Offprint from "Metrologia"

Journal International de Métrologie scientifique — International Journal of Scientific Metrology
Internationale Zeitschrift für wissenschaftliche Metrologie. Vol. 6, No. 2, April 1970, Pages 56—60

© by Springer-Verlag 1970 · Printed in Germany

# The Hydrogen Storage Beam Tube, a Proposal for a New Frequency Standard\*

H. HELLWIG

Atomic Frequency and Time Standards Section, National Bureau of Standards, Boulder, Colorado 80302 USA

Received 16 January, 1970

## Abstract

The basic design features and the frequency stability and accuracy capabilities of a proposed new frequency standard

\* Contribution of the National Bureau of Standards, not subject to copyright.

are projected. The hydrogen storage beam tube combines the virtues of the hydrogen maser with those of beam tubes and eliminates some of the problems associated with these devices. The projected frequency stability for 1 s averaging is better than  $10^{-14}$ . The long-term stability should be equally good. Ways for investigating the wall shift are discussed.

#### 1. Introduction

The definition of the second is at present based on a certain hyperfine transition in the cesium atom. The utility of this definition obviously depends on the accuracy in the measurement of this transition. Research and development at several laboratories around the world are directed towards improving this accuracy. Recently published results indicate that several effects simultaneously impose accuracy limitations of the order of 10<sup>-13</sup> on the performance of the cesium beam tube [1, 2, 3]. These effects<sup>1</sup> are associated partly with the beam tube design and partly with the electronics, and it seems difficult to improve on all of them sufficiently in order to go well beyond the value of  $10^{-13}$ . But even if one assumes that these problems can all be solved by some, yet unknown, means, then a more basic effect enters the picture at accuracy values beyond 10<sup>-13</sup> to 10<sup>-14</sup>: the second order Doppler shift. This frequency shift which is proportional to the square of the particle velocity can not be avoided, and its knowledge depends on the knowledge of the value of the mean square velocity or in a more general sense of the temperature. In the case of beam tubes where the velocity depends not only on the oven temperature but also on the beam optics performance, it appears quite difficult to measure adequately (in the above sense) the mean square velocity of the particles.

Projecting into the future for accuracies beyond 10<sup>-14</sup>, the second order Doppler shift limitation therefore favors the gas cell approach of the kind used in the hydrogen maser or in the laser absorption technique [4]. Here, the kinetic energy of the atoms under interrogation is in thermal equilibrium with the storage vessel, and its temperature may be kept and/or measured with an accuracy adequate for frequency accuracies beyond 10<sup>-15</sup>. Following this line of thinking the application of the hydrogen storage technique

was reevaluated.

Shortcomings of the present hydrogen maser<sup>2</sup> are mainly the lack of accuracy due to inadequate methods of determining the wall shift and lack of longterm stability due to cavity pulling. In addition thermal noise in the cavity limits its frequency stability<sup>3</sup> [5]. It was noted that for beam tubes, the effect of thermal noise in the cavity is suppressed relative to its effect in a maser. The frequency stability of a beam tube is limited by shot noise of the beam and the performance of the slave oscillator [1]. In addition cavity pulling is much less pronounced.

As a result of these considerations, a new device is proposed which potentially combines the advantages of the hydrogen maser with those of a beam tube while offering in addition means to investigate the wall effects due to the storage of hydrogen atoms. This device may be called a hydrogen storage beam tube. Its concept is a logical extension of Ramsey's original storage beam proposal and his "bounce box" experiments with cesium [6] which led Ramsey to the

given in Ref. [5].

development of the hydrogen maser. As will be discussed in the following a revival of the idea of a bounce box in a passive device appears to be a promising approach for a frequency standard of high precision.

# 2. Design of the Proposed Hydrogen Storage Beam Tube

A beam of hydrogen atoms will be used which travels from a dissociator-source through a magnetic state selector into a storage bulb which is located in a cavity tuned to the hydrogen hyperfine transition frequency. This portion of the device functions analogously to the concept of a regular hydrogen maser. However the storage bulb will have a second (exit) opening. The beam of hydrogen atoms leaving the bulb through the exit then passes into a second state selector magnet and is finally focused on a beam detector suitable for hydrogen atoms (e.g., electron bombardment type). This configuration corresponds to a "flopout" mode of operation in regular beam tubes. Fig. 1 depicts this basic design.



