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HYDROGEN MASER FREQUENCY STANDARD Final
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STUDY OF IMPROVEMENT OF HYDROGEN
MASER FREQUENCY STANDARD

Final Report: Contract NAS⁵-23520

Prepared for: National Aeronautics and Space
Administration, Greenbelt, Maryland

Submitted by: Professor Stuart B. Crampton
Principal Investigator

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TABLE OF CONTENTS

Introduction	1
Part One: Reduction of Atom Leakage Rate	1
Part Two: Investigation of New Surfaces	7
Part Three: Expenditures	11
Part Four: Final Inventory	12

INTRODUCTION

The long term frequency stability of hydrogen maser standards is ultimately set by the minimum usable linewidth. The minimum achievable drift of the hydrogen maser microwave cavity resonance frequency affects the oscillation frequency in direct proportion to the minimum atomic resonance linewidth at which acceptable oscillation power levels can be achieved. Changes of the magnetic field environment of the radiating atoms inside the best achievable magnetic shielding affect the oscillation frequency even after corrections for observed changes of average magnetic field in proportion to the atomic resonance linewidth. The minimum atomic resonance linewidth at which acceptable oscillation power (enough for acceptable short term stability) can be achieved depends in turn on the rate at which radiating atoms leak out of the storage bottle and on the rate at which the radiation is damped by collisions between the radiating atoms and the walls of the storage bottle. This work dealt primarily with reducing the atom leakage rate using as storage surfaces the FEP Teflon surfaces conventionally used in contemporary hydrogen maser frequency standards. Some work was also done on a possible alternative to the conventional surfaces, but the results here and elsewhere suggest that the alternative surface is not promising enough to warrant much further work.

PART ONE: Reduction of Atom Leakage Rate

This work was done in accordance with paragraph one of the statement of work accompanying NASA contract NAS5-23520, which reads as follows:

"1. The use of coated, capillary arrays to increase the bulb storage time will be investigated. Measurements will include the effects of the capillary arrays on the ratio of atoms entering the storage bulb to molecules pumped."

In a hydrogen maser standard the energy pump is provided by state selecting a hydrogen atom beam with a six pole magnet and introducing the state selected beam into a storage bottle through some sort of aperture. The radiating atoms leak out of the same aperture, and the

contribution of that leakage rate to the atomic resonance linewidth is directly proportional to the effective area presented to the atoms radiating inside the storage bottle. A simple hole does not make a successful aperture because the maximum attainable atomic beam intensities in atoms/cm²/sec do not provide a high enough atomic density inside the storage bottle for self-sustained oscillation on the atomic transition if the radiating atoms see an effective exit area equal to the entrance area. In the past, the effective exit area has been reduced relative to the effective entrance area by using a long stem as an aperture. Because the state selected atomic beam travels axially, it sees an entrance aperture equal to the area of the stem, but because the directions of the atoms radiating inside the storage bottle are random, they see an exit area equal to the area of the stem reduced by the kappa factor (roughly four times the diameter of the stem divided by three times the length of the stem for a long stem). In practice, there is a limit to the length of the stem set by (a) the geometrical space between the state selecting magnet and the microwave cavity in which the storage bottle must be centered, (b) the practicality of coating a storage bottle with a very long, thin stem, and (c) the introduction of annoying frequency shifts and instabilities as the volume of the stem becomes appreciable compared to the volume of the storage bottle itself.

This work focused on replacing the stem with a multitube collimator, coated with a material which would not relax radiating atoms while in the collimator and attached to the storage bottle after coating.

The collimators used are bundles of pyrex tubes having pore diameters from 0.050 mm on up, thicknesses of 0.5 mm on up, and maximum achievable transparency of 65% before coating or clogging the pores. These are available from Galileo Electro-Optics Corp., Sturbridge, Mass. at about \$50 for a capillary array 13 mm in diameter with many sizes of pore and length in stock.

