outside the cavity to avoid the problems associated with a variable storage bulb inside the microwave cavity. It will also not have any electronic feedback to avoid the instabilities associated with amplifiers in the feedback loop. Figure 9 demonstrates that a reasonable external volume can be achieved without electronic feedback. The extra filling factor obtained with the elongated cavity design used in NASA masers has proved instrumental in achieving this. The plans are to operate this device either as a zero wall shift maser or in the generalized mode just described. Recent experiments have shown that FEP teflon film, after a bake out at 120°C, has a low enough vapor pressure at 100°C to allow a zero wall shift maser to function. With this device and future developments with some of the others outlined here, the promise of 10^{-14} accuracy should become a reality in the near future.

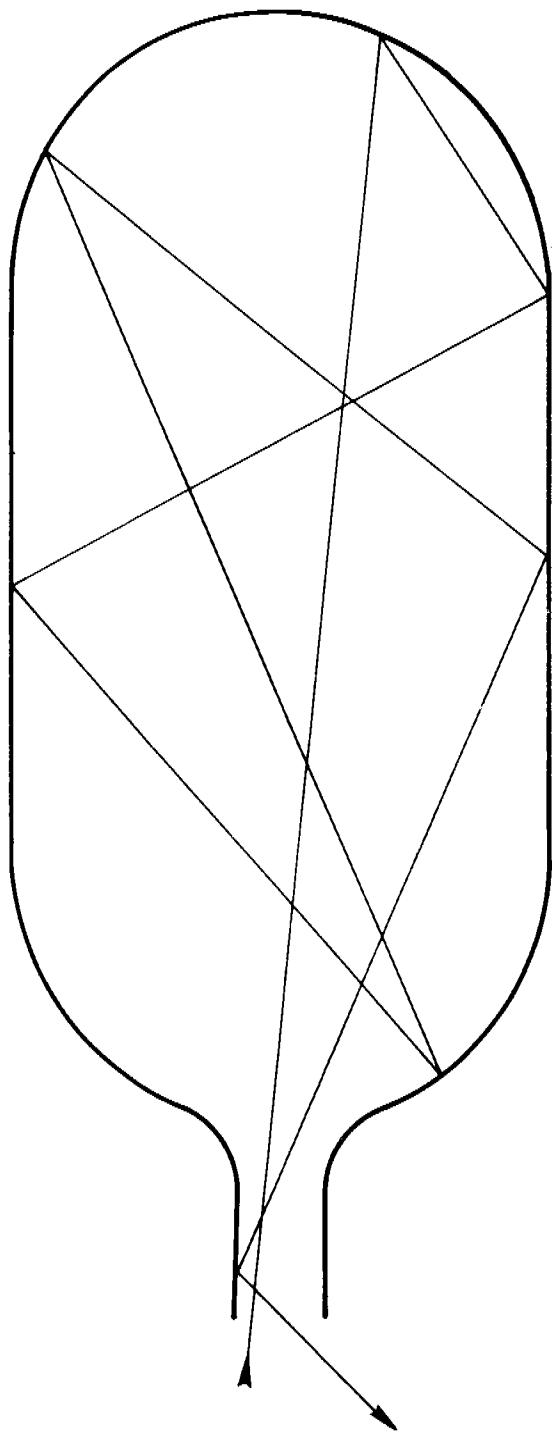
ACKNOWLEDGEMENTS

The author would like to acknowledge Dr. Peter Cervenka of Phoenix Corporation for providing Figure 9 and for his theoretical work demonstrating the feasibility of the external bulb variable volume maser.

