Properties of the Hydrogen Maser

Daniel Kleppner, H. Mark Goldenberg, and Norman F. Ramsey

Properties of the hydrogen maser and details of the apparatus are discussed. Experimental work is described which yields a new value for the hyperfine separation of hydrogen in its ground state. The result is

$$\Delta\nu = 1{,}420{,}405{,}762 \pm 4 \text{ cps.}$$

This result is based on a value of the hyperfine separation in Cs^{133} which is taken to be

$$\Delta\nu(Cs^{133}) = 9{,}192{,}631{,}840 \text{ cps.}$$

I. Introduction

Amplification through the use of stimulated emission was first accomplished with the 3-3 line in ammonia, using a molecular beam technique to give the required state inversion.[1] Since then a great many devices utilizing numerous resonance systems and a variety of inversion techniques have been successfully operated. Until recently, however, perhaps the simplest possible resonance system—the ground state of a free atom—was not utilized. This is because the magnetic dipole transition, the only transition possible in the ground state of a free atom, has radiation matrix elements which are characterized by the Bohr magneton, μ_0, approximately one hundred times smaller than μ_e, the electric dipole moment characteristic of molecular transitions. To compensate for the small size of the moment either the beam flux must be increased by approximately $(\mu_e/\mu_0)^2$, a prohibitive factor of 10^4, or the time in which the atom interacts with the field must be increased by (μ_e/μ_0). The latter possibility has been achieved recently by the use of the "storage box" technique in which the atoms are constrained to move within the volume of the storage box by collisions with the walls.[2,3] In this sense the atoms are not strictly "free," for they experience large forces during the time of a collision. However, if the time of the collision is very short, and if the forces do not seriously upset the spacing of the energy levels of interest, the atom is indeed very close to being entirely free. This has turned out to be the case for atomic hydrogen with suitably inert wall surfaces, and it has enabled the operation of a maser oscillator utilizing the $(F = 1, m_F = 0) \rightarrow (F = 0, m_F = 0)$ hyperfine transition in hydrogen. This article summarizes some of the features of the hydrogen maser and describes experimental work which has led to a new value for the hyperfine separation in hydrogen. A more complete discussion of the maser's theoretical properties is being prepared for publication elsewhere.[4] One of the authors has presented an unpublished account of the experimental work which is described here.[5]

II. Analysis of the Maser Oscillator

The operation of a beam type maser has been analyzed by Shimoda *et al.*[6] and with a few modifications their results can be applied to the hydrogen maser. An important difference between a conventional beam maser and the hydrogen maser is that in the former the time spent in the rf field is characterized by the fixed path length each molecule travels across the cavity and a velocity which may either be considered fixed (as in ref. 6) or is given by a modified Maxwellian distribution, while in the latter the time is described by an exponential distribution function with a decay time equal to the mean time necessary for an atom to effuse from the box. Another important difference is due to the effect of wall collisions on the resonance states in the hydrogen maser, but this will be neglected initially.

The familiar energy level diagram for the ground state of hydrogen is shown in Fig. 1. The transition of interest, $(F = 1, m_F = 0) \rightarrow (F = 0, m_F = 0)$, occurs at a frequency $\Delta\nu = 1420$ Mc/s approximately. A schematic diagram of the apparatus is shown in Fig. 2. Atoms from the source pass through the state selector which deflects toward the axis those in the $(F = 1, m_F = 1)$ and $(F = 1, m_F = 0)$ states. The beam is

The authors are with Harvard University, Cambridge, Massachusetts.

Work supported by the Office of Naval Research and the National Science Foundation.

Received July 1961.

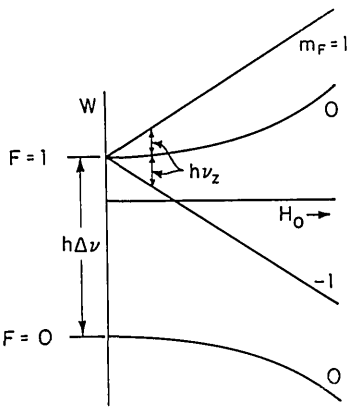

Fig. 1. Energy levels of hydrogen in its ground state.

focused on the aperture of the storage bulb, which is located in a microwave cavity tuned to the transition frequency. If the density and lifetime of atoms in the ($F = 1$, $m_F = 0$) state are great enough, oscillation due to stimulated emission occurs and a signal at the resonance frequency is detected by means of a small coupling loop in the cavity.