The pores are a bit small for coating with FEP Teflon by conventional techniques, and so the collimators were coated using $(CF_3CH_2CH_2)_2SiCl_2$ = "BCFT" obtained from the Dow Corning Company in

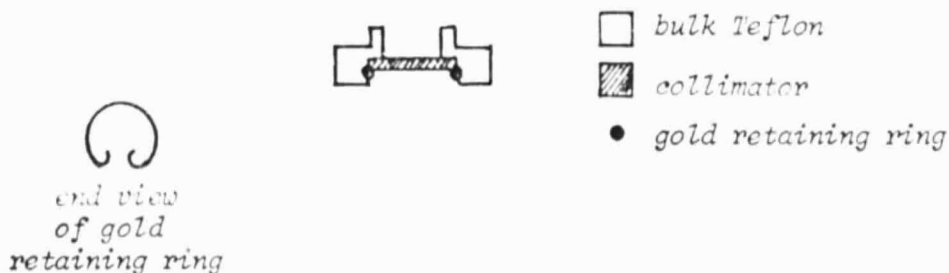
Midland, Michigan. Coatings made with this material had been studied here at Williams in 1971 by Kennedy Richardson, and his preliminary results are contained in his (unpublished) undergraduate honors thesis. He had found that the coating procedure described below produces an invisible film which water will not wet and which as a hydrogen maser storage surface introduces a wall shift typically twice as large and a wall relaxation rate typically twice as large as FEP Teflon.

In the first set of experiments a collimator having 0.050 mm pore diameter and 1 mm length was coated and attached to a 5 inch diameter spherical storage bottle coated with FEP Teflon. For comparison, measurements were made on a similar 5 inch diameter sphere having instead a 100 mm long by 8 mm diameter stem coated, as was the bottle, with FEP Teflon.

The procedure for coating the collimator was as follows:

- a. The collimator was soaked for several hours in undiluted BCFT and drained by standing it on some clean, absorbant material (kimwipe).
- b. The collimator was dipped for a few minutes in distilled water at a temperature between 40 degrees C and 50 degrees C. The purpose of the water dip is to react away any BCFT which has not already reacted with unreacted BCFT which has not already reacted with and been bound to the glass surface. The reaction of water with unreacted BCFT produces an oily residue which may keep water away from residual unreacted BCFT, so that it was thought advisable to agitate the collimator during the water dip.
- c. The oily residue was removed by an agitated dip in ethyl alcohol.

The Collimator was held in place as shown below:



The top of the collimator (as shown here) holder was a snug fit to a round hole in the bottom of the 5 inch storage bottle. The bottle was then placed in our hydrocarbon-free experimental hydrogen maser, and linewidths and power levels of oscillation at various atomic beam intensities were measured and compared to the results of similar measurements for the 5 inch storage bottle with stem. The results of the comparison were reported to the Eighth Annual Precision Time and Time Interval Planning Meeting held at the Naval Research Labs at the end of November 1976, and they appear in the published proceedings of that meeting. This preliminary work indicated both an improvement (reduction) of the range of linewidths over which the maser standard would oscillate at acceptable power levels and a concomitant reduction of necessary hydrogen atom beam intensity and pump capability and an indication that further reduction of atom storage time should be possible without unduly degrading the oscillation power. The limits to reduction of the leakage rate by longer and narrower collimating tubes are set by (a) the rate at which radiating atoms are relaxed by atom-surface collisions in the storage bottle itself but not removed by recombination = R_w , (b) the rate at which radiating atoms are lost by recombination during atom-surface collisions in the bottle itself = L_w , (c) the rate at which atoms are relaxed without recombining in the collimator = R_c , and (d) the rate at which atoms recombine in the collimator = L_c . If R_w is too large compared to the rate at which atoms leave the storage bottle or recombine, the hydrogen maser will not oscillate at any beam intensity. Our preliminary experiments indicated a total $R_w + R_c$ consistent with other measurements of R_w alone and small enough to allow a significantly reduced rate of recombining and/or leaving the bottle, if that could be achieved.