Fig. 1. Basic configuration of a hydrogen storage beam tube

Since the atoms are to be stored for times of the order of 1 s, the storage bulb has to be designed accordingly. In particular, some measures have to be taken to prevent atoms from traversing directly from the entrance of the bulb to its exit. This could be done by an axial offset of the two openings, or by a Teflon4 beam stop within the bulb, or by any other suitable means. However, if the conventional TE<sub>011</sub> cavity is used it is desirable to keep entrance and exit of the bulb close to the cavity axis. The exit opening should be designed for an adequate degree of collimation, and its flow resistance as compared to the beam entrance should be chosen such as to allow the majority of stored atoms to leave the bulb through the exit. Both requirements will be easily satisfied because the extremely low atomic hydrogen gas pressure allows the very effective use of multi-channel apertures (made from Teflon).

It also appears possible to use only one opening as a combined entrance-exit together with only one state selector and an annular detector configuration. Furthermore the bulb need not be confined to be within the cavity but may extend outside of the cavity in a fashion somewhat similar to Ramsey's large storage box maser [7].

The interrogation of the stored, excited hydrogen atoms is accomplished by injecting a microwave signal at the atomic transition frequency into the cavity. Technical details will correspond to the well documented methods used in cesium beam tubes [1, 2, 3, 8].

As compared to the hydrogen maser one new source of frequency error is introduced because of the operation as a passive device: the uncertainty due to impurities in the exciting microwave signal. However,

<sup>&</sup>lt;sup>1</sup> D. J. Glaze gives a summary of these effects for NBS III, which is in operation at the National Bureau of Standards, USA [1].

<sup>2</sup> A comprehensive description of the hydrogen maser is

<sup>&</sup>lt;sup>3</sup> In a practical hydrogen maser at present this effect is covered by other noise processes. However, here it is assumed that those effects can be eliminated.

<sup>&</sup>lt;sup>4</sup> DuPont's polytetrafluoroethylene.

based on results obtained with cesium [1] it can be predicted that this uncertainty will be close to  $10^{-14}$ .

#### 3. Short-Term Frequency Stability

One can distinguish between different methods of obtaining the response of an atomic ensemble which is being interrogated by radiation of the atomic transition frequency: (1) Detection (counting) of atoms which have completed the desired transition. This is the standard detection method in beam tubes. (2) Detection of an increase or decrease in the power of the interrogating radiation (passive devices) or of the generation of radiation by self-oscillations (active device). This method is used in masers and lasers. (3) A third detection method involves optical pumping as for example in the rubidium gas cell device; however, we shall not consider its aspects in detail in this paper.

A frequency standard using the first detection method is ultimately limited in its stability by the shot noise of the incoming atoms. In the case of the second method the stability limiting effect is the influence of thermal noise (the black body radiation in the resonance cavity) on the stimulated emission, absorption, or oscillation of the ensemble of atoms. A quantitative comparison is given in the following. The frequency stability of a maser oscillator was given by Shimoda, Wang, and Townes [9], and Cutler and Searle [10], to be approximately

$$\sigma_{\rm M} \approx \frac{1}{\sqrt{2}Q_1} \sqrt{\frac{kT}{P}} \frac{1}{\sqrt{\tau}}$$
 (1)

where  $\sigma=$  fractional standard deviation,  $Q_1=$  Q-value of the transition line, k= Boltzmann constant, T= temperature, P= power of the maser, and  $\tau=$  averaging time. A typical hydrogen maser would exhibit  $\sigma\approx 10^{-14}$  for  $\tau=1$  s. Actually this stability figure can not be realized because of other noise processes which are associated with the detection of the maser signal. The frequency stability of a beam tube system was given by Cutler and Searle [10], and by Lacey, Helgesson, and Holloway [11] as approximately

$$\sigma_{\rm T} \approx \frac{0.388}{Q_1 \sqrt{\tau}} \frac{I_{\rm n}}{I_{\rm s}} \tag{2}$$

where  $I_n$  is the noise current in a 1 Hz bandwidth and  $I_s$  is the signal current. The ratio  $I_n/I_s$  may be replaced by  $(n/2)^{-1/2}$  where n is the flux of signal atoms. n can be expressed by the total microwave power P available from the beam and one can rewrite Eq. (2)  $(\nu = \text{transition frequency})$  as approximately

$$\sigma_{\rm T} \approx \frac{1}{2Q_1} \sqrt{\frac{h\nu}{P}} \frac{1}{\sqrt{\tau}} \ .$$
 (3)