A. Threshold for Oscillation

The minimum flux necessary for oscillation, I_{th}, is given[4] by

$$I_{th} = \frac{h}{8\pi^2}\frac{V}{Q}\frac{1}{\eta(\mu_0 T_r)^2} \tag{1}$$

where Q is the loaded quality factor of the cavity, T_r is the mean radiation lifetime of an atom in the cavity, V is the volume of the cavity, μ_0 is the Bohr magneton, and η is a geometrical factor depending on the distribution of rf magnetic field in the cavity. Typically, $\eta \approx 3$.

For an order of magnitude estimate of the threshold flux we may take the following values: $V = 10^4$ cm^3, $Q = 3 \times 10^4$, $T = 10^{-1}$ sec. The result is $I_{th} \approx 10^{12}$ pps.

B. Noise in the Maser

The ultimate stability of the maser oscillator is governed by the influence of thermodynamic noise on the oscillation. If the maser is observed for a time t, the relative fluctuation in frequency which is observed due to noise is given[4] by

$$\frac{\langle \delta\nu^2\rangle^{1/2}}{\nu} = \frac{1}{2\pi\sqrt{2}}\frac{1}{Q_l}\sqrt{\frac{kT}{Pt}}. \tag{2}$$

Here P is the power delivered to the cavity by the beam ($P = {}^1/_2 h\nu$ approximately in a typical case). The quality factor for the resonance is defined by $Q_l = \nu/\delta\nu_r$, where $\delta\nu_r$, the resonance width of the transition, is given by $\delta\nu_r = (\pi T_r)^{-1}$. This expression takes into account the effect of atomic amplification of the noise. With $T_r = 0.3$ sec, $I = 3 \times 10^{12}$ pps, and an observation time of 1 sec, we have $\langle \delta\nu^2\rangle^{1/2}/\nu = 4 \times 10^{-15}$. The spectral purity of the maser is consequently very high. However, its long term stability in practice depends upon how well external influences can be controlled, some of which will be discussed below.

C. Limitations to Radiative Lifetime

The fundamental limit to the radiative lifetime of atoms in a given bulb is due to the rate at which atoms escape from the bulb. The characteristic time for this, T_b. is the mean time an atom spends in the storage bulb before effusing from the bulb aperture. This time may readily be determined from the geometry of the bulb. If the total volume of the bulb is V_b, and if the total escape area is A_e, then it can be shown that $T_b = 4V/(A_e\bar{v})$, where $\bar{v}$ is the mean velocity. As an example, a bulb 15 cm diam with a 2 mm diam aperture yields a lifetime $T_b = 0.8$ sec. There are, however, a variety of other effects which may relax the radiative state of an atom before it escapes from the bulb. Among the more important of the processes are:

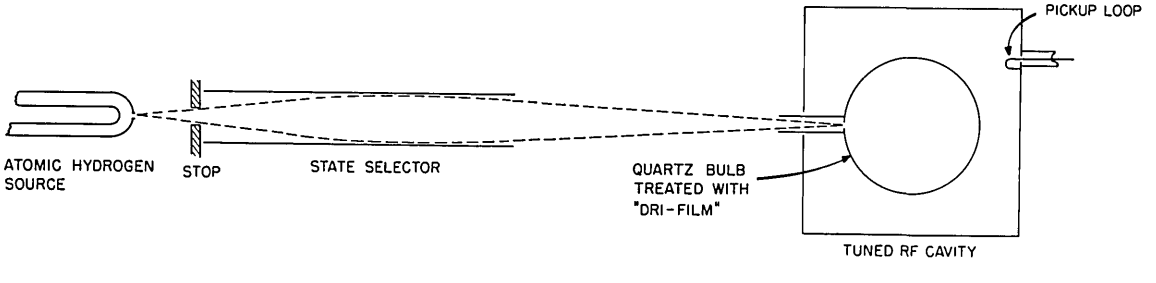

Fig. 2. Schematic diagram of hydrogen maser.