REFERENCES

1. D. Kleppner, H. Goldenberg, and N. Ramsey [1962], Theory of the hydrogen maser, Phys. Rev., Vol. 26, 603
2. VANIER, J., LAROUCHE, R., and AUDOIN, C. [May 1975] The hydrogen maser wall shift problem. Proc. 29th Annual Symposium on Frequency Control (U.S. Army Electronic Command, Ft. Monmouth, N. J. 07703, May 28-30, 1975), in press (Electronic Industries Association, Washington, D. C. 20006).
3. BRENNER, D. [1969] Bull Am Phys. Soc., Vol. 14, 943.
4. BRENNER, D. [1970] Absolute frequency of the hydrogen maser using a flexible storage bulb. J. Appl Phys., Vol. 41, 2942.
5. REINHARDT, V. [1973] Flexible bulb-large storage box hydrogen maser. Proc. 5th Annual Precise Time and Time Interval Planning Meeting (NASA, Goddard Space Flight Center, Greenbelt, MD).
6. PETERS, H. E. [May 1975] The concertina hydrogen maser. Proc. 29th Annual Symposium on Frequency Control (U.S. Army Electronic Command, Ft. Monmouth, N. J. 07703, May 28-30, 1975), in press (Electronic Industries Association, Washington, D. C. 20006).
7. PETERS, H. E. [1971], Topics in Atomic Hydrogen Standard Research and Applications, Proceedings of the frequency standards and metrology seminar (Quebec).
8. CRAMPTON, S. B. and WANG, H. T. M. [May 1974] Density-dependent shifts of hydrogen maser standards. Proc. 28th Annual Symposium on Frequency Control (U.S. Army Electronics Command, Ft. Monmouth, N. J. 07703, May 29-31, 1974), pp. 355-361 (Electronic Industries Association, Washington, D. C. 20006).
9. REINHARDT, V. and PETERS, H. [May 1975] An improved method for measuring the magnetic inhomogeneity shift in hydrogen masers. Proc. 29th Annual Symposium on Frequency Control (U. S. Army Electronics Command, Ft. Monmouth, N. J. 07703, May 28-30, 1975), in press (Electronic Industries Association, Washington, D. C. 20006).
10. BERG, H. [March 1965], Spin exchange and surface relaxation in the atomic hydrogen maser. Phys. Rev. A., Vol. 137, A1621.

11. CRAMPTON, S. B. and WANG, T. M. [October 1975], Duration of hydrogen-atom spin-exchange collisions. Phys. Rev. A, Vol. 12, A1305.
12. REINHARDT [March 1974], The flexible bulb-large storage box hydrogen maser. Thesis (unpublished), Harvard University.
13. DEBELY, P. E. [1970] Hydrogen maser with deformable bulb. Rev. Sci. Inst., Vol. 41, 1290.
14. REINHARDT, V. [1973] Flexible bulb-large storage box hydrogen maser. Proc. 5th Annual Precise Time and Time Interval Planning Meeting (NASA, Goddard Space Flight Center, Greenbelt, MD).
15. UZGIRIS, E. E. and RAMSEY, N. F. [1970] Multiple region hydrogen maser with reduced wall shift. Physical Review A, Vol. 1, page 429.
16. WALLS, F. L. and HELWIG, H. [1976] A new kind of passively operating hydrogen standard, 30th Symposium on Frequency Control (USAEC, Ft. Monmouth, N. J.).
17. REINHARDT, V. S. and LAVANCEAU [1974] A comparison of the cesium and hydrogen hyperfine frequencies by means of portable clocks and Loran C, 28th Symposium on Frequency Control (USAEC, Ft. Monmouth, N. J.).
18. HELWIG, H. et. al. [1970] Measurement of the unperturbed hydrogen hyperfine transition frequency, IEEE Trans., Vol. IM-19, 200.
19. Dupont Trademark.
20. ZITZEWITZ, P. W. and RAMSEY, N. F. [1971] Studies of the wall shift in the hydrogen maser. Physical Review A. Vol. 111, page 51.
21. VESSOT, R. and LEVINE, M. [October 1970] A method for eliminating the wall shift in the hydrogen maser. Metrologia, Vol. 6, No. IV, pp 116-117.
22. VESSOT, R., et. al. [1971] Recent developments affecting the hydrogen maser as frequency standard. NBS Special Publication 343, Precision measurements and fundamental constants, August 1971. pp 22-37. (See also U. S. Patent No. 370-2972, Atomic hydrogen maser with bulb temperature control to remove wall shift).