We next attempted to coat collimators having the same (0.05 mm) pore diameter but three times the length (3 mm). Unfortunately, BCFT clogged an appreciable proportion of the pores despite efforts to dislodge it using water, alcohol and compressed nitrogen gas. We went instead to a special order of collimators 8 mm in length with 0.20 mm pore diameters and guaranteed 65% transparency before coating. The

coating procedure used previously was successful, provided that the fluids were blown out of the pores using compressed nitrogen gas after each dip. One such collimator was installed polished side up (they had been ordered with one polished face in order to minimize the surface area presented to the inside of the storage bottle) in a holder similar to the one pictured on page 3 and used with a 7" diameter storage bottle freshly coated with FEP Teflon. The larger diameter was chosen to more nearly approximate the mean free path between atom-surface collisions in the large NASA hydrogen maser standards, and the results are easily scaled to what would have occurred for a 5" sphere or the larger volume of the NASA standard storage bottles.

The oscillation frequency of the 7" storage bottle with the long collimator was about 4 millihertz below the frequency of the NASA NX-1 comparison standard, whereas previous 5" diameter storage bottles had had oscillation frequencies about 11 millihertz below the NX-1 standard. These results are consistent within a measurement uncertainty of a few millihertz with the expected wall shifts of the different sized storage bottles without any contribution from wall shifts in the collimators themselves and indicate that even the long collimator with small pore diameters used with the 5" sphere did not significantly degrade the wall shift.

The relaxation parameters which could be measured simply by observing the relative oscillation power levels at different atomic beam intensities and degrees of cavity mistuning were measured by the technique outlined in the appendix to our previous publication Phys. Rev. A12, 1305 (1975). The results were

$$\frac{1}{\pi T_0} = 0.21 \pm .01 \text{ Hz}$$

$$\frac{8s_0}{\pi m T_B} = 2.84 \pm .09 \text{ Hz}$$

where $\frac{1}{\pi T_0}$ is the atomic resonance linewidth in the absence of any spin exchange collisions at all, s_0 is the ratio of the state-selected atomic beam intensity into the storage bottle and is normally close to $\frac{1}{2}$, $\frac{1}{\pi T_B}$ is the contribution to the resonance linewidth from recombination and

escape from the storage bottle, and m is the "maser quality factor:"

$$m = \frac{\bar{v}\sigma}{\pi} \frac{h}{\mu_0^2} \frac{V_c}{\eta Q V_B}$$

in standard hydrogen maser notation defined in the reference cited above. Another standard characterization of the oscillation conditions is

$$q = \frac{mT_B}{8T_t s_0}$$

which is just the ratio of the parameters above (provided that the T_1 relaxation at the wall is equal to the T_2 relaxation at the wall, which it is within the limit of precision of these measurements), or

$$q = .074$$

Another way of stating that the oscillation conditions were very good is to point out that the minimum linewidth for oscillation at all was 0.24 Hz while the maximum linewidth for oscillation at all was 1.17 Hz. Since .21 Hz of the linewidth was not due to spin exchange, the range of atomic beam intensities over which the maser oscillated was of the order of a factor of 30.

Assuming $s_0 = \frac{1}{2}$ and estimating m at 0.19 allows us to estimate

$$\frac{1}{\pi T_B} = .13 \text{ Hz,}$$

while if the only escape of atoms were through the collimator pores without recombination, that rate would be 0.09 Hz. In other words, the total amount of loss due to recombination on the storage bottle walls, recombination within the collimator walls or leakage around the collimator was tiny compared to the other contributions to linewidth.

Comparing dissociator pressures and storage bottle densities to those observed for the 5" bottle with stem, we believe that we do obtain the full 65% transparency to the beam, so that few pores are clogged and the alignment procedures are not stringent.

In terms of molecular hydrogen pumping requirements, the beam intensity at maximum oscillation power for the 7" sphere with the long collimator corresponded to a molecular hydrogen pumping rate of order 0.017 moles per year, while the pumping rate at maximum oscillation power for the 5" sphere was of order 0.09 moles per year.

A collimator with this geometry and quality of coating could reduce the minimum usable linewidth of NASA hydrogen maser standards to about 0.3 Hz times the ratio of the volume of a 7" sphere to the volume of one of the large NASA storage bottles, representing an improvement of linewidth and therefore also potential long term frequency stability by a factor of five or so.