Collisions with the Walls

So far the effect of surface collisions has been neglected. The collisions can both cause a shift in the resonance frequency and broaden the resonance line. The two effects are related and can be described in terms of the parameter φ, the phase shift introduced into the wave function by a single wall collision.[3] It is shown in ref. 3 that the atom will lose its coherence with the radiation field after a number of collisions $n \approx 2/\varphi^2$. If the mean time between collisions is t_0, then the time for relaxation due to wall collisions is $T_w = 2t_0/\varphi^2$. As will be described below, φ may be determined from measurements of frequency shifts in the maser. This has so far only yielded an upper limit to φ given by $\varphi < 8 \times 10^{-4}$ rad/collision, corresponding to $T_b = 100$ sec. This is extremely large, and due to the reciprocal fashion in which relaxation times add up, may be neglected.

Spin Exchange Collisions

If all the hydrogen atoms in the cavity were in a pure state their radiation properties would be unaffected by collisions with each other. However, because of the presence of atoms in the $(F = 1, m_F = 1)$ state, and because atoms in the radiation state have different life histories in the cavity, a collision between two hydrogen atoms will in general cause a change in the wave function of each. The most important collision process is that of spin exchange. This process has been considered in some detail by Wittke and Dicke.[7] Wittke and Dicke estimate that the relaxation time for spin exchange is related to the density of hydrogen atoms, N, by

$$T_{se} = 10^9/N \text{ sec.}$$

For a flux of 10^{12} pps, and a bulb volume of 10^3 cm^3, this corresponds to a lifetime of 1 sec. Spin exchange does not place a fundamental limit on the stability of the maser, however, since if oscillation is once achieved and if no other relaxation mechanisms are important, decreasing the flux increases the radiative lifetime in inverse proportion. There is a proportional decrease in power but, as indicated in Eq. (2), $\langle \delta\nu^2 \rangle^{1/2}/\nu$ depends linearly on the lifetime but only on the square root of the emitted power, so the net effect of decreasing the flux is to increase the stability.

Motion through Inhomogeneous Magnetic Fields

If there is an inhomogeneous magnetic field in the region of the bulb, an atom, by virtue of its motion, experiences a field which varies randomly in time. Components of the varying field occurring at $\Delta\nu$ are negligible. However, there may be appreciable intensity at the Zeeman frequency, indicated by ν_z in Fig. 1. Decay of the $(F = 1, m_F = 0)$ state by either of the Zeeman transitions limits the radiation lifetime of the atom. In the absence of all other relaxation processes this mechanism can be characterized by the relaxation time T_H. There are two extreme conditions, corresponding to the Zeeman frequency being either large or small compared to the collision frequency of the atom with the walls. It is usually desirable to run the maser in a low magnetic field in which case the latter condition holds. The relaxation time is then related to the rms field inhomogeneity, $\langle H_i^2 \rangle$, by[4]

$$T_H = 3/(4\gamma^2 \langle H_i^2 \rangle \tau).$$

Here γ is the gyromagnetic ratio for the $(F = 1)$ state, 1.4 Mc/s per gauss, and τ is a correlation time which may be taken as t_0, approximately 3×10^{-4} sec. If $\langle H_i^2 \rangle^{1/2}$ is 0.1 milligauss the radiation lifetime is 3 sec. Although lifetimes this long have not been observed so far, it appears possible to achieve fields of this homogeneity in the laboratory by the use of suitable shields. It is evident, nevertheless, that the requirements for uniformity of the magnetic field are severe.