$$\Delta f = \frac{\phi}{2 \pi t_0}$$

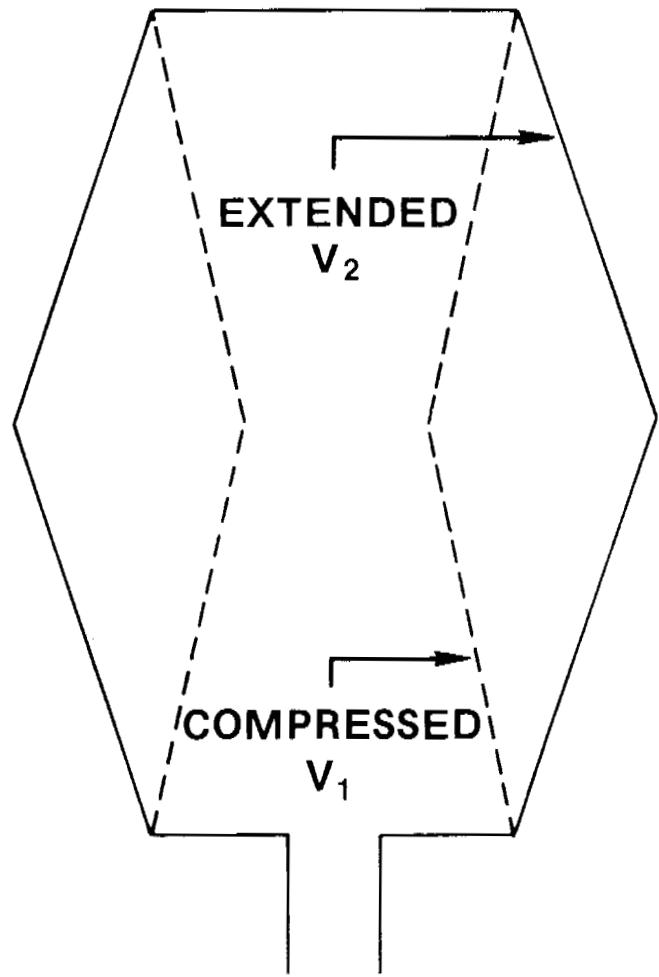
$$t_0 = \frac{\lambda}{\sigma}$$

$$\lambda = \frac{4 V}{A}$$

$$\Delta f = \frac{\phi \sigma A}{8 \pi V}$$

$$\frac{\Delta f}{f} \simeq 2 \times 10^{-11}$$

Figure 1. The Wall Shift



$$\Delta f = \frac{\phi \sigma A}{8 \pi V}$$

$$f_1 = f_0 + \Delta f_1$$