Shimoda, Wang, and Townes [9] pointed out that a maser will not "suffer" from shot noise because the effect of thermal noise on the stability is larger by the factor

$$\frac{\sigma_{\rm M}}{\sigma_{\rm T}} \approx \sqrt{2} \sqrt{\frac{kT}{hv}}$$
 (4)

as can be seen by comparing Eqs. (1) and (3). For the hydrogen maser versus hydrogen storage beam tube one calculates a value of  $\sigma_{\rm M}/\sigma_{\rm T}\approx 90$  (at room temperature). Interpreting the result differently we conclude that a beam tube operating with the same beam intensity and the same radiation interaction

time as a corresponding maser oscillator will display a stability which is better by the same factor. For the proposed hydrogen storage beam tube this results in a one second frequency stability of approximately  $1 \times 10^{-16}$  as compared to  $10^{-14}$  for the regular hydrogen maser<sup>5</sup>. This, of course, is true only if the hydrogen storage beam tube operates under conditions comparable to the present hydrogen maser, and provided the detection of atoms leaving the bulb is 100% efficient. The first condition is fully realizable; the second one is not. Based on a detector efficiency of several percent [12], it appears reasonable to assume at least 0.01% overall detection efficiency, in which case the hydrogen storage beam tube would exhibit  $\sigma(1 \text{ s}) \approx 1 \times 10^{-14}$ . We assume that slave oscillators and multiplier chains will be available with a performance which is compatible with this stability figure. The beam tube has a further advantage over the maser in that additive noise in practical beam detection electronics has only a negligible influence on the short-term stability performance at common beam intensities of  $> 10^8$  particles per second. One could even tolerate fairly large noise figures of the beam detector preamplifier although values of less than 1 dB can be realized. In contrast, the short-term stability performance of the practical maser oscillator will always be covered up by additive noise of the microwave receiver thus being worse than the calculated  $\sigma$  (1 s)  $\approx 10^{-14}$ .

## 4. Frequency Accuracy and Long-Term Stability

A variety of effects, such as cavity phase shifts, magnetic field problems, and Doppler shifts which limit the performance of cesium beams, do not pose a limitation for a hydrogen storage device (both masers and beam tubes) down to parts in  $10^{15}$ . Requirements on the servo for the slave oscillator are also relaxed due to the higher line Q of the proposed device as compared to cesium.

Two problems which do affect seriously the accuracy of the hydrogen maser and which have some bearings on the hydrogen storage beam tube remain to be discussed, the cavity pulling and the wall shift. If methods can be found to cope adequately with these two effects the hydrogen storage beam tube may be expected to be capable of accuracy figures of better than 10<sup>-14</sup>.

#### 4.1 Cavity Pulling

The hydrogen storage beam tube displays an excellent performance with respect to cavity pulling. Cavity pulling is a result of a frequency dependent change of the power level in the cavity which follows the cavity resonance. A formula describing this effect for beam tubes was given by Holloway and Lacey [13]

$$\frac{\Delta v}{v} = F \left(\frac{Q_c}{Q_1}\right)^2 \frac{\Delta v_c}{v} \tag{5}$$

where  $Q_c = Q$  value of the cavity,  $\Delta \nu$  and  $\Delta \nu_c =$  offset of the apparent atomic resonance and cavity, respectively, F = factor which is a function of the excitation power. The magnitude of F is of the order of unity.

Eq. (5) holds strictly only if the atoms under interrogation do not contribute power to the cavity.

<sup>&</sup>lt;sup>5</sup> This figure may be compared with the present 1 stability of laboratory-type cesium beams of  $10^{-12}$  (NBS X4) and a projected figure of  $2 \times 10^{-13}$  [1] (NBS 5).

For an experimental hydrogen storage beam device we can expect typically a beam contributed power of  $10^{-2}$  to  $10^{-4}$  of the driving microwave power. The corresponding change in the susceptibility of the cavity can lead to a small detuning of the cavity which would modify  $\Delta \nu_c$  in Eq. (5). However, this modification is negligible for all practical aspects.