III. Frequency Perturbations

A. Wall Collisions

During the time of collision with the wall the energy levels of the hydrogen atom may be seriously perturbed and this will be reflected in a shift in the resonance frequency of the transition, since the resonance frequency depends on the average energy separation. The frequency shift, $\delta\nu$, is related to the phase shift per collision, φ, by[3]

$$\delta\nu = \varphi/(2\pi t_0).$$

It is apparent that the shift depends on the collision rate within the bulb and will reveal itself as a systematic shift in frequency with size of the storage bulb. A search with bulbs treated with dimethyldichlorosilane, the wall coating used in the experimental work described here, has failed to reveal any such shift, which implies an upper limit for the shift in a 16.5 cm diam bulb, $|\delta\nu| < 4$ cps $\approx 3 \times 10^{-9}\Delta\nu$. A lower limit may be obtained from the measured frequency shift of the hyperfine resonance in hydrogen caused by collisions of hydrogen atoms with molecular hydrogen in a buffer gas.[8] If the pressure shift in ref. 8 is extrapolated to the collision rate characteristic of the maser the frequency shift is found to be $\delta\nu > 2 \times 10^{-14}\Delta\nu$ and this may be taken as a lower limit. The reproducibility and stability of a given surface are of fundamental importance to the operation of a maser, but as yet there is no information on these points.

B. Doppler Shift

The effects of the first order Doppler shift are suppressed in the hydrogen maser because the effective average velocity of the hydrogen is very low due to its confinement in the storage bulb. The second order

Doppler shift is unavoidable, and because of the small mass of the hydrogen atom, it is fairly sizable. The frequency shift is given by

$$\delta\nu_D = + \frac{1}{2}\frac{V^2}{c^2}\Delta\nu$$

$$= + \frac{3kT}{2mc^2}\Delta\nu$$

$$= (1.4 \times 10^{-13}T)\Delta\nu$$

where T is the temperature in °C. If the second order Doppler shift is the limiting perturbation it may be reduced by a factor of 3 by using tritium instead of hydrogen.

C. Cavity Pulling

If the cavity is mistuned from the resonance by an amount $\delta\nu_c$ then the oscillator frequency will be displaced by an amount $\delta\nu = \delta\nu_c(Q_c/Q_c)$. Since the shift decreases with increasing Q_l it does not become relatively any more significant as the lifetime increases. This expression is not exact for the case of a conventional beam maser, but must be multiplied by a slowly varying function of the power level. In the case of an exponential distribution for radiation lifetime, however, the pulling effect is independent of the power level.

D. Other Effects

Because of the lack of structure of the resonance line and the symmetry of the ($F = 1$, $m_F = 1$) and ($F = 1$, $m_F = -1$) states, a number of perturbations which affect a molecular maser are either absent or very small in the case of the hydrogen maser.

IV. Apparatus

A. Wall Coating

The storage bulbs which were used in the present experiments were treated with dimethyldichlorosilane (General Electric "Dri-Film," SC-02). This compound was used by Wittke and Dicke[7] to prevent surface recombination of atomic hydrogen, and has been used to prevent hyperfine relaxation of alkalis in optical pumping experiments.[8] The surface is treated either by placing a few drops of the liquid in a carefully cleaned bulb and letting the vapor interact with the walls, or by flowing the vapor through the bulb in an inert carrier gas.

B. Source

The machine used for the present experiments has a Wood's discharge source. However, an rf discharge appears to work equally satisfactorily, and has the advantages of smaller volume and much less power dissipation. Some of the relevant characteristics of rf source are: hydrogen prsesure, 0.3 mm Hg; discharge tube size, 9 mm i.d. and 11 mm o.d.; exit orifice, 0.8 mm diam; frequency, 14 Mc/s; coupling, electric; electrodes, external, 8 cm separation; power, about 30 watts.

C. State Selector

State selection is accomplished by means of a hexapolar magnetic field. Both electromagnets and permanent magnets have been used. The latter seems more satisfactory and is much simpler in construction. A magnet similar to the design of Christenson and Hamilton,[10] but with slightly larger poles, yields a maximum field at the pole tips of 9900 gauss. The gap is 3 mm diam and 7.5 cm long. The calculated solid angle for the desired state, assuming a 2 mm image diameter, is 3×10^{-5} ster.