$$f_2 = f_0 + \Delta f_2$$

$$B = \frac{\Delta f_1}{\Delta f_2} = \frac{V_2}{V_1}$$

$$f_0 = \frac{B f_2 - f_1}{B - 1}$$

Figure 2. The Variable Volume Technique

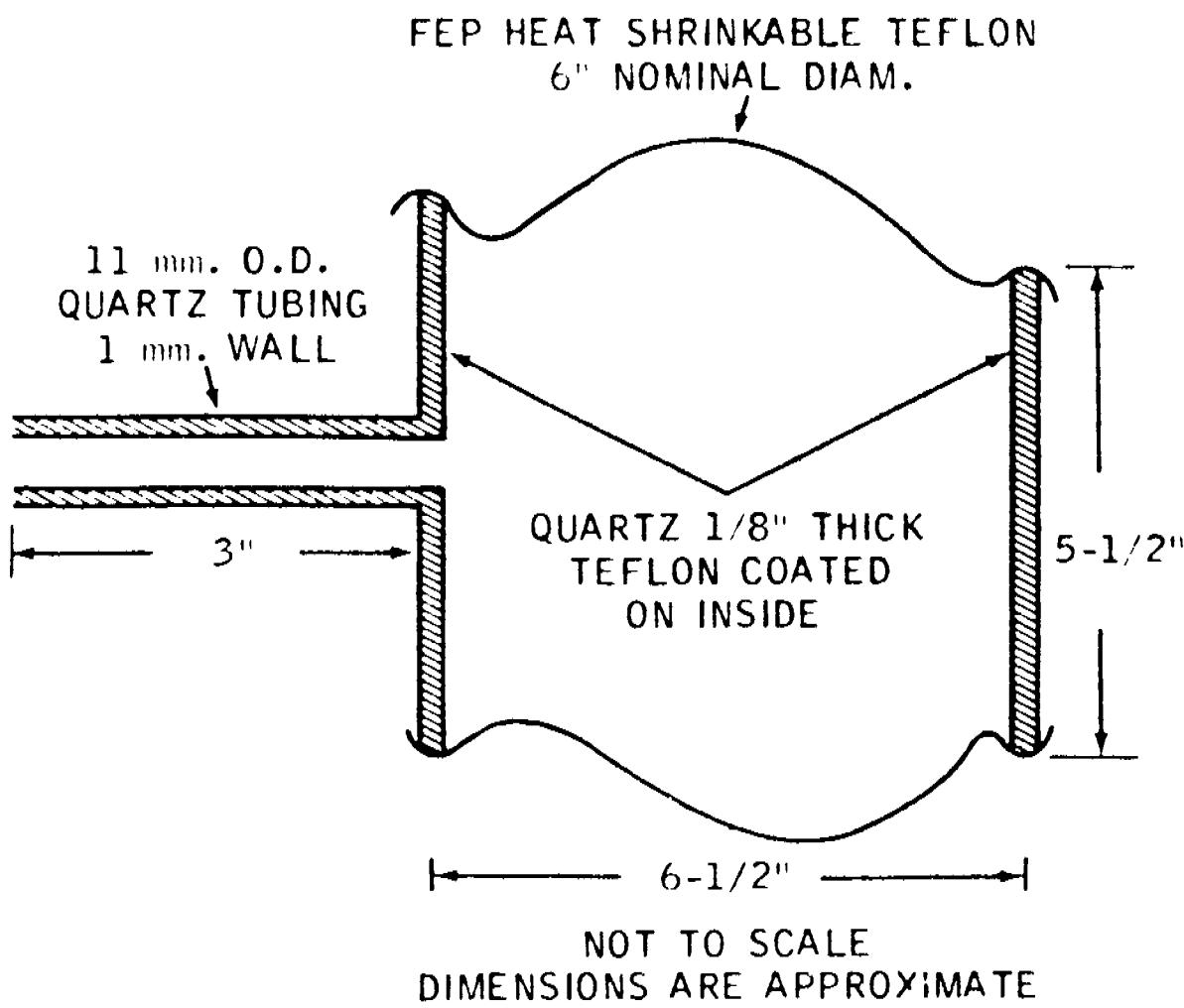


Figure 3. Brenner Variable Volume Storage Bulb (Reprinted From:
D. Brenner [1970], J. Appl Phys., Vol. 41, 2942).