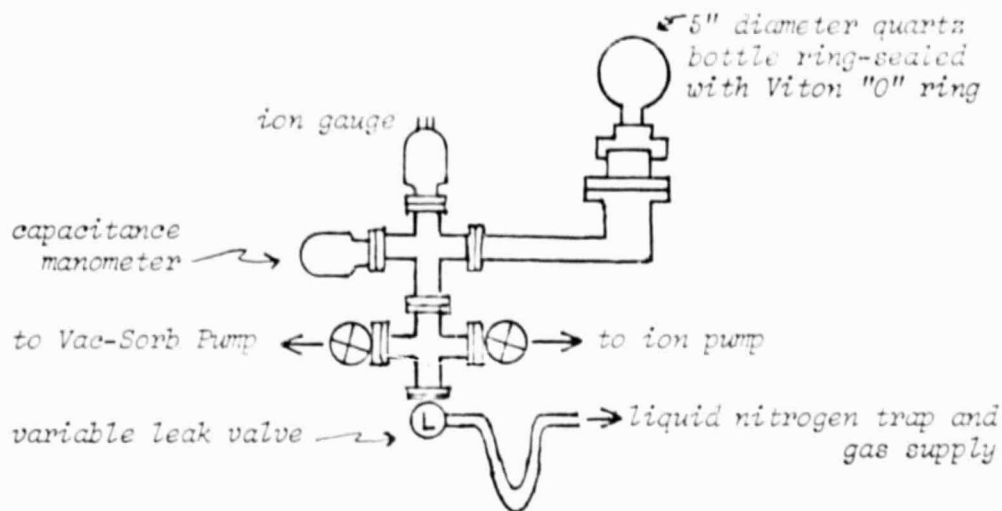
Other tricks with collimators could be tried, but the collimator used on our 7" sphere did not appreciably degrade the performance while providing very nearly the limit of storage time allowed by present storage bottle FEP Teflon wall coatings. We believe, therefore, that the improvement of storage times by improved collimation alone is essentially complete with this work.

PART TWO: Investigation of New Surfaces

This work was done in accordance with the second paragraph of the statement of work accompanying this contract:

"2. The improvement of wall relaxation times by the use of new storage bulb materials, and new storage bulb coating techniques will be investigated. Materials investigated will include Teflon polymerized by ultraviolet techniques and frozen fluorocarbons. Full documentation of techniques and materials used will be provided."

It was not possible to investigate frozen fluorocarbons, because we are not yet able to cool a storage bottle reliably to a low enough temperature. Polymerization of Teflon from C_2F_4 gas right in a storage bottle was investigated, using a clean gas handling system constructed for that purpose.



Initially, the ion pump was a large Vac-Ion pump which had pumped our H-10 hydrogen maser standard, and the variable leak was a Granville-Phillips servo valve controlled by the signal from the capacitance manometer. Using this system, one storage bottle was coated by the following procedure:

- a. The system was roughed out with the Vac-Sorb pump.
- b. The ion pump was started, the Vac-Sorb pump valved off, and the system pumped for several days with mild baking to a pressure of order 10^{-7} Torr, limited by the vac-ion pump itself.
- c. The liquid nitrogen trap was filled, and the ion pump was valved off.
- d. C_2F_4 gas was leaked into the system through the liquid nitrogen trap and held at a pressure of 2 Torr.
- e. The storage bottle was irradiated for a couple of hours by four u.v. lamps placed symmetrically around the bottle inside an aluminum reflector.

On the first run the pressure in the system actually rose above 2 Torr when the u.v. lamps were turned on, whereas we had expected the pressure to drop dramatically as the C_2F_4 polymerized. We surmised that the problem was outgassing by the system in the presence of the strong u.v. light. There was a little vacuum epoxy around the neck of

the storage bottle to seal a leak in the quartz ring seal (quartz to quartz, not the viton seal), but there was also much other contamination in the system, considering the high base pressure, and of course, there was the viton "O" ring itself. We pumped out the C_2F_4 and tried again.

On the next run we exposed the storage bottle to the u.v. light for several hours under high vacuum before valving off the ion pump and admitting the C_2F_4 . This time the pressure in the system did drop from 1.5 to .4 T when the u.v. light was turned on, indicating less out-gassing in competition with the polymerization. After a few minutes the pressure stabilized, but we continued the irradiation for a few hours, removed the storage bottle from the system, and installed it on our experimental hydrogen maser.