D. Electronics

The present measurements were made completely in terms of a National Company Atomichron which is located in the laboratory of J. A. Pierce. The frequency measuring system is indicated in Fig. 3. Auxiliary apparatus for tuning the cavity, and for measuring the radiation lifetime by a transient technique, are not shown.

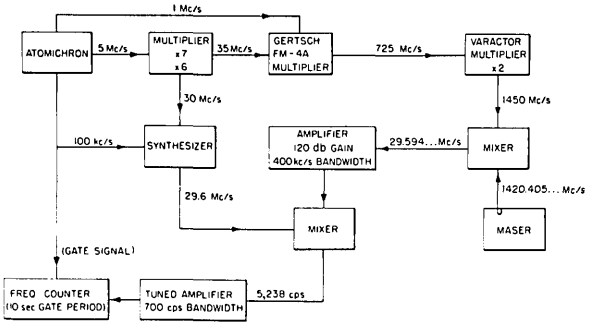

Fig. 3. Frequency measuring system.

E. Magnetic Field

The measurements described here were all carried out with the maser cavity surrounded by three orthogonal pairs of correction coils which served to cancel most of the earth's magnetic field. However, the combination of inhomogeneities imposed by the coils and residual inhomogeneities in the laboratory prevented oscillation at fields below 60 mgauss, and most measurements were taken at 80 mgauss. Fluctuations in the magnetic field due to activity of a nearby subway during the day were large enough to require measurements to be taken late at night.

The magnetic field was measured by a double resonance technique in which a signal at the Zeeman frequency is applied to the maser by means of an external coil while the oscillation level is measured. The effect of the low frequency signal is to mix the Zeeman states, thereby reducing the radiative lifetime of atoms in the ($F = 1$, $m_f = 0$) state, with a consequent diminution in radiated power. The line width of the Zeeman resonance was typically 2 kc/s.

F. Cavity

A cylindrical cavity operating in the TE_{011} mode was used. The unloaded Q of the cavity is approximately 53,000. Tuning is accomplished by a sliding end plate, and a fine tuning plunger. The resonant frequency could be measured to 250 cps by monitoring the absorption curve, using a phase-locked oscillator as the signal source.

V. Experimental Procedure

The routine followed in measurements of the hyperfine frequency of hydrogen was as follows: the cavity resonance frequency and loaded Q were first measured. This was followed by a Zeeman frequency measurement as described in Section IV, E. A series of ten measurements of the 5.7 kc/s beat frequency between the maser and the Atomichron were then made with a frequency counter, each measurement taking 10 sec and having an uncertainty due to the counter of ±0.1 cps. Zeeman measurements alternated with hyperfine measurements for six such sequences. The run was completed by remeasurement of the cavity resonance frequency.

The large number of measurements was necessary in order to average out a short term fluctuation in the Atomichron which was adjusted for long term stability and consequently has an underdamped servo loop. Measurements taken with a second Atomichron which was properly adjusted for short term stability consistently gave readings differing only by the inherent counting error. However, since the first Atomichron was monitored against other standards all the data were taken with it as the standard.

The radiative lifetime in all the measurements was approximately 0.3 sec.

VI. Results

A total of 25 runs was made with two bulbs, one 16.5 cm diam and the other 11.4 cm diam. The results are shown in Fig. 4. The following corrections were made to the data:

Magnetic field. The field present, about 80 mgauss, shifts the resonance frequency about 18 cps. The Zeeman frequency could be determined to an accuracy of 100 cps, and the consequent uncertainty introduced by this into the final result is 0.03 cps.

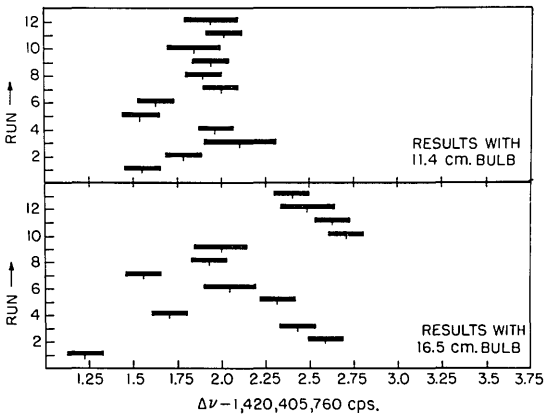

Fig. 4. Experimental results with two different size bulbs (uncorrected for second order Doppler shift).