The cavity of the hydrogen storage beam tube may have a very low Q value in order to reduce cavity pulling according to Eq. (5). In the case of the hydrogen storage beam tube the required power level of the exciting microwave signal is fairly low so that spoiling of the cavity Q will not push the power beyond technically feasible levels. First order Doppler and phase shifts which could be more pronounced with a very low cavity Q are negligible in this device due to the storage process within a single cavity.

A value  $Q_{\rm c}\approx 10^3$  shall now be assumed. With  $Q_1\approx 10^9$  this leads to a "pulling factor" of only  $(Q_{\rm c}/Q_{\rm l})^2=10^{-12}$ . If frequency pulling shall be kept below  $\Delta v/v\approx 10^{-15}$  this means that the cavity has to be kept within  $\Delta v_{\rm c}/v\approx 10^{-3}$  only. Cavity detuning is mainly caused by temperature change and storage bulb adjustments. The temperature dependence of the cavity resonance may be written as

$$\frac{\Delta v_{\rm c}}{v} = \alpha \Delta T \tag{6}$$

with  $\alpha \approx 3 \times 10^{-6} \, \mathrm{K^{-1}}$  as a typical value for a quartz cavity. Using this value and the above assumptions, one concludes that, for  $\Delta \nu / \nu \approx 10^{-15}$ , temperature changes, without retuning, of  $\Delta T \approx 300 \,\mathrm{K}$  can be tolerated! We conclude that cavity pulling is virtually of no concern for the hydrogen storage beam tube. For this reason spin exchange frequency shifts [5] which do occur at higher beam intensities or atomic densities in the storage bulb can be measured directly by varying the intensity of the beam. We can expect that usually the spin exchange frequency shift is a relatively small effect. Its magnitude is a linear function of the beam intensity, and one calculates for a flux of 1012 atoms/s into a storage bulb of  $10^3$  cm<sup>3</sup> ( $t_E = 1$ s) a fractional frequency shift of  $3 \times 10^{-13}$  [14]. The longterm stability should exceed the performance of the hydrogen maser which is apparently limited by cavity detuning effects even if the cavity is servoed [15, 16].

## 4.2 Wall Shift

The hydrogen atoms stored in the bulb will collide with its walls many times, about  $10^4$  collisions within 1 s. Although the disturbance of the atomic energy levels due to these collisions is largely reduced by suitable coating of the bulb walls (Teflon), a finite influence remains which is conveniently described by a certain average phase shift  $\phi_0$  per collision. The resultant shift in frequency is proportional to the product of  $\phi_0$  with the frequency of the wall collisions. For a spherical bulb of radius R and atoms of the mean velocity  $\bar{v}$  we have [17]

$$\frac{\Delta v}{v} = \text{const } \phi_0 \frac{v}{R} \ . \tag{7}$$

The total shift for a Teflon coated bulb of about 15 cm diameter is of the order of  $10^{-11}$  [18]. From Eq. (7) it appears possible to study the wall shift by varying v. However, it has to be noted that a temperature variation not only changes the mean velocity but also

the collisional interaction, i.e.,  $\phi_0$  is strongly temperature dependent. Taking temperature as the variable we may write

$$\frac{\Delta v}{v} = \text{const } \mathbf{f} (T) \sqrt{T} . \tag{8}$$

The function f(T) is not yet understood very well. Some experimental data are available [18] and experiments with the hydrogen maser are in progress at Harvard University [19] and the Smithsonian Astrophysical Observatory [20] which indicate a very interesting and surprising behavior of f(T), most notably a change of the effect from a negative to a positive frequency shift at a temperature of about 373 K.

Experiments with the hydrogen storage beam tube will lead to quantitative results with regard to Eq. (8) and therefore hopefully to a better understanding of the wall shift. The hydrogen storage beam tube will be better suited for such experiments than the maser oscillator because cavity tuning is not a problem, and the lack of a threshold condition permits investigations at parameter settings which are not realizable in the maser. These are also important assets of the beam tube with respect to other wall shift experiments based on the variation of the radius, i.e., a change of the surface to volume ratio of the storage bulb. Finally, it must be emphasized that the second order Doppler shift, a frequency shift linear with the temperature, has to be accounted for in all experiments relating to Eq. (8). However this can adequately be done by calculation based on measurements of the storage bulb temperature if we assume that the kinetic energy of the stored atoms is in thermal equilibrium with the storage bulb<sup>6</sup>.