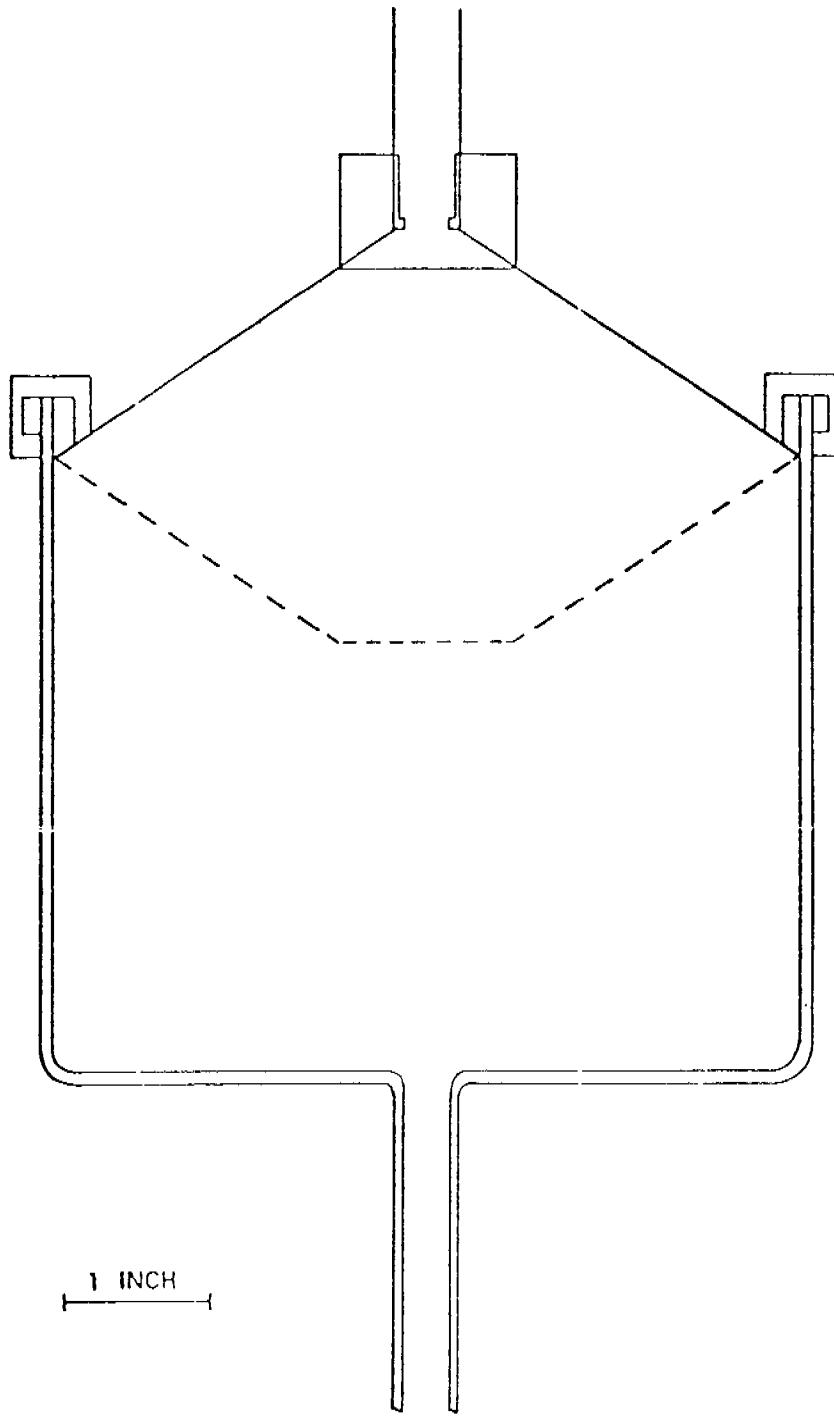


Figure 4. Debely Variable Volume Storage Bulb (Reprinted From:
P. E. Debely [1970], Rev. Sci. Inst., Vol. 41, 1290)

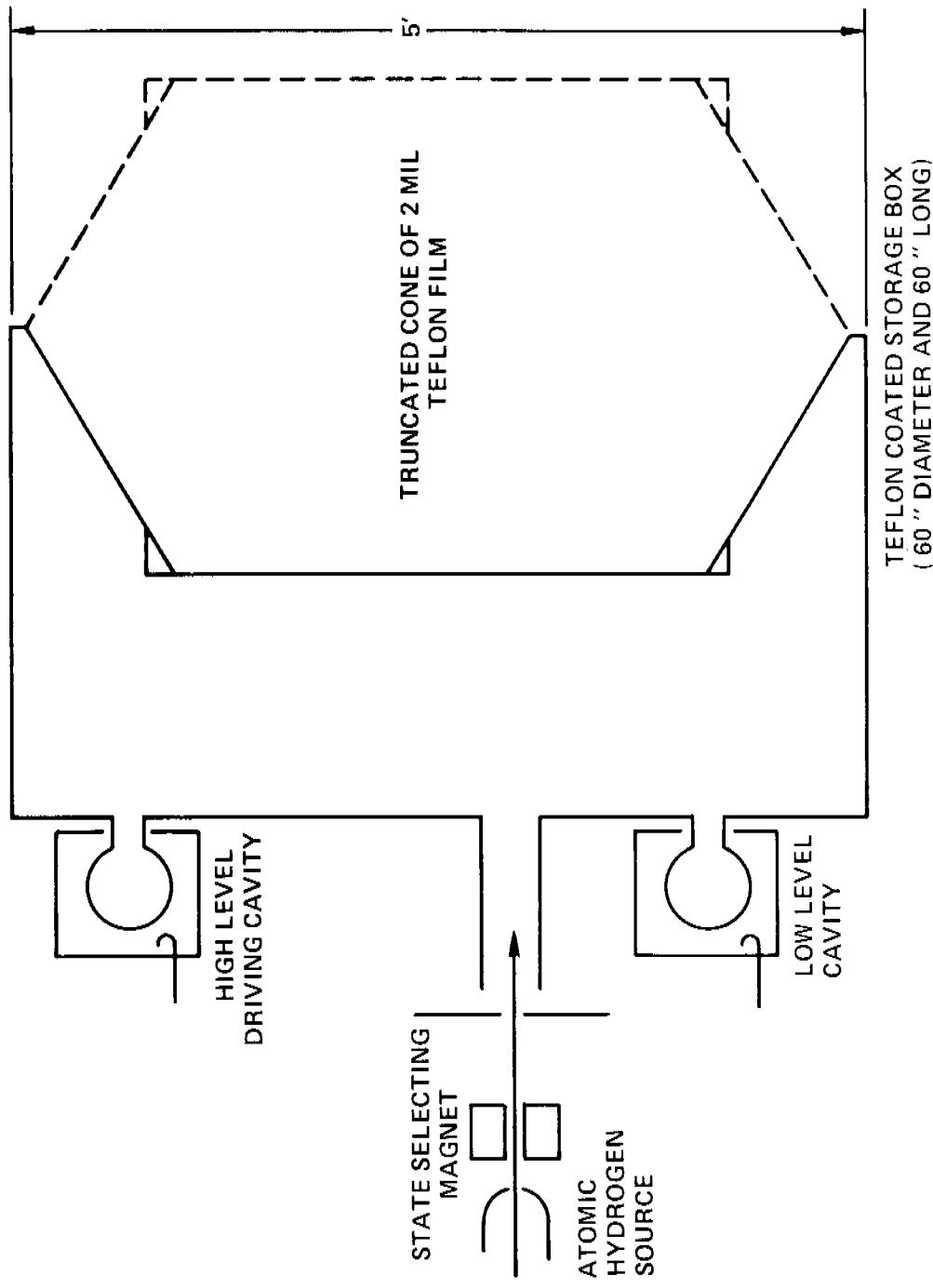


Figure 5. The Flexible Bulb-Large Storage Box Hydrogen Maser

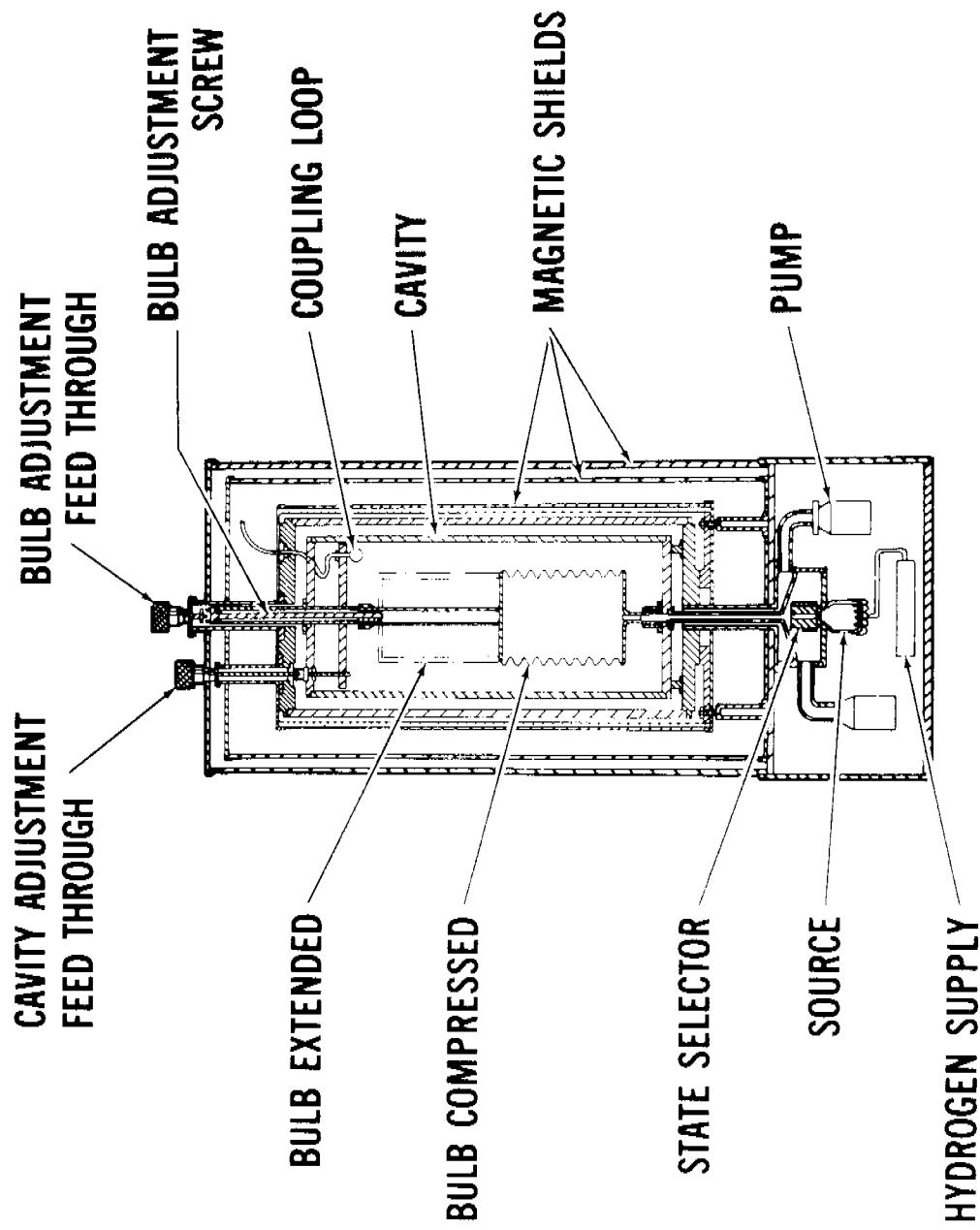