The results were disappointing. The hydrogen maser did oscillate with the storage bottle, but the wall shift was 28 millihertz greater than the wall shift for a similar bottle coated with FEP Teflon, and the linewidth at oscillation threshold (1.7 Hz) was about three times that attributable to the rate of escape through the aperture. We conclude that the coating of polymerized C_2F_4 was only partial and was contaminated by unknown impurities. Much of the relaxation not attributable to escape through the aperture must have been recombination, or the bottle would not have oscillated at all.

We have since improved the system somewhat. We have replaced the old ion pump by a new ion pump supplied by NASA, we have replaced the Granville-Phillips variable leak (which clogged with polymerized C_2F_4 by a 1½" bakeable valve, so that there are now three such bakeable valves on the system, and we have replaced the viton seal by a gold "O" ring sealing a copper flange fused by a graded seal to a new storage bottle. With mild baking (of the rather delicate system for sealing the storage bottle on the system) we have achieved a system pressure of 10^{-9} Torr. That represents a considerable improvement, but not enough of an improvement to warrant much confidence in the quality of surfaces produced by this technique. We have shown that coatings can be produced by this technique, and we have analyzed the property of one such coating carefully for what we believe to be the first time. The results suggest

That these techniques are not likely to produce coatings as good as the conventional FEP Teflon coatings or the more promising Type L Film bottles being investigated at NASA by Dr. Reinhardt. The results of our work have been simply to demonstrate low feasibility for the polymerized C_2F_4 coatings.

It would be interesting to see how well someone could do with these techniques by working hard on the cleanliness of the coating system and by experimenting with the polymerization conditions, including backfilling with various gases. We would like to keep the system intact and offer it next year as an undergraduate research project leading to a senior thesis at Williams, but we recommend bringing the intense phase of development with support by NASA of a postgraduate scientist to an end, considering the work contracted for under NASA contract NAS5-23520 to be complete insofar as it is likely to succeed.

EXPENDITURES:

a. Partial support of the summer salary of the principal investigator during the summer of 1976.	\$2,400.00
b. Partial support of the salary of a postdoctoral research associate during the fiscal year 1977 and partial support during the summer of 1977 for a recent graduate.	8,569.00
c. Computer costs (not charged to the contract).	0
d. Supplies, including repairs and replacement parts for equipment.	7,265.22
e. Permanent Equipment (see attached inventory).	5,408.71
f. Stipends for undergraduate summer research assistants.	2,940.00
g. Fringe benefits for staff in proportion to NASA support of their salaries.	1,953.85
h. Overhead (50.06% of salaries and stipends).	6,962.97
	<hr/>
	\$35,500.00

FINAL INVENTORY as of September 17, 1977

Equipment Supplied by NASA or Purchased by the Contractor and Reimbursed or to be Reimbursed by NASA.

A. Government Furnished Property

1. Experimental hydrogen maser apparatus parts

- a. Pump and Manifold assembly
- b. State selector magnet and source assembly
- c. Magnetic shield assembly
- d. Upper Vacuum enclosure
- e. Cavity assembly
- f. 3 quartz storage bottles
- g. Electronics module assembly

2. Controlled Parts

- a. Frequency Counter (NASA experimental) (NASA 198072)
- b. Tuning Recorder (NASA experimental) (NASA 198071)
- c. Crystal Oscillator (Austron 1270) (NASA 246482)
- d. Transmitter (NASA experimental) (NASA 198073)
- e. Power supply (HP 6284A, SN: H1384) (NASA 229616)
- f. Dist. Syst. Eng. Ins. (5008B, AN1164) (NASA 187599)

3. 1 Perkin-Elmer Model 203-2000 Ion Pump and Cable

B. Equipment Purchased by the Contractor

- a. 4 Phillips #2628990-140 HgCdZn ultraviolet lamps without power supplies, acquisition cost: \$274.93
- b. 1 Granville-Phillips Model #213 Pressure Controller, second-hand, acquisition cost: 907.74
- c. 1 Varian Model #971-0003 Ionization Gauge Controller, second-hand, acquisition cost: 302.24
- d. 1 Varian 1½" bakeable valve, second-hand, acquisition cost: 232.24
- e. 1 Varian Model #971-50080J nude ionization gauge, acquisition cost: 261.52
- f. Matheson #E15F-679 regulator, #68-1008 trap and #940F valve for a cylinder of fluorine, acquisition cost: 343.38