## 5. Conclusions

The hydrogen storage beam tube appears to offer capabilities surpassing considerably those of any existing frequency standard in terms of frequency stability. The frequency accuracy expectations are also promising. All known limitations on the accuracy are expected to be below a fractional uncertainty of 10<sup>-14</sup> with the wall shift being the only exception. However, the proposed hydrogen storage beam tube facilitates wall shift determinations, and offers new possibilities for wall shift studies. It is hoped that this will lead to a better understanding and more accurate measurements of the wall shift than previously possible. If this expectation can be experimentally realized, the hydrogen storage beam tube will uniquely combine unexcelled short-term frequency stability with a frequency accuracy capability equal to or better than cesium beam tubes currently in the development

Finally it shall be emphasized that the proposed hydrogen storage beam tube in its design and performance expectations, as outlined in this paper, is based on a straightforward extension of principles already proven in the hydrogen maser and in beam tubes. Its experimental realization appears entirely feasible although a considerable amount of effort will be necessary.

<sup>&</sup>lt;sup>6</sup> At present there is no strong experimental evidence for or against this assumption. The proposed hydrogen storage beam tube is expected to greatly facilitate experiments directed at this question, and it offers a technical solution by pre-thermalization in a separate storage vessel.

Acknowledgements. The author wishes to express his sincere appreciation to Dr. Donald Halford for many helpful discussions, in particular for suggesting the comparison of the stability performance of masers and beam tubes.

#### References

1. Glaze, D. J.: Presented at the XVIth General Assembly of the International Scientific Radio Union (URSI) in Ottawa, Canada, by J. A. Barnes (August, 1969); submitted to IEEE Trans. on Instr. and Measurements.

2. Becker, G., Fischer, B., Kramer, G., Müller, E. K.: PTB-Mitteilungen 2, 77 (1969). 3. Mungall, A. G., Bailey, R., Daams, H., Morris, D.: Metro-

logia 4, 165 (1968).

- 4. Barger, R. L., Hall J. L.: Phys. Rev. Letters 22, 4 (1969). 5. Kleppner, D., et al., Vessot, R. F. C., et al.: Phys. Rev.
- 138 Å, 972 (1965). 6. Kleppner, D., Ramsey, N. F., Fjelstad, P.: Phys. Rev.
- Letters 1, 232 (1958).
- 7. Uzgiris, E. E., Ramsey, N. F.: IEEE Journal of Quantum
- Elec. QE-4, 563 (1968). 8. Beehler, R. E., Mockler, R. C., Richardson, J. M.: Metrologia 1, 114 (1965).

- 9. Shimoda, K., Wang, T. C., Townes, C. H.: Phys. Rev. 102,
- 10. Cutler, L. S., Searle, C. L.: Proc. IEEE 54, 136 (1966).
- 11. Lacey, R. F., Helgesson, A. L., Holloway, J. H.: Proc. IEEÉ 54, 170 (1966).
- 12. Andresen, S.: Private communication.
- 13. Holloway, J. H., Lacey, R. F.: In Proc. of the Congrès International de Chronométrie, Lausanne, Juin 1964, V. 1, p. 317.
- 14. Hellwig, H.: NBS Technical Note, No. 387 (1970).
- 15. Vessot, R. F. C., Levine, M., Mueller, L., Baker, M.: In Proc. of the 21st Annual Symposium on Frequency Con-
- trol, Ft. Monmouth, N. J., 1967, p. 512. 16. Hellwig, H., Pannaci, E.: Proc. IEEE 55, 551 (1967).
- 17. Berg, H. C.: Phys. Rev. 137 A, 1621 (1965).
- Uzgiris, E. E., Zitzewitz, P. W.: Proc. 23rd Annual Symp. on Freq. Control, Ft. Monmouth, N. J., 284 (May, 1969).
- 19. Zitzewitz, P.: Private communication; to be presented at the 24th Annual Symp. on Freq. Control, Ft. Monmouth, N. J., and published in the Proceedings (1970).
- 20. Vessot, R. F. C.: Private communication; to be presented at the 24th Annual Symp. on Freq. Control. Ft. Monmouth, N. J., and published in the Proceedings (1970).