Cavity pulling. The cavity was always tuned to within 1.5 kc/s of the hyperfine frequency, and the correction for pulling was at most 0.05 cps. The uncertainty of 300 cps in the cavity resonance frequency corresponds to an uncertainty of 0.01 cps in the cavity pulling correction.

In addition, a correction of −0.06 cps, due to the second order Doppler shift, was made to the final results (not indicated in Fig. 4). With this correction, the results for the two bulbs are:

16.5 cm diam bulb $\Delta\nu = 1{,}420{,}405{,}761.78 \pm 0.7$ cps.

11.4 cm diam bulb $\Delta\nu = 1{,}420{,}405{,}762.11 \pm 0.4$ cps.

These frequencies are in terms of the cesium hyperfine frequency which is taken as $\Delta\nu_{Cs} = 9{,}192{,}631{,}840$ cps. There is no apparent systematic shift in frequency between the bulbs. However, the deviations among individual runs are much larger than can be explained by the uncertainties mentioned above. The cause for these deviations was not determined. However, if the difference between the two results is assumed to be due entirely to a wall shift then the phase shift per collision is found to be $\varphi = +1.9 \times 10^{-4}$ rad/collision. This corresponds to a frequency shift $\delta\nu = 0.8$ cps for the larger bulb. If the extreme frequency difference between the two bulbs is used to determine maximum possible limits to the wall shift the result is $|\varphi| < 8 \times 10^{-4}$ rad/collision, or $|\delta\nu| < 4$ cps. This latter figure will be taken to be the maximum uncertainty in the final result, which is

$$\Delta\nu = 1{,}420{,}405{,}762 \pm 4 \text{ cps.}$$

Although this is slightly higher than the published value of Anderson and Pipkin,[9]

$$\Delta\nu = 1{,}420{,}405{,}726 \pm 30 \text{ cps},$$

it is in close agreement with the more recent unpublished value of Lambert and Pipkin[11]

$$\Delta\nu = 1{,}420{,}405{,}748 \pm 7 \text{ cps},$$

and falls within the limits of measurements by Wittke and Dicke[7] and Kusch.[12]

The authors wish to acknowledge the help of Norval Fortson and Howard Berg in the construction of some of the apparatus and for assistance in taking data, and wish to thank J. A. Pierce for providing the frequency standard. One of the authors (H. M. G.) wishes to acknowledge the support of an NSF Predoctoral Fellowship during the period of this work.

References

1. J. B. Gordon, H. J. Zeiger, and C. H. Townes, Phys. Rev. **95**, 2821 (1954).
2. H. M. Goldenberg, D. Kleppner, and N. F. Ramsey, Phys. Rev. Letters **5**, 361 (1960).
3. H. M. Goldenberg, D. Kleppner, and N. F. Ramsey, Phys. Rev. **123**, 530 (1961).
4. D. Kleppner, H. M. Goldenberg, and N. F. Ramsey (to be published).
5. H. M. Goldenberg, Ph.D. Thesis, Harvard, 1960 (unpublished).
6. K. Shimoda, T. C. Wang, and C. H. Townes, Phys. Rev. **102**, 1308 (1956).
7. J. P. Wittke and R. H. Dicke, Phys. Rev. **103**, 620 (1956).
8. Carroll Alley (to be published).
9. L. W. Anderson and F. M. Pipkin, Phys. Rev. Letters **4**, 69 (1960).
10. R. L. Christenson and D. R. Hamilton, Rev. Sci. Instr. **30**, 356 (1959).
11. R. H. Lambert and F. M. Pipkin, private communication.
12. P. Kusch, Phys. Rev. **100**, 1188 (1955).

Contents of Optics and Spectroscopy

VOL. XI OCTOBER 1961 *This issue will be published in complete translation by the Optical Society of America in January 1962*

Single copies of this issue $3.00 each

Yearly subscription (monthly) $15.00 ($7.50 to OSA members)