- g. 1 Mill Lane Engineering aluminum jacket
for cooling the cavity of the experimental
maser, custom made, acquisition cost: \$1,700.00
- h. 2 Varian Model #971-5005 glass ionization
gauges on conflat flanges, acquisition cost: 424.25
- i. L MKS #2214S-10 capacitance manometer with
power supply and cable, acquisition cost: 751.00

PROBLEMS IN HYDROGEN MASER DESIGN AND
SUGGESTED IMPROVEMENTS*

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ABSTRACT

Frequency shifts due to changing magnetic field inhomogeneities can limit the stability of atomic hydrogen maser standards. They can be minimized by careful design of the magnetic shields, by centering the storage bottle in the microwave cavity, and by operating at relatively high ambient magnetic fields, so as to make these frequency shifts less than one part in 10^{-13} and the instabilities due to changes of field much smaller than 10^{-13} . The inhomogeneity shift is also reduced by increasing the atom storage time, as are both the instabilities due to changes of cavity tuning and the amount of hydrogen atom flux required for self-sustained oscillation. Strategies for improved atom storage times include new surface materials and improved collimation techniques.

INTRODUCTION

Atomic hydrogen maser frequency standards are normally operated at very low average magnetic fields, in order to take advantage of the quadratic dependence of frequency on magnetic field so as to minimize errors due to changes of the average magnetic field.¹ Unfortunately, in low average magnetic fields the hydrogen maser oscillation frequency is subject to appreciable errors due to changes of gradients in the magnetic field. Motion through static and microwave magnetic field gradients by the radiating atoms induces hyperfine transitions which pull the oscillation frequency by amounts which can vary as the static magnetic field gradients change over time.^{2,3} This report provides rough estimates of frequency shifts to be expected because of this mechanism and suggests strategies for minimizing the effects on long term frequency stability.

MOTIONAL FREQUENCY SHIFTS

Changes $\delta\nu_{osc}$ of the spin exchange tuned hydrogen maser oscillation frequency¹ due to changes of static magnetic field gradient are of the order of³

$$\delta\nu_{osc} \approx \pm(3 \text{ to } 6) \times 10^{-3} (\pi T_B)^{-1} (\rho_{11} - \rho_{33})_0 [1 + (1.5 \times 10^{-5} \omega_z)^4]^{-1/2} \delta(\pi T_M)^{-1/2}. \quad (1)$$

$(\pi T_B)^{-1}$ is the contribution to the atomic resonance linewidth from the rate at which atoms escape from the storage bottle. It can be reduced by using a small effective exit area, but it cannot be reduced much below the contribution $(\pi T_W)^{-1}$ to resonance linewidth from wall collision relaxation plus the contribution $(\pi T_M)^{-1}$ from motion through static magnetic field gradients, without limiting the oscillation power level and spin exchange timing range. $(\rho_{11} - \rho_{33})_0$ is the net average electron polarization of the hydrogen atom beam as it enters the storage bottle. It can be reduced with some loss of oscillation power and tuning range by driving Zeeman transitions in the atomic beam before it enters the storage bottle, or it can be reduced without loss of power or tuning range using the double focusing technique to eliminate atoms in the uppermost hyperfine state from the atomic beam.^{4,5} ω_z is the angular frequency of the $\Delta F=0$ Zeeman transitions and is directly proportional to the static magnetic field averaged over the storage bottle. The contribution $(\pi T_M)^{-1}$ of magnetic field gradient relaxation to the atomic resonance linewidth is proportional to the mean square deviation of the static magnetic field from its average, so that $\delta(\pi T_M)^{-1/2}$ is proportional to the change of amplitude of the static magnetic field gradient. $(\pi T_M)^{-1}$ depends on ω_z roughly as $[1 + (1.5 \times 10^{-5} \omega_z)^4]^{-1/2}$, so that $\delta\nu_{osc}$ falls off with increasing static magnetic field roughly as ω^{-4} and can be made arbitrarily small by operating the hydrogen maser at relatively high magnetic fields such that ω_z is large compared to the rate at which atoms bounce back and forth across the storage bottle. In that case care must be taken to precisely measure and make relatively large corrections for the magnetic field dependence of the oscillation frequency in such high magnetic fields. The factor $\pm(3 \text{ to } 6) \times 10^{-3}$ in eq. (1) depends in sign and magnitude on the correlation between static magnetic field inhomogeneities and the configuration of the microwave magnetic field in the hydrogen maser cavity. Carefully centering the storage bottle in the cavity eliminates oscillation frequency pulling from first and all odd order static magnetic field gradients.