Figure 6. The Concertina Hydrogen Maser

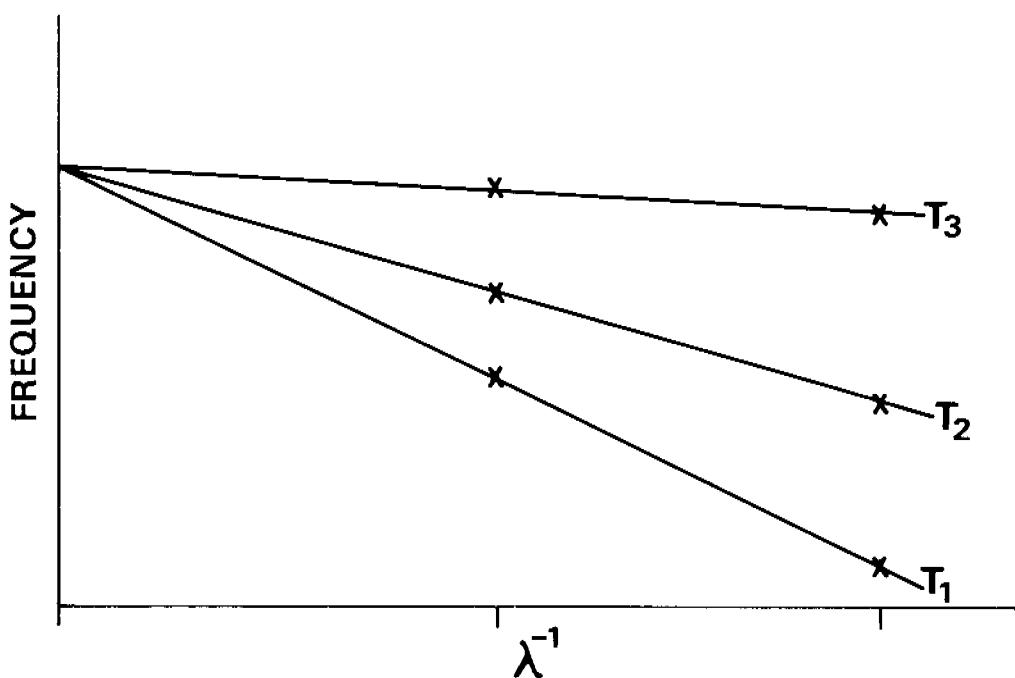


Figure 7. Extrapolation Method for Determining the Zero Wall Shift Point

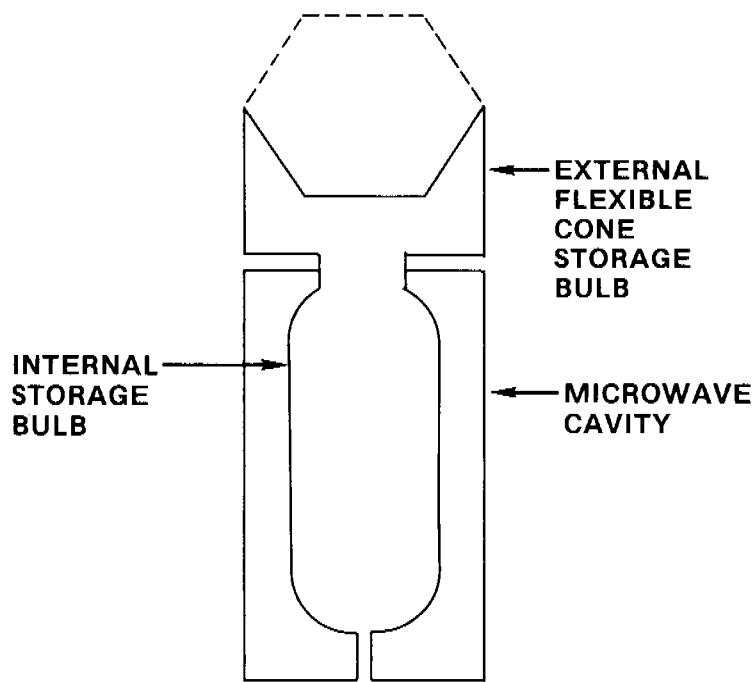


Figure 8. The External Bulb Variable Volume Maser

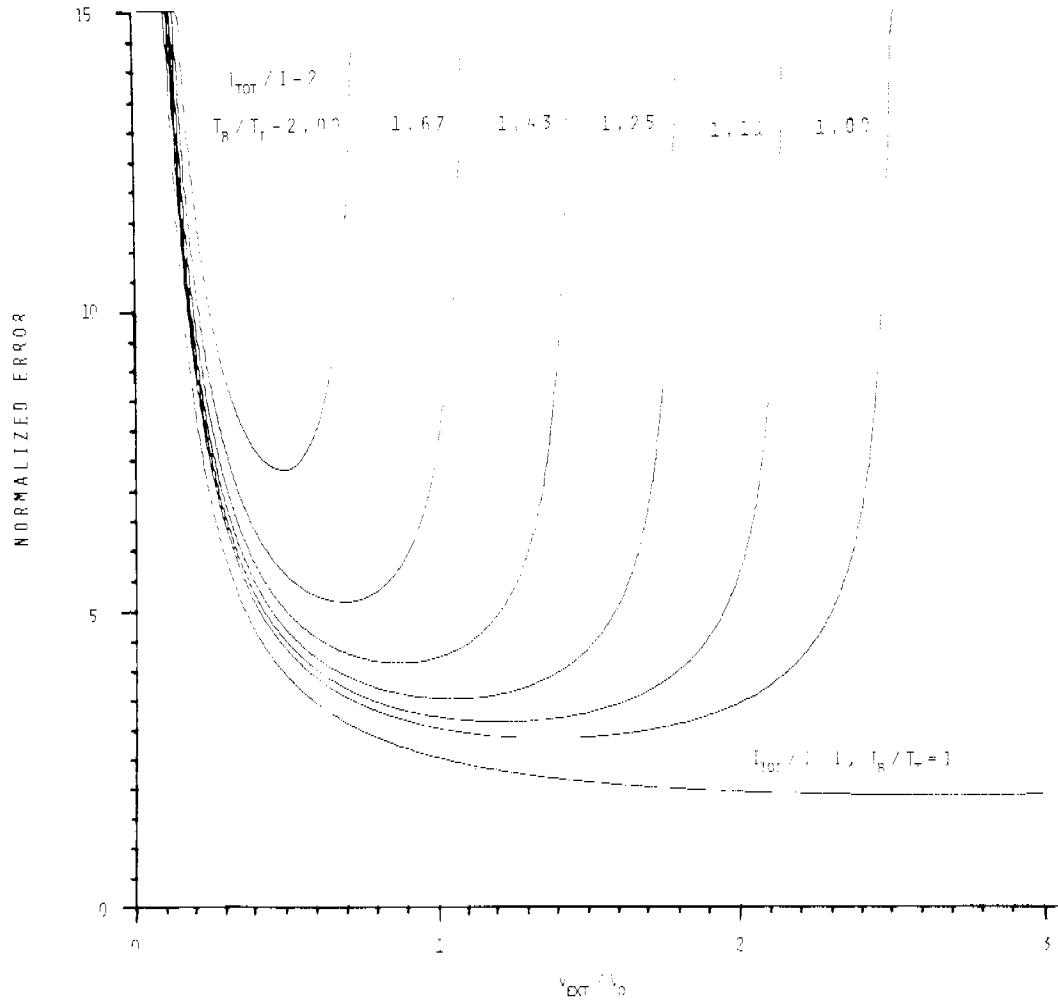


Figure 9. External Bulb Variable Volume Maser Error