Eq. (1) predicts frequency shifts which are small enough to be hard to detect yet large enough to contribute to long term frequency instability. For example, for $(\pi T_M)^{-1} \approx 0.1$ Hz, a change of static magnetic field gradient of 10% may give a shift of order 5×10^{-14} of the oscillation frequency. The shift may be larger or smaller depending on the configuration of static magnetic field gradient,

but model calculations based on plausible field configurations suggest this order of magnitude at very low average magnetic field and $(\rho_{11} - \rho_{33})_0 \approx 1/2$. Of course, this source of oscillation frequency instability can be reduced by reducing changes of static magnetic field gradient by careful magnetic shield design, or by reducing any of the other factors in eq. (1).

INCREASED HYDROGEN ATOM STORAGE TIME

We have been doing experiments to maximize hydrogen atom storage time using multitube collimators to confine the atoms in the storage bottles longer, thereby reducing both the factor $(\pi T_B)^{-1}$ in eq. (1) and the overall linewidths at which the hydrogen maser is tuned and operated. Lower overall linewidth means less oscillation frequency instability due to drifts of the microwave cavity tuning. The beam intensity required for oscillation is also less, so that requirements of pumping speed and pump element life are eased. The only disadvantage is that the oscillation power level is reduced, so that longer averaging times are required.

Table one shows a comparison between relaxation rates and relative beam intensities measured for two 5" diameter spheres coated with FEP Teflon, one of which had a conventional stem to limit egress by the atoms and the other a multitube collimator in place of the stem.

TABLE ONE

	With Stem	With Collimator
$(\pi T_B)^{-1}$.67 Hz	.27 Hz
$(\pi T_O)^{-1}$.87 Hz	49 Hz
I	1	.27
V_C	5.16 cm ³	.1 cm ³

The stem was 7.54 mm I.D. by 115.6 mm long and was coated with FEP Teflon. The collimator was a bundle of tubes having .05 mm pore diameters by 1 mm long and said by the manufacturer⁶ to have 50% transparency before being coated with fluorinated drifilm. The $(\pi T_B)^{-1}$ contribution to the resonance linewidth due to atom escape from the storage bottle was substantially reduced for the bottle with the collimator. The overall density-independent linewidth $(\pi T_O)^{-1}$, including $(\pi T_B)^{-1}$, $(\pi T_M)^{-1}$ and wall collision relaxation, was also substantially reduced but by a smaller ratio because of a similar .2 Hz contribution by wall collision relaxation in both cases. Comparing the measured $(\pi T_B)^{-1}$ for the bottle with the collimator to what would be predicted by geometry suggested that the transparency of the collimator after coating was only 38%. The relative input beam intensity I for similar source conditions

was even less than .38, probably due to alignment problems. Despite the alignment loss, the ratio of maximum available resonance linewidth to $(\pi T_0)^{-1}$, which determines the spin exchange tuning range, was 1.2 times greater for the bottle with the collimator because of the much longer storage time. The volume V_c of the collimator, which is an important factor in the motional averaging frequency shifts discussed by Brenner,⁷ was only .02 as large as the volume of the stem.

These results are promising, and we plan to try some collimators having larger pore diameters for easier alignment and better transparency after coating but with some increase of volume. We also plan to try some new coating materials, in order to reduce the contribution to $(\pi T_0)^{-1}$ from wall collision relaxation, but we have not yet found anything better than FEP Teflon